ADVANCES IN FOOD RESEARCH

SUPPLEMENT 4

Spices and Condiments:

Chemistry, Microbiology, Technology

J. S. PRUTHI

Spices and Condiments:
Chemistry, Microbiology, Technology

ADVANCES IN FOOD RESEARCH

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Spices and Condiments: Chemistry, Microbiology, Technology. 1980
J. S. PRUTHI

Spices and Condiments:
Chemistry, Microbiology, Technology

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Ludhiana
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ACADEMIC PRESS
A Subsidiary of Harcourt Brace Jovanovich, Publishers
New York London Toronto Sydney San Francisco 1980

ACADEMIC PRESS, INC.
111 Fifth Avenue, New York, New York 10003

United Kingdom Edition published by
ACADEMIC PRESS, INC. (LONDON) LTD.
24/28 Oval Road, London NW1 7DX

Library of Congress Catalog Card Number: 75–84257

ISBN 0–12–016464–7

PRINTED IN THE UNITED STATES OF AMERICA

80 81 82 83 9 8 7 6 5 4 3 2 1

DEDICATION

The author sincerely and wholeheartedly dedicates this work to his
ALMA MATER

THE CENTRAL FOOD TECHNOLOGICAL RESEARCH INSTITUTE
Mysore, India
(Council of Scientific and Industrial Research, New Delhi, India).

The author has had the privilege of serving as one of its founding staff members since March 1, 1950.

CONTENTS

ABOUT THE AUTHOR

Dr. J. S. Pruthi has received considerable worldwide recognition for his contributions to food science and technology during the last 35 years of his career. His work on spices has been especially noteworthy. His professional and academic accomplishments have been varied and impressive. He was the first Scientist-In-Charge of the Spice Technology Centre of the Central Food Technological Research Institute at the Council of Scientific and Industrial Research Complex, Trivandrum, Kerala, India.

Dr. Pruthi joined the Ministry of Food and Agriculture, Government of India, in 1964, and served as the first Director of Agmark Laboratories for a number of years. He established the Central Agmark Laboratory in Nagpur, and under his guidance as its Founder and Director, brought national and international recognition to the work being done at the facility. He also helped to establish a network of twelve regional Agmark laboratories throughout India for research on food and spice chemistry, microbiology, and technology.

Dr. Pruthi's experience in the planning, implementation, and coordination of research programs in food science and spice technology has contributed to the excellent training of student scientists and professionals in the areas of food science and technology for the food industries, quality control, and standardization of 95 agricultural and food commodities.

As Honorary Executive Secretary of the Association of Food Scientists and Technologists of India, and Convener of the Editorial Board, he has contributed and edited the annual publications of *Reviews in Food Science and Technology*. He has authored or co-authored 4 patents and over 250 scientific publications on the chemistry, microbiology, and technology of spices, as well as other topics of interest in the food science and technology field. He has led Indian delegations overseas to the International Organization for Standardization meetings for the formulation of ISO Standards for Spices, in addition to his heading several national committees for the formulation of national standards for spices and other foods. He has also received several national and international awards for patents, outstanding publications, and other literary achievements.

Dr. Pruthi now serves as Head of the Central Food Technological Research Institute Experiment Station at Ludhiana, Punjab, India.

C. O. CHICHESTER

PREFACE

Since antiquity, spices and condiments have been considered virtually indispensable in the culinary arts, as they are used to flavor foods and beverages the world over. They add savor to insipid dishes, tang to beverages, and zest to appetizers. Some are also used in perfumery and cosmetics, and others soothe and heal through medicine. The preservative and antioxygenic faculties of spices are also esteemed throughout the world. It is no wonder, therefore, that by virtue of these very qualities, spices were valued like gold in ancient times and ranked with precious stones in the inventory of royal possessions. They determined the wealth and policies of nations, played an important role in ancient medicine, and served as an incentive for the discovery of new waterways and continents.

Even now, spices travel far and wide. There is a considerable volume of international trade, amounting to about 365 million dollars annually, and hence spices play quite an important role in the national economy of several producing, exporting, and importing countries. Spices are grown in most of the tropical countries, notably in South and Southeast Asia. India is one of the major spice-producing and spice-exporting countries of the world, and, on that basis, it is rightly called the Home of Spices.

According to the International Organization for Standardization (ISO), there are as many as seventy spices and condiments grown in different parts of the world. During the past sixty years, a vast amount of interesting and valuable information has been published in thousands of journals, bulletins, and reports. The tremendous quantity of information available justifies the publication of a comprehensive technical compendium, as previous publications, although useful, are either of a popular nature or deal with historical, morphological, agricultural, and botanical aspects. To my knowledge, there is as yet no single book dealing with the chemistry, microbiology, and technology of at least the most important spices collectively. In this supplement to *Advances in Food Research,* an attempt has been made only to highlight some of the major findings relating to some important spices and to point out avenues for further research. Thus, this publication is by no means exhaustive, and I am fully aware of its limitations. There are over 4000 literature references available, but

this publication covers only about 1700. It was a rather difficult job to make this selection. However, in no way are aspersions cast on other valuable works not cited here owing to lack of space.

To gather, process, and compress the available published information into this supplement to *Advances in Food Research* has been no easy task, but the assignment has been rewarding in the valuable experience gained and in the feeling of having been, in a limited way, of some service to the spice industry and to spice scientists in projecting the importance of spices to man, recent developments in the field, and future research needs.

This supplement covers, in brief, the nomenclature, properties (flavoring, antioxygenic, preservative, antimicrobial or antibiotic, physiological, and medicinal),use in perfumery and cosmetics, chemistry and chemical constituents, general and specialized techniques of analysis, adulterant detection, marketing, quality control and standardization on national and international levels, postharvest technology, processing technology, utilization of wastes, packaging, storage, transportation, and sanitation (microbial and insect infestation and their control) of spices and condiments. The final chapter, which discusses future research needs, highlights several important gaps and bottlenecks in spice trade, research, and quality control enforcement so vital for the development of the spice industry on an international level.

In the preparation of this publication, I have relied heavily on earlier published works in thousands of journals, bulletins, and so on. I wish to express my warm appreciation for the excellent cooperation extended by various spice scientists, institutions, universities, and governmental departments in freely supplying information, reprints, and publications. The task of publishing this first technical volume of its kind, on important spices collectively, was made easier by the fine cooperation of the Editors and Academic Press. I also warmly appreciate the excellent cooperation and help given by my wife, Ujjal K. Pruthi, and my sons, Premjit, Gurjeet, and Harjeet during the course of the preparation of this manuscript.

I should also like to express my gratitude to the Director, Central Food Technological Research Institute, Mysore, and the Director General, Scientific and Industrial Research, Council of Scientific and Industrial Research, New Delhi, for their kind encouragement.

J. S. PRUTHI

Spices and Condiments:
Chemistry, Microbiology, Technology

1

Introduction

From the hieroglyphics on the walls of the pyramids to the Scriptures of the Bible, we find constant mention of the important part spices have played in the life of ancient peoples. An outline of the past history of the spice industry has been aptly described by Parry (1953), who gives an interesting account of how spices ranked with precious stones in the inventory of royal possessions. Spices were monopolized by the few; they determined the wealth and policies of nations, played an important role in ancient medicine, and served as an incentive for the discovery of new waterways and new continents! Thus, the fascinating history of spices is a story of adventure, exploration, conquest, and wars at sea.

Even now, spices and condiments travel far and wide (1) to flavor numerous foods and beverages, (2) to soothe and heal through medicine, and (3) to be used in cosmetics and perfumery. They have become particularly indispensable in the culinary arts, adding savor to insipid dishes, tang to beverages, and zest to appetizers. The preservative and antioxygenic faculty of spices was esteemed on a par with their flavoring properties by the people of ancient times. These qualities of spices constituted the main reason why they were valued like gold in those days and why men from distant lands sought them so eagerly.

Even in present times there is a considerable volume of international trade, amounting to over $165 million or an equivalent £63 million or 1134 million rupees (Commonwealth Secretariat, 1969). About 340 million rupees worth of spices were exported from India alone during 1969 (Directorate General of Commercial Intelligence and Statistics, 1970), and 1000 million rupees worth in 1977–1978, and 1610 million rupees worth in 1978–1979 (Spices Export Promotion Council, Cochin, India, 1975, 1979).

According to recent statistics, the United States is the largest importing country

in the world, taking about 40% of the total international trade. During 1968–1969, the United States imported about 250 million pounds of spices valued at $66 million (U.S. Department of Agriculture, 1970). Evidently, spices play an important role in international trade and hence affect the national economy of several producing, exporting, and importing countries.

According to the International Organization for Standardization (ISO) (1968, 1972), there are as many as seventy spices and condiments grown in different parts of the world. A systematic survey of the available literature for the past sixty years revealed that the valuable and interesting published information was quite voluminous (running into a few thousand references), enough to justify a comprehensive technical compendium or a monograph in two or three volumes.

In the present review, an effort has been made to cover only the important findings relating to a few important spices. A separate, detailed monograph is being prepared by the author.

The earlier brief reviews relate mostly to individual spices such as asafoetida (Subrahmanyan *et al.*, 1954), black pepper (Pruthi, 1968a,b; Govindarajan, 1977), caraway (Sandermann, 1942; Pomini, 1963), cardamom (Kulkarni and Pruthi, 1967; Shankaracharya and Natarajan, 1971d), coriander (Guenther, 1943; Shankaracharya and Natarajan, 1971c), chilies (Natarajan *et al.*, 1968b), cinnamon (Lawrence, 1967), cloves (Molnar, 1942), curry powder (Pruthi, 1964, 1966), cumin (Shankaracharya and Natarajan, 1971a), fenugreek (Shankaracharya and Natarajan, 1972), ginger (Srinivasan *et al.*, 1959; Pruthi *et al.*, 1960a; Srivas *et al.*, 1963b), garlic (Krishnamurthy and Sreenivasamurthy, 1956; Pruthi *et al.*, 1959a; Shivrina, 1962; Petkov, 1966), horseradish (Babichev and Lukovnikova, 1961), juniper (Pomini, 1962), leafy spices (Shankaracharya and Natarajan, 1971b), mace and nutmeg (Weil, 1965), onion (Shifrina, 1961), poppyseed (Pfiefer, 1962), saffron (Zofia, 1948; Fernandez Pizzaro and Samper, 1953; Sastry *et al.*, 1955a), turmeric (Shankaracharya and Natarajan, 1973a), vanilla and vanillin (Hora, 1961; Horst and McGlumphy, 1962; Guenther *et al.*, 1959, 1961, 1963, 1965, 1966, 1967, 1969, 1971, 1973, 1975, 1977), piperine (Pruthi, 1969b, c,d, 1970a,b), capsaicin (Pruthi, 1969a, 1970c), paprika, red pepper, and chilies (Pruthi, 1969a, 1970c), and other spices (Boyle, 1955; Natarajan, 1967; Sreenivasamurthy and Krishnamurthy, 1959; Srinivasan *et al.*, 1959; Srivas *et al.*, 1963a,b; Abraham *et al.*, 1976; Pruthi, 1976, 1980a,b).

The general methods of analysis and the approximate composition of some spices are found scattered in books on food analysis (Leach and Winton, 1941; Woodman, 1941; Winton and Winton, 1945; Nichols, 1952; Jacobs, 1958; Cox and Pearson, 1962; Association of Official Analytical Chemists, 1975; Pruthi, 1976; American Spice Trade Association, 1968; Indian Standards Institute, 1973–1976). Some aspects of the detection of adulteration and the microscopy of spices and vegetable foods are partially covered in earlier books (Winton and Winton, 1916, 1939; Schneider, 1921; Blyth *et al.*, 1927; Liverseage, 1932).

Guenther (1948–1952), Guenther *et al.* (1959, 1961, 1963, 1965, 1967, 1969, 1971, 1973, 1975, 1977), and Kulka (1962) have published interesting books and reviews on essential oils and related products, including some spice essential oils. The *Handbook of Spices* by Parry (1945) covers morphological aspects with photomicrographic illustrations, but it covers very little of the chemical aspects, etc. The Canadian and American standards for important spices are included briefly; these have since been amended from time to time during the past thirty-two years. The *Story of Spices* by Parry (1953) covers only the history of spices. *Spices—Their Morphology, Histology and Chemistry* by Parry (1962) deals mostly with botanical aspects, with only one small chapter on proximate composition, which is by no means comprehensive. The *Handbook of Spices* by Rosengarton (1969) is also of a popular or semitechnical nature; it is well illustrated but covers only general information on about 40 spices. Similarly, a well-illustrated handbook, *Spices and Condiments,* by Pruthi (1976), that describes as many as 70 spices is also of a semitechnical nature.

Thus, none of these books or publications review the latest scientific literature covering the chemical, physiological, medicinal, and microbiological aspects of spices or discuss the application of the latest analytical techniques, such as chromatography, thin-layer chromatography and mass spectroscopy, packaging, storage, insect and microbial infestation and their control, marketing, quality control, and detection of adulteration and standardization of spices. In the present work, an attempt has been made to cover all these aspects of some of the important spices of international interest, but lack of space does not permit us to consider in detail all the seventy spices and condiments.

The present publication, in brief, covers nomenclature, properties and uses of spices (including flavoring, antioxygenic, antimicrobial, and physiological and medicinal properties), and chemical aspects, including analytical techniques for major constituents, chemical composition, and detection of adulteration. It also covers several technological aspects, such as postharvest technology, processing technology, packaging, storage and transportation, sanitation, including microbial contamination and control, and insect infestation and control, marketing, international standards, and other areas of interest. Processing technology includes sun drying and dehydration, freeze grinding, freeze drying, processing of spice essential oils and oleoresins, and manufacture of new spice products such as liquid spices, instant spices, sterilized spices, imitation spices, encapsulated spices, seasonings, spice blends, and mixtures, spice essences, emulsions, salts, pastes, concentrates, and decoctions, dispersed spices or dry soluble spices, and other new spice products, their quality and storage stability, etc. Utilization of spice waste forms the subject matter of a separate chapter. At the end, additional research needs are listed, and selected references (about 1700) are presented.

The correct nomenclature of spices is of paramount importance and is therefore discussed first.

I. NOMENCLATURE

In international trade questions and disagreements sometimes arise relating to (1) nomenclature of spices and condiments, (2) methods of sampling, (3) methods of analysis, and (4) limits of quality specifications. There are also frequent controversies over the definitions of the terms spices and condiments. In a review such as this, it is desirable first to define such terms clearly and then to identify the correct botanical origin of the variety or type of spices of the particular aromatic plants. There are over seventy spices now officially recognized by the International Organization for Standardization (ISO). This organization represents the various spice-producing, spice-exporting, and spice-importing countries. The ISO has arrived at a commonly accepted definition of spices for international trade, after a prolonged and careful consideration during the thirteen years of its existence. The ISO found it difficult to differentiate between spices and condiments, and therefore decided to keep both terms.

According to the ISO (1968, 1972), the term "spices and condiments" applies to natural vegetable products or mixtures thereof, without any extraneous matter, that are used for flavoring, seasoning, and imparting aroma to foods; the term applies to the product either in the whole form or in the ground form.

The nomenclature of spices and condiments, as now accepted by all the member countries of the ISO at a meeting held in Paris in May–June, 1970, is given in Table I. In addition to the common English name, the table gives the botanical name, the name of the family to which it belongs, the nature of the plant—whether it is an annual, perennial, or evergreen, and whether it is a tree, creeper, or bush—and the part of the plant used as a spice. This information is thus available for ready reference by both the spice-exporting and spice-importing countries, and by the quality standards enforcement authorities on national and international levels.

A review of Table I reveals that spices and condiments come from the following parts of aromatic plants:

1. Fruits (capsicum, black pepper, cardamom, etc.).
2. Seeds (aniseed, caraway, celery, coriander, cumin, fennel, fenugreek, mustard, etc.).
3. Rhizomes or roots (ginger, turmeric, etc.).
4. Leaves (bay leaves, marjoram, parsley, sage, thyme, etc.).
5. Barks (cinnamon and cassia).
6. Floral parts (saffron, cloves, caper, etc.).
7. Bulbs (onion, garlic, shallot, etc.).

TABLE I NOMENCLATURE OF SPICES AND CONDIMENTS[a]

English name	Botanical name	Family[b–d]	Nature of plant[b–d]	Part used as spice
Ajowan	*Trachyspermum ammi* (Linn.) Sprague Syn. *Carum copticum* Hiern.	Umbelliferae	Perennial plant	Fruit (dried)
Allspice or pimento	*Pimenta dioica* (Linn.) Merrill Syn. *Pimenta officinalis* Lindley	Myrtaceae	Evergreen tree	Berry (dried) and leaf
Amchoor	*Mangifera indica* Linn.	Anacardiacea	Evergreen tree	Unripe fruit (dried slices)
Anardana	*Punica granatum* Linn.	Punicaceae	Shrub or small tree	Seed (dried with flesh)
Angelica	*Angelica archangelica* Linn.	Umbelliferae	Perennial plant	Fruit, young stem and root
Anise or aniseed	*Pimpinella anisum* Linn. Syn. *Anisum vulgare* Gaertner *Anisum officinarum* Moench	Umbelliferae	Annual herbaceous plant	Fruit
Asafetida	*Ferula asafoetida* Linn.	Umbelliferae	Perennial herb	Rhizome
Balm or lemon balm	*Melissa officinalis* Linn.	Labiatae	Leafy perennial	Leaf
Basil or sweet basil	*Ocimum basilicum* Linn.	Labiatae	Annual plant	Leaf
Bay or laurel leaf	*Laurus nobilis* Linn.	Lauraceae	Evergreen tree	Leaf
Bay, West Indian	*Pimenta racemosa* (P. Miller) J. W. Moore	Myrtaceae	Small tree	Leaf, fruit
Caper	*Capparis spinosa* Linn.	Capparidacae	Trailing shrub	Unopened flower bud
Capsicum or chili	*Capsicum annum* Linn.[e]	Solanaceae	Shrub	Fruit
	Capsicum frutescens Linn.[e]	Solanaceae	Perennial plant	Fruit
Caraway	*Carum carvi* Linn. Syn. *Apium carvi* (Linn.) Crantz *Seseli carvi* (Linn.) Lamarck et De Candolle	Umbelliferae	Biennial plant	Fruit
Cardamom				
Madgascar cardamom	*Aframomum augustifolium* (Sonnerat) K. Schumann	Zingiberaceae	Herb	Fruit and seed
Cameroon cardamom	*Aframomum hanburyi* K. Schumann	Zingiberaceae	Herb	Fruit and seed
Korarima cardamom	*Aframomum korarima* (Pereira) Engler	Zingiberaceae	Herb	Fruit and seed
Grains of Paradise or Guinea grains	*Aframomum melegueta* (Roscoe) K. Schumann	Zingiberaceae	Herb	Fruit and seed

(*continued*)

TABLE I (*Continued*)

English name	Botanical name	Family[b–d]	Nature of plant[b–d]	Part used as spice
Greater cardamom				
Bengal cardamom	*Amomum aromaticum* Roxburgh	Zingiberaceae	Herb	Fruit and seed
Round cardamom or Chester cardamom or Siam cardamom	*Amomum kepulaga* Sprague et Burkill Syn. *Amomum cardamomum* Roxburgh	Zingiberaceae	Herb	Fruit and seed
Cambodian cardamom	*Amomum krervanh* Pierre et Gagnepain	Zingiberaceae	Herb	Fruit and seed
Greater Indian cardamom or Nepal cardamom	*Amomum subulatum* Roxburgh	Zingiberaceae	Herb	Fruit and seed
Lesser cardamom	*Elettaria cardamomum* (Linn.) Maton (a) var. minuscula Burkill (b) var. major Thwaites	Zingiberaceae	Herb	Fruit and seed
Cassia				
Batavia cassia or Java cassia or cassia vera or Padang cinnamon	*Cinnamomum burmannii* C. G. Nees et Blume	Lauraceae	Tree	Bark
Cassia China or cassia	*Cinnamomum aromaticum* C. G. Nees	Lauraceae	Tree	Bark
Saigon cassia	*Cinnamomum loureirii* C. G. Nees	Lauraceae	Tree	Bark
Celery seed	*Apium graveolens* Linn. var. rapaceum de candalle	Umbelliferae	Biennial herb	Fruit
Chervil	*Anthriscus cerefolium* (Linn.) Hoffmann	Umbelliferae	Annual plant	Leaf
Chive or cive	*Allium schoenoprasum* Linn.	Liliaceae	Perennial	Leaf
Cinnamon	*Cinnamomum zeylanicum* Blume	Lauraceae	Evergreen tree	Bark
Clove	*Eugenia caryophyllus* (C. Sprengel) Bullock et Harrison Syn. *Syzygium aromaticum* (Linn.) Merrill et Perry	Myrtaceae	Evergreen tree	Unopened flower bud
Coriander	*Coriandrum sativum* Linn.	Umbelliferae	Annual herb	Leaf and seed
Cumin black	*Nigella sativa* Linn.	Umbelliferae	Annual plant	Fruit
Cumin seed	*Cuminum cyminum* Linn.	Umbelliferae	Annual plant	Fruit
Curry leaf	*Murraya koenigii* (Linn.) Sprengel	Rutaceae	Small tree	Leaf
Dill seed	*Anethum graveolens* Linn.	Umbelliferae	Annual plant	Fruit

Fennel seed	*Foeniculum vulgare* P. Miller	Umbelliferae	Perennial herb	Fruit and seed
Fenugreek seed	*Trigonella foenumgraecum* Linn.	Leguminosae	Annual plant	Seed
Galanga	*Alpinia galanga* (Linn.) Willdenow	Zingiberaceae	Perennial plant	Rhizome
Garlic	*Allium sativum* Linn.	Liliaceae	Perennial plant	Bulb
Ginger	*Zingiber officinale* Roscoe	Zingiberaceae	Perennial plant	Rhizome
Horseradish	*Armoracia rusticana* (Gaertner) Meyer et Scherbius	Cruciferae	Perennial plant	Root
Hyssop	*Hysopus officinalis* Linn.	Gilibert	Perennial shrub	Leaf
Juniper	*Juniperus communis* Linn.	Coniferae	Evergreen plant	Berry
Leek or stone leek or Welsh onion	*Allium fistulosum* Linn.	Liliaceae	Tall biennial (bulbous)	Leaf
Lovage	*Levisticum officinale* Koch	Ammaceae	Perennial herb	Fruit, root
Mace	*Myristica fragrans* Houttuyn	Myristicaceae	Tall evergreen tree	Aril
Marjoram	*Majorana hortensis* Moench Syn. *Origanum majorana* Linn.	Labiatae	Perennial herb	Leaf and flowering top
Mint				
Mint	*Mentha arvensis* Linn. var. piperascens	Labiatae	Perennial herb	Leaf
Peppermint	*Mentha piperita* Linn. var. piperita	Labiatae	Perennial herb	Leaf
Spearmint	*Mentha spicata* Linn. Syn. *Mentha viridis* Linn.	Labiatae	Perennial herb	Leaf
Mustard				
Brown or Indian mustard	*Brassica juncea* (Linn.) Czernjoyev	Cruciferae	Annual plant	Seed
Black mustard	*Brassica nigra* (Linn.) Koch.	Cruciferae	Annual plant	Seed
White mustard	*Sinapsis alba* Linn. Syn. *Brassica hirta* Moench	Cruciferae	Annual plant	Seed
Nutmeg	*Myristica fragrans* Houttuyn	Myristicaceae	Evergreen tree	Kernel
Onion	*Allium cepa* Linn.	Liliaceae	Biennial plant	Bulb
Origanum or oregano or Mexican oregano or wild marjoram	*Origanum vulgare* Linn.	Labiatae	Perennial plant	Leaf and flowering top
Parsley	*Petroselinum crispum* (Miller) Nymann et A. W. Hill	Umbelliferae	Biennial herb	Leaf and seed
Pepper				
Black pepper	*Piper nigrum* Linn.	Piperaceae	Perennial climber	Berry

(*continued*)

TABLE I (*Continued*)

English name	Botanical name	Family[b–d]	Nature of plant[b–d]	Part used as spice
West African or Benin pepper	*Piper guineense* Schumacher et Thonning	Piperaceae	Perennial climber	Berry
Long pepper (Pipli or pipal)	*Piper longum* Linn.	Piperaceae	Perennial climber	Fruit, leaf, seed
Poppyseed	*Papaver somniferum* Linn.	Papaveraceae	Annual plant	Seed
Rosemary	*Rosmarinus officinalis* Linn.	Labiatae	Evergreen shrub	Leaf
Saffron	*Crocus sativus* Linn.	Iridaceae	Annual plant	Stigma
Sage	*Salvia officinalis* Linn.	Labiatae	Perennial herb	Leaf
Clary or clary sage	*Salvia sclarea* Linn.	Labiatae	Biennial herb	Leaf
Savory	*Satureia hortensis* Linn.	Labiatae	Annual herb	Leaf and flower
Summer savory	*Satureia montana* Linn.	Labiatae	Annual herb	Top stem, leaves
Shallot	*Allium ascalonicum* Linn.	Liliaceae	Bulbous herb	Bulb
Star anise	*Illicium verum* Hooker	Magnoliaceae	Evergreen tree	Fruit
Sweet flag or calamus root	*Acorus calamus* Linn.	Araceae	Perennial plant	Rhizome
Tamarind	*Tamarindus indica* Linn.	Leguminasae	Evergreen tree	Fruit
Tarragon	*Artemisia dracunculus* Linn.	Compositae	Perennial herb	Leaf and flowering top
Thyme	*Thymus vulgaris* Linn.	Labiatae	Perennial plant	Leaf and flowering top
Wild thyme	*Thymus serpyllum* Linn.	Labiatae	Perennial plant	Leaf and flowering top
Turmeric	*Curcuma longa* Linn. Syn. *Curcuma domestica* Valeton	Zingiberaceae	Herbaceous plant	Rhizome
Vanilla	*Vanilla fragrans* (Salisb) Ames Syn. *Vanilla planifolia* Andrews	Orchidaceae	Climbing plant	Pod (cured fruit)
Vanilla	*Vanilla tahitensis* J. W. Moore	Orchidaceae	Climbing plant	Pod (cured fruit)
Vanillon	*Vanilla pompona* Schneider	Orchidaceae	Climbing plant	Pod (cured fruit)
Xylopia	*Xylopia aethiopica* (Dunal) A. Richard	Anonaceae		Fruit

[a] From International Organization for Standardization (1968, 1972).

[b] Willis (1951).

[c] Parry (1945).

[d] Council of Scientific and Industrial Research, Publication Directorate (1949–1976).

[e] Generally, these two names cover the capsicums of the trade—namely, paprikas of Central Europe, red pepper of Southern Europe, North Africa, and North America, and red chilies of the tropical regions of Southeast Asia and Africa.

These aromatic plants are generally grown in different parts of the world and are known by different names in different languages in different countries, as given in Table IA.

II. WORLD PRODUCTION AND CONSUMPTION

Although spices are cultivated in most of the tropical countries, their production on a commercial scale is confined to a few regions. South and Southeast Asia produces black and white pepper, cinnamon, cassia, and cardamoms. India and Pakistan produce chilies and turmeric. Tanzania (Zanzibar) and the Malagasy Republic are the principal sources of the world's supplies of cloves, and the latter country is also the major source of vanilla. Jamaica is important for ginger and pimento, and Grenada for nutmeg. Sierra Leone, Nigeria, and India export sizable quantities of ginger, and the Seychelles and Ceylon export cinnamon. Indonesia is also an important source for a number of spices, particularly black and white pepper and cassia. The International Trade Centre (1977a,b) has covered in two impressive volumes the world production, harvest calendar, important spice-producing, exporting, and importing countries, their spice consumption patterns, per capita spice consumption, and international marketing of spices.

III. INTERNATIONAL MARKETING

The value of world trade in spices during the year 1975 was around £129 million (or $290 million), of which India, the major spice-producing country, alone contributed about 25%. During 1978–1979, according to the latest statistics available (Directorate General of Commercial Intelligence and Statistics, 1979), India exported spices and condiments worth about 1610 million rupees ($200 million). These figures vary considerably from year to year, however, owing to fluctuations in the volume of trade and in the prices of different spices. Although there are many spices of commercial importance, international trade is largely concentrated in a few of them. Pepper alone may account for about 30% of the total international trade, and, together with cloves, cardamom, cinnamon, and nutmeg, it may represent about 70% of the whole spice trade. The export of nine spices—pepper, cloves, cardamom, cinnamon, nutmeg, mace, cassia, ginger, and pimento—accounts for about 90% of the total international trade. The annual trade in pepper, which is by far the most important of all the spices grown, is valued at around £21 million, followed by capsicum and chilies at £8 million, cinnamon and cassia at £6 million, cloves at £5.3 million, vanilla at £5 million, cardamom at £5 million, nutmeg and mace together at £3.5 million, and ginger

TABLE IA NAMES OF IMPORTANT SPICES AND CONDIMENTS IN DIFFERENT LANGUAGES

English name	Family	Arabic	Chinese	Dutch	French	German	Hindi (Indian)
Allspice	Umbelliferae	Bahar	—	Pimint	Piment	Piment	—
Aniseed	Umbelliferae	Yanisun	Huei-Hsiang	Anys	Anis	Anis	Valaiti-Saunf
Basil	Labiatae	Raihan	Lo-le	Basilicum	Basilic	Basilien-	Babui Tulsi
Bay leaf	Lauraceae	Ghar	Yueh-Kuei	Laurier	Laurier	Lorbeer	—
Capsicums							
Cayenne	Solanaceae	Filfil ahmar	La-Chiao	Caynene pepper	Poivre rouge	Cayenene pfeffer	Lal Mireh
Chili	Solanaceae	Filfil	Hung-fan-Chiao	Spaanse pepper	Piment enrage	Beissfee	Lal Mireh
Paprika	Solanaceae	Filfil ahmar	Hsiung ya-li-chao	Spaanse pepper	Poivre de guinee	Paprika	Simla Mirch
Caraway	Umbelliferae	Karouya	Yuan-sui	Karwij	Carvi	Kümmel	Siah Zira
Cardamom	Zingiberaceae	Hal	Poi-tau-kou	Kardemom	Cardamome	Kardamom	Chhota ilaichi
Cassia	Lauraceae	Darasini	Kuei or kwell	Kassia	Cochinchine	Zimtkassie	Jangli Dalchini
Celery	Umbelliferae	Karafs	Chin	Selderij	Celeri	Sellerie	Shalari, Ajmud
Chervil	Umbelliferae	Mardunis-Afranji	San-lo-po	Kervel	Cerfeuil	Kerbel	Baz Atrila
Chive	Liliaceae	Basal	Hsia-ye-Tsung	Biesiook	Giboulette	Schnittlauch	—
Cinnamon	Lauraceae	Qurfa	Jau-Kuei or Jow-Kevei	Kaneel	Cannella	Zimt	Darchini
Clove	Myrtaceae	Qaranful	Jing-Hsiang	Kruidnagel	Girofle	Gewürznelke	Laung
Coriander	Umbelliferae	Kuzbara	Hu-sii	Koriander	Coriandra	Koriander	Dhanya
Cumin seed	Umbelliferae	Kamnun	Ma-chin	Komyer	Cumin	Lomisch kommel	Safaid Zira
Dill	Umbelliferae	Shibith	Sheh-lo	Dille	Aneth	Dill	Sowa, Soya
Fennel	Umbelliferae	Shamar	Hiu-Hsiang	Venkel	Fenouil	Fenchel	Sounp
Fenugreek	Leguminosae	Hulba	Ku-Tou	Fenegriek	Fenugrec	Bockshornkbe	Methi, Metha

Garlic	Liliaceae	Thum	Suan	Knoflook	Ail	Knoblauch	Lasan, Lassan
Ginger	Zingiberaceae	Zanjabil	Chiang	Gember	Gingembre	Ingever	Adrak
Horseradish	Cruciferae	Fiyl Ha	Lagen	Mierikwortel	Raifort	Meerrettich	—
Mace	Myristaceae	Basbasa	Jou-Tou-Kou	Notemuskaat	Macis	Muskatnuss	Jaivetri
Marjoram	Labiatae	Marzaujush	Ma-Yueh-hanhua	Marjolein	Marjolaine	Majoran	Marwa
Mint	Labiatae	Nana	Yang-Po-Ho	Pepermunt	Menthe	Pfefferminze	Pudina
Mustard	Cruciferae	Khardal	Chiep	Mosterd	Moutarde	Senfsaat	Rai, Banarsi Rai, Safaid Rai
Nutmeg	Myristicaceae	Basbasa	Jou-Tou-Kou	Notemuskaat	Muscade	Muskatnuss	Jaiphal
Onion	Liliaceae	Basal	Yang-Tsung	Ui	Oignon	Zwiebel	Piyaz
Oregano	Labiatae	Anrar	—	Wilde marjolein	Origan	Dost	Sathra, Mirzan-Josh
Parsley	Umbelliferae	Baqdunis	Yang-Hu-Sui	Peterselie	Persil	Petersilie	Ajmood
Pepper	Piperaceae	Filfil Aswad	Hu-Chiao	Peper	Poivre	Pfeffer	Kali Mirch
Poppyseed	Papaveraceae	Khashkhash	Ying-Shu	Slaapbol	Pavot	Mohn	Kaskash
Rosemary	Labiatae	Iklil Al Jabal	Mi-Tieh-Hsiang	Rozemarijn	Romarin	Rosmarin	Rusmary
Saffron	Iridaceae	Zafaran	Tan-Hung-Hua	Saffraan	Safran	Safrin	Zaffron
Savory	Labiatae	Nadgh	Hsiang PoHo	Bonenkruid	Sarriette	Bohnenkraut	Salvia-Sefakups
Sage	Labiatae	Mariyamiy	Ching-Chieh	Salie	Sauge	Salbei	—
Tarragon	Compositae	Tarkhun	Ai-Hao	Dragon	Estragon	Estragon	—
Thyme	Labiatae	Satar	Pai-Li-Hsiang	Tijm	Thym	Thymian	Banajwain
Turmeric	Zingiberaceae	Kurkum	Yu-Chin	Geelwortel	Curcuma	Kurkuma Gelbivarzel	Haldi
Vanilla	Orchidaceae	Wanila	Hsiang-Tsao	Vanilla,	Vanille	Vanille	Vanilla

(*continued*)

TABLE IA (*Continued*)

English name	Italian	Japanese	Portugese	Russian	Spanish	Swedish
Allspice	Pepe Di Giamaic	—	Pimenta da Jamaica	Yamayski Pyereto	Pimienta gorda	Krydd peppar
Aniseed	Anice	—	Erva-Loce	Anis	Anis	Anis
Basil	Basilies	Meboki	Manjiricao	Bazilik	Albahaca	Basilkort
Bay leaf	Alloro	Gekkeijie	Laureiro	Laur	Lawiel	Lager
Capsicums						
Cayenne	Pepe Oli caiena	—	Pimentao-de-caiene	Kayenski Pyerets	Pimenton	Kajenn pepper
Chili	Peperone	Jogarashi	Pimentao picante	Stsuchkovy Pyerets	Chile	Spank peppr
Paprika	Peperane	Jogarashi	Pimentao	Stsuchkovy Pyerets	Pimenton	Spansk pepper
Caraway	Carvi	—	Aliaravia	Twin	Alearavea	Kummin
Cardamom	Cardamoma	Karudaman	—	Kardaman	Cardamomo	Kardamumma
Cassia	—	Bokie	—	—	Canela de la China	Kassia
Celery	Sedano	Serorii	Aip o	Syelderey	Apio	Selleri
Chervil	Cerfoglio	—	Carefotho	Kervel	Cerafolio	Karvel
Chive	Apollina	—	Cebolinha	Luk-Rezanyeto	Cebolleta	Grastok
Cinnamon	Lanuella	Seiron-Nikkei	—	Koritsa	Canela	Kanel
Clove	Garafano	Chaji	Cravo	Gvozolika	Clavo	Kryddnezlik
Coriander	Coriandolo	Koendoro	Coentro	Koriander	Culantro	Koriander
Cumin seed	Comino	—	Cominho	Kmin	Comino	Spiskumin
Dill	Aneto	—	Endro	Uprop	Eneldo	Dill
Fennel	Finacchio	Uikyo	Funcho	Fyenkhel	Hinojo	Fankal

Fenugreek	Fieno Greco	Koroha	Alforva	Pazhitnik	Alholva	Bockhornskover
Garlic	Aglio	Ninniku	Alho	Chesnok	Ajo	Vitlok
Ginger	Zenzero	Shoga	Gengibre	Imbir'	Jengibre	Ingefara
Horseradish	Rafano	Wasabi	Rabanopicanta	Khren	Robanopicante	Pepparot
Mace	Noce Mascota	Nikuzuku	Noz-Moscada	Oryckh Musatny	Nuez Mescada	Muskot
Marjoram	Maggiorana	Mayorana	Manjerona	Mayoran	Amaraco	Mejram
Mint	Menta	Seiyo-Hakpa	Hortela	Myata	Menta	Pepparmynta
Mustard	Senape	Shiro-Karshi	Mostarda	Gorchista	Mostaza	Senap
Nutmeg	Noce Maskata	Nikuzuku	Noz-Moscada	Oryekh Muckatny	Nuez Mascada	Muskot
Onion	Lipolla	Tamanegi	Cebola	Luk	Cebolla	Redhek
Oregano	Regamo	—	Ouregao	Dushitsa	Oregano	Vild Mejram
Parsley	Prezzemolo	Paseri	Salsa	Pyetrushka	Perijil	Persilja
Pepper	Pepe	Kosho	Pimenta	Pyerets	Pimienta	Peppar
Poppyseed	Papavero	Keshi	Dormideira	Mak	Adormidera	Vallmo
Rosemary	Ramerino	Mannenro	Alecrin	Rozmarin	Romero	Rosmarin
Saffron	Zafferano	Safuran	Scafrao	Shafran	Asafran	Saffran
Savory	Santoreggio	—	Segurelha	Chabyor	Ajedrea	Kyldel
Sage	Salvia	Sage	Salva	Shalfey	Salvia	Salvia
Tarragon	Targone	—	Estragao	Estragon	Estargon	Dragon
Thyme	Timo	Jakoso	Tomilho	Jimyan	Tomilo	Tiimjan
Turmeric	Curcuma	—	Acfrao-Da. Zholti-India	Imbir	Curcuma	Gurkmeja
Vanilla	Vaniglia	Banira	Baunilha	Vanil	Vanilla	Vanilj

and pimento at more than £2 million each (Commonwealth Secretariat, 1969).

Among the importers of spices and condiments, the United States is by far the largest, with annual spice imports worth about £21 million (Commonwealth Secretariat, 1969). During 1975, the United States imported about 63,622 tons valued at $100 million (International Trade Center, 1977a,b). Imports into the countries of the European Economic Community together account for just over £14 million. In recent years, the Soviet Union has been an important market with imports valued at around £5 million annually. Other principal markets are the United Kingdom with around £3 million and Ceylon with £5 million, mainly because of her large imports of capsicums and chilies (Commonwealth Secretariat, 1969). According to the Australian Bureau of Census and Statistics, Australia imported spices worth over $1.5 million during 1968–1969. Although Singapore features as a large importer, second only to the United States, it re-exports practically the entire amount (Commonwealth Secretariat, 1969).

India continues to occupy the privileged top position among the commonwealth countries in the world supply of spices and condiments, her total exports during 1974–1975 being of the order of about 620 million rupees (Spices Export Promotion Council, Cochin, India, 1975) and 1610 million rupees in 1978–1979.

Zanzibar, the main source of the world's supply of cloves, depends heavily on this trade, which in 1967 accounted for 80% of her total exports. Among the other producers in the Commonwealth, Grenada relies heavily on her exports of nutmegs and mace, and Malagasy Republic exports vanilla, cloves, and pepper (Commonwealth Secretariat, 1969).

IV. PER CAPITA SPICE CONSUMPTION

Although direct information on per capita spice consumption in many countries is generally not available, the trends in world consumption may be assessed roughly by using export statistics of the main producers.

Development of consumption of all spices taken together in grams per capita in individual countries is shown in Table II. Estimates of all nine major spices selected are available for only three countries—Germany, The United Kingdom, and the United States. It is not possible to say for the other countries whether absence of consumption estimates implies an absence of consumption, although the absolute levels of consumption are not strictly comparable, their trends can be compared, and in fact they show a similar pattern in each case (Food and Agriculture Organization, 1962). Of course, the nature and extent of spice consumption varies from country to country. Enquiries made by the spice trade and manufacturers' associations elicited estimates that the proportion of spices sold directly to

TABLE II PER CAPITA CONSUMPTION OF MAIN SPICES IN SELECTED COUNTRIES[a,b]

Country	1937–1939	1951–1953	1958–1960	Spices[c]
Canada	141	92	108	Pepper, clove, ginger, nutmeg, mace
France	64	35	58	Pepper, clove
Germany	127	69[d]	108[d]	All nine selected spices
India	86	77	74	Pepper, clove, cinnamon, cassia, cardamom, ginger, capsicum
Indonesia	219	139	229	Pepper, clove
Italy	42	18	37	Pepper
Sweden	161	65	200	Pepper, cardamom
United Kingdom	129	83	105	All nine selected spices
United States	212	151	161	All nine selected spices

[a] From Food and Agriculture Organization (1962).

[b] Values in grams.

[c] Spices included are capsicum, cardamom, cassia, cinnamon, clove, ginger, mace, nutmeg, pepper.

[d] Western Germany only.

the retail trade is about 60% in the United States, 30 to 50% in the United Kingdom, and 15% in Germany. Of the remainder, 50% or more is thought to be consumed by the meat processing industry (mostly sausage manufacturers), and the rest is consumed by bakers and by the pharmeceutical and perfumery industries (Commonwealth Secretariat, 1969).

2

Properties and Uses

The different properties and uses of spices and condiments were known even to the ancient people in prehistoric times. Their uses have been inscribed on the walls of ancient Egyptian temples and burial chambers. They are often described in the Scriptures too. During the past fifty years, several reports have appeared in the literature on the different uses of spices, such as in the flavoring of foods, and also on their properties, such as bacteriocidal, bacteriostatic, fungistatic, antifertility, anthelmintic, and medicinal. Other reports have discussed their role in animal physiology with special reference to the secretion of saliva and amylasis, gastroenterology, the effect of spices on the cortex of the suprarenal gland, the circulatory organs, blood pressure, blood cholesterol, blood sugar, blood coagulation and fibrinolysis, prophylaxis and therapy of thrombosis, spasmolytic and diuretic activity, uses in rheumatism, leprosy, and tuberculosis, and other pharmacological and medicinal properties. These aspects will be briefly reviewed in this chapter.

I. FLAVORING PROPERTIES

The aromatic ingredients of spices improve the flavor of our foods. Whereas some spices are warming and pervasively aromatic with a tangy bitterness that gives them zest and a refreshingly clean taste, others have a subtle and suave flavor that intrigues the palate and stimulates the appetite. Some exceptional spices are very pungent, bitingly hot, racy, and strong smelling; others are crisp and succulent, with almost no pungency. The aromatic and pungent principles

that render spices valuable are contained in their volatile oils and oleoresins. The volatile oils, called essential oils, are responsible for the characteristic aroma of spices. Some of the spices that contain essential oils in considerable proportion are cloves, turmeric, cumin, caraway, cardamom, coriander, cinnamon, fennel, fenugreek, nutmeg, and mace. The oleoresin content (nonvolatile extract) of the spices, which can be extracted with alcohol or other chemical solvents, is responsible for the typical taste and flavor peculiarities of the spices concerned.

A vast store of knowledge on spices and condiments and an armory of answers to the specialized problems of food manufacturing and processing industries have been built up over the years. Natural and synthetic spiced foods and other flavors in industrial quantities and spice seasonings in distinctive blends can now be produced. Spices and condiments are being sought after for more definite uses than previously to enhance the innumerable varieties of instant cooked and semicooked foods, soups, sauces, pickles, chutneys, breads, beverages, canned curried vegetable and meat products, and fruit jellies, jams, and preserves that our modern food industry is producing. A healthy appetite appreciates the presence of an appropriate pungency and flavor added by means of spices and condiments in the food. The need is even greater for a jaded appetite, where the palate or digestion has been weakened by sickness, environment, or other causes.

The food industry, by using different types of flavoring extracts and spices, proves their versatility and value in many ways. In the forefront of the flavoring field come the essential oils, essences, liquors, and fruit juices. The most widely used essential oils in food flavoring are those derived from spices and herbs, such as anise, cassia, cinnamon, celery, cloves, ginger, nutmeg, mint, and thyme. The use of spices and their extracts is not limited to the flavoring of foods, however. It extends to making the appearance of the foods pleasing to the eye by the addition of coloring materials derived, for example, from turmeric, paprika or chilies, and saffron.

Space does not permit a detailed listing of the numerous reports published from time to time on the uses of spices in the flavoring of foods. Neale and Klis (1964) have published an exhaustive check list of about 140 different food products grouped under seventeen main categories such as bakery products, beverages, cheeses, condiments, sauces, chutneys, etc., confections, ethnic dishes, fish and seafoods, fruits and fruit-based products, gravies and curries, meat products, pickles, poultry products, salad dressings, sausages, soups, and many vegetables and vegetable products. Such uses of spices have also been described (Anonymous, 1969).

II. ANTIOXYGENIC PROPERTIES

In view of the fact that natural spices are widely used in a variety of food products, it is important to know the effects they have on the keeping qualities of

such products. A number of studies have been made on the bactericidal and bacteriostatic properties of spices to evaluate their effectiveness in preventing or retarding spoilage caused by microorganisms. A few scattered reports have also appeared in the literature on the antioxidant effect of spices on fats in certain foods. Thus, Maveety (1938) patented the use of certain spice fractions (spice oils such as clove oil) for the prevention or retarding of rancidity in edible oils and fats. DuBois and Tressler (1943) reported that black pepper, sage, mace, and ginger retarded the development of rancidity in frozen ground pork and beef. Antioxidant properties were attributed to black pepper, red pepper, and sage in stored frozen pork sausage (Atkinson *et al.*, 1947). Ramaswamy and Banerjee (1948) found that the phenolic pigment curcumin, present in turmeric, was responsible for the antioxidant properties of this spice. Sethi and Aggarwal (1950, 1952, 1956) indicated that chilies, cinnamon, ginger, turmeric, nutmeg, mace, black pepper, cloves, onions, and betal leaf, when heated with groundnut (peanut) oil to incipient charring of the spice at 275° to 280°C (527° to 536°F), all retarded the development of peroxides and free fatty acids, when the oil was aerated at 100°C (212°F). However, as a process for adoption by the industry, it is considered uneconomical. Besides, such treatment might destroy appreciable quantities of some types of natural antioxidants. However, numerous investigators have also shown that heat treatment of many biological materials, either before or after addition to a fat or oil, produces fat-stabilizing substances. The results reported by Sethi and Aggarwal (1950, 1952) are probably due, in large measure, to the development of such stabilizers produced during heating of the materials at high temperature. The same authors further reported that dried ginger and nutmeg had weak antioxidant properties. The highest activity was that from onion extract, which, however, was much less than that from betel leaves (Sethi and Aggarwal, 1957).

Chipault *et al.* (1952) examined seventy-eight samples representing thirty-six spices for their antioxidant properties. First, the stabilizing effect of the ground spices in lard was tested by the active oxygen method at 98.6°C (210°F). Also, because the ground spices contain relatively large proportions of antioxygenically inert substances and because the antioxidant properties of the spices may be due to combinations of several primary and synergistic antioxidants and also pro-oxidants, at least one sample of each spice was extracted successively with two solvents to separate and concentrate some of the active compounds. The fractions so obtained were also tested in lard by the active oxygen method. The results of the stability measurements for the thirty-six spices and equivalent solvent fractions in terms of antioxidant indices are given in Table III. In most cases, there was little difference between the antioxidant indices of various samples of the same spice, indicating that, in general, the antioxidant activities of spices tend to be relatively independent of their origin.

With only one exception (ginger) in the seventy-eight samples examined, all

TABLE III ANTIOXIDANT PROPERTIES OF GROUND SPICES AND OF PETROLEUM ETHER AND ALCOHOL-SOLUBLE FRACTIONS[a]

Spice	Antioxidant index[b]		
	Ground spice	Petroleum ether-soluble fraction	Alcohol-soluble fraction
Allspice	1.8	1.1	1.0
Aniseed	1.9	1.1	1.2
Basil leaf	1.2	1.0	0.8
Bay leaf	2.1	1.3	1.3
Cardamom	1.3	0.8	1.1
Caraway	1.8	0.9	1.2
Cassia	1.4	1.0	1.3
Celery seed	1.2	0.9	1.1
Chili	1.5	1.3	1.4
Cinnamon	1.3	0.9	1.4
Clove	1.8	1.4	1.7
Coriander	1.3	0.9	1.0
Cumin	1.3	1.1	1.2
Dill	1.3	0.8	1.0
Fennel	1.3	0.9	1.1
Fenugreek	1.6	0.9	1.2
Ginger	1.8	1.0	1.5
Mace	2.6	1.1	1.8
Marjoram	2.2	1.0	1.3
Mustard	2.0	1.1	1.4
Nutmeg	3.1	1.0	2.2
Oregano A	3.8	1.4	2.7
Oregano B	3.3	1.8	2.6
Paprika	2.5	1.2	1.4
Pepper, black	1.4	1.0	0.9
Pepper, red	1.5	1.1	1.3
Pepper, white	1.2	1.0	0.9
Poppyseed	1.2	0.7	1.2
Rosemary A	17.6	2.2	5.5
Rosemary B	17.6	2.5	5.2
Sage A	14.2	2.2	5.4
Sage B	16.5	2.2	5.2
Savory	1.6	1.3	1.3
Thyme	3.0	1.5	3.0
Turmeric A	2.9	1.0	2.0
Turmeric B	2.0	1.0	2.1

[a] From Chipault *et al.* (1952).

[b] Active oxygen method at 98.6°C, employing as substrate prime steam lard with a stability of 6.5 hours.

the ground spices exhibited an antioxidant effect on lard when tested by the active oxygen method at 98.6°C (210°F). Rosemary and sage exhibited particularly pronounced antioxidant effects. Citric acid exerted a synergistic effect on the antioxidant properties of some spice fractions, and no effect on others.

In general, the antioxidant activity of the spices in pie crust was considerably less than that in lard, owing probably to a partial destruction of antioxidants during baking. The antioxidants of some of these spices appeared to be less susceptible to destruction by heating than those of other spices. The studies on "carry-through" antioxidant activity in baked pie crusts revealed that most of the spices were only slightly antioxygenic, six were pro-oxygenic, and four prolonged the stability of pie crusts by factors of 2 to 4.5 (Chipault *et al.*, 1955). This was in agreement with the results of others (Higgins and Black, 1944; Lundberg *et al.*, 1944), who had found that typical phenolic antioxidants were much less effective in protecting baked goods than in protecting lard.

Mattil and Black (1947) related the water solubility of an antioxidant to its ability to stabilize the fat in baked goods. Their results indicated that in baked goods, where the fat containing the antioxidant is in contact with water, if the solubility of the antioxidant in water is appreciable, it may be extracted from the fat and thus be ineffective. Lehmann and Watts (1951) reported that of the five phenolic antioxidants tested in dry lard, and in lard in contact with an aqueous phase, three had approximately the same effectiveness in both substrates, one was more effective in the dry lard, and one was more effective in the lard–water system.

Spices are seldom used in pure fats or in pie crusts. Furthermore, antioxidants used in pie crusts are not only in contact with an aqueous phase but are also subjected to relatively high temperatures for various lengths of time, a treatment that may well destroy much of the antioxidant. Spices, however, are used in a number of other fat-containing food products in which the fat phase is in intimate contact with an aqueous phase, and many of these products are not subjected to any heat treatment. Chipault *et al.* (1955), therefore, studied the antioxidant effectiveness of spices in a two-phase aqueous–fat system. They observed that the best spice in the oil-in-water emulsion system was cloves. The samples of cloves in three different emulsions gave antioxidant indices of 72.5, 82, and 103, approximately fifty times as large as that obtained with lard alone (Table IV). When tested in lard, rosemary and sage were the best antioxidants of all the spices studied, and their activities were approximately equal. In pie crusts, however, rosemary was definitely superior to sage; in the emulsion studies, rosemary was also definitely superior to sage. In addition to cassia, cinnamon, allspice, turmeric, and cloves, it was found that ginger, mace, nutmeg, and oregano were also better than sage; black and white peppers were nearly equal to sage in protecting emulsions. All these spices were much inferior to sage when studied in lard (Chipault *et al.*, 1955).

TABLE IV ANTIOXIDANT PROPERTIES OF SPICES IN AN OIL-IN-WATER EMULSION[a]

Spice	Mean stability (hours)	Anti-oxidant index[b]	Spice	Mean stability (hours)	Anti-oxidant index[b]
Control	12.5	—	Mace	294.0	12.8
Aniseed	59.5	4.8	Control	19.0	—
Basil	49.5	4.0	Sage A	148.0	7.8
Cardamom	64.5	5.2	Sage B	133.0	7.0
Control	17.0	—	Rosemary A	193.5	10.2
Caraway	49.0	2.9	Rosemary B	217.0	11.4
Celery seed	55.0	3.2	Control	13.0	—
Chili	42.0	2.5	Paprika	53.0	4.1
Cassia	148.0	8.7	Oregano[c]	103.0	7.9
Cinnamon	142.0	8.4	Turmeric[c]	207.0	15.9
Control	15.5	—	Turmeric	384.0	29.6
Fenugreek	26.0	1.7	Control	15.0	—
Mustard	36.0	2.3	Dill	32.0	2.1
Poppy	31.5	2.0	Bay leaf	66.0	4.4
Ginger	135.0	8.7	Red pepper	49.0	3.3
Control	13.0	—	Control	15.0	—
Coriander	33.0	2.5	Thyme	101.5	6.8
Cumin	33.5	2.6	Savory	118.0	7.9
Fennel	35.0	2.7	Allspice	250.0	16.7
Control	23.0	—	Clove	1228.0	81.8
Marjoram	94.0	4.1	Control	13.0	—
Black pepper	140.0	6.1	Clove	1340.0	103.0
White pepper	115.0	5.0	Control	17.8	—
Nutmeg	211.0	9.2	Clove	1290.0	72.5

[a] From Chipault *et al.* (1955).

[b] Ratio of mean stability of sample to mean stability of control.

[c] Emulsion unstable.

All the spices studied protected the oil-in-water emulsion against oxygen absorption. In most cases, the spices were more effective in the oil-in-water emulsions than in plain lard or baked pie crusts (Table V), but the order of their effectiveness was different for the different substrates (Chipault *et al.*, 1956). Cloves were extremely effective in preventing oxidation of the emulsions. Other spices with antioxidant indices greater than 5.0 were allspice, cardamom, cassia, cinnamon, ginger, mace, nutmeg, oregano, black pepper, white pepper, rosemary, sage, savory, thyme, and turmeric (Table IV).

Many fat-containing food products in which spices are used have a fat phase in intimate contact with an aqueous phase. Other factors, such as the physical state of the fat–water system, the kind of fat, the pH of the aqueous medium, or the presence of other substances (for example, pro-oxidants, antioxidants, synergists)

TABLE V ANTIOXIDANT PROPERTIES OF SEVERAL SPICES IN SIX SUBSTRATES[a]

Spice	Lard	Pie crust	Oil-in-water emulsion	Ground pork		Mayonnaise		French dressing	
				−5°C	−15°C	A	B	37°C	63°C
Spice concentration in fat (%):	0.2	0.2	0.1	0.25	0.25	0.20	0.56	1.0	1.0
				Antioxidant indices					
Allspice	1.8	1.1	16.7	5.3	10.0	1.4	3.1	1.1	1.2
Clove	1.8	1.3	85.8[b]	5.3	10.0	2.0	4.6	2.0	1.2
Oregano	3.8	2.7	7.9	7.2	3.7	8.5	9.1	2.6	2.4
Rosemary	17.6	4.1	10.2	5.3	10.0	2.2	—	—	—
Sage	14.2	2.7	7.8	5.3	10.0	2.4	3.4	2.2	2.2
Thyme	3.0	1.9	6.8	6.0	3.2	1.8	—	—	—

[a] From Chipault *et al.* (1956).

[b] Average of three determinations.

affecting the stability of fats, may markedly influence the antioxidant effects of spices in different foods (Chipault *et al.*, 1956).

Chipault *et al.* (1956) further studied the antioxidant properties of several spices in different types of foods such as ground pork, two types of mayonnaise, and a French dressing. Allspice, cloves, oregano, rosemary, sage, and thyme increased the stability of all the fat substrates in which they were tested, but the relative effectiveness of the spices varied with the different substrates. Sage and rosemary were more effective than the other spices in lard alone. Cloves were a powerful antioxidant in simple oil-in-water emulsions and in ground pork, but oregano was more effective in mayonnaise and French dressing.

Bickoff (1951) reported the antioxidant properties of vanillin in preserving carotene. Narayanan *et al.* (1957) also found antioxidant properties of vanillin when incorporated in shortenings (m.p. 37°C).

Sahasrabudhe and Bhatia (1953) reported the antioxygenic effect of ground spices (mustard, ginger, nutmeg, cloves, black pepper, and combinations thereof, with or without citric acid and salt), which increased either when the concentration of spice was increased or when the spices were treated with hot oil. Heating the oil to 250°C probably destroyed the antioxygenic principles in spices.

Citric acid had a synergistic effect in some spices, particularly in chilies. Combinations of turmeric, chilies, ginger, and citric acid had a remarkable antioxygenic effect. The results appear to be of practical value in the processing of spiced cakes and in the preparation of oil pickles.

Lewis and Watts (1958) observed that the juices of onions and garlic exhibited antioxidant properties. The extracts prepared from the onion skins had a more pronounced effect, especially that prepared from yellow onion skins. Onion skin extracts also exhibited copper chelating abilities and varying degrees of synergism with ascorbic acid. These activities are presumed to be due to flavonoid compound from the onion skin. The 2,4,5-trihydroxybutyrophenone (THBP) exhibited strong copper chelating activity, antioxidant properties, and synergism. Furthermore, both THBP and the extract of yellow onion skins were effective as antioxidant when incorporated in cooked meat, both in the absence and in the presence of ascorbic acid.

Kihara and Inoue (1962) stated that the thiobarbituric acid reaction of lard oil, with which seventeen powders were mixed separately, was followed up by them for six months. Clove, thyme, pimento, ginger, laurel, and mace had a relatively strong antioxidant activity. Cloves, pimento, and ginger had antioxidant activity in cookies. Cinnamon, ginger, and cloves were effective as antioxidants in potato and sweet potato chips.

Herrmann (1962) reported that the known antioxidant action of cloves, oregano, rosemary, sage, thyme, marjoram, and savory was due in part to the labiatic acid which they contain and is depside of caffeic acid and *p*-hydroxyhydrocaffeic acid. Labiatic acid, caffeic acid, powdered sage, rosemary, and

marjoram leaves inhibited rancidity of lard at 20°C, 37°C, and 50°C. The seasonings also contain flavonol, which may impart some antioxidant activity.

Cort (1974) studied the antioxidant activity of several spices (allspice, cloves, mace, marjoram, nutmeg, oregano, rosemary, sage, thyme, and turmeric). By their direct addition (without any extraction), they all exhibited antioxidant activity. The highest activity was shown by cloves, followed, in decreasing order, by turmeric, allspice, and rosemary. In experiments on extraction with water and hexane, only cloves, marjoram, and thyme showed activity. When extracted with ethanol, they all showed increased activity, especially mace and rosemary. Cloves contain 15% eugenol and have an antioxidant activity equivalent to 14% of BHT activity; since eugenol has 90% of the activity of BHT, most of the activity of cloves is due to eugenol. Eugenol isomerizes to isoeugenol, which has a more pleasant aroma than that of eugenol, but equivalent antioxidant activity. Isoeugenol is found in nutmeg.

Oregano was also found to have an antioxidant activity equivalent to 6% of that of BHT (Cort, 1974).

Curcumin—the major pigment found in turmeric—had 75% of the activity of BHT. The curcumin content of turmeric is about 8%, which probably accounts for the 6% antioxidant activity of turmeric (Cort, 1974).

Revankar and Sen (1974) studied the antioxidant effect of a spice mixture and its oleoresin. Both of them had an antioxidant effect, the latter being more effective. Scher and Ivanov (1976) reported antioxidants in fennel oil.

Palitzsch (1974) investigated the effect of natural spices, spice extracts, essential oils, and synthetic antioxidants on the breakdown of pork fat and model lipid systems. Oil-free spice extracts were often found to be very antioxidative. A petroleum ether extract of rosemary particularly inhibited oxidation of fats. Chang *et al.* (1977) reported natural antioxidants from rosemary and sage.

III. PRESERVATIVE ACTION

Numerous workers have dealt with the preservative action of several spices and condiments on foods. Although some of them do exert a definite inhibiting action against microorganisms, many of them are without any significant action. Most of the workers have also agreed that mustard, cinnamon, and cloves are among the most potent spices in checking the growth of molds, yeasts, and bacteria. Their preservative qualities are due to the presence of some active antimicrobial principles contained in them, which are briefly discussed below.

A. The Major Active Principles in Spices and Spice Essential Oils

Cloves contain essential oil ranging from 15 to 20%, of which 85 to 92% is eugenol—the major active principle that inhibits the growth of microorganisms, thereby helping in the preservation of some foods.

Cinnamon yields 0.5 to 3.5% volatile oil, which contains 42 to 75% cinnamic aldehyde, the major active principle. Similarly, ajowan contains thymol (45 to 55%), turmeric contains curcumin, and garlic contains allicin.

All mustards contain certain pungent principles, which differ in different varieties, and which form the basis for the use of mustard as a condiment, a preservative, or a medicinal component in drug preparations. The seeds of different species of mustard yield varying percentages of a pungent volatile oil containing allyl isothiocyanate. It is the bactericidal properties of this compound that are responsible for the preservative action of mustard.

Black mustard and, to a lesser extent, brown mustard contain the glucoside sinigrin, or potassium myronate, which, on enzymatic hydrolysis, yields one mole each of allyl isothiocyanate, dextrose, and potassium acid sulfate according to the following reaction (Kosker *et al.*, 1949):

$$\underset{\text{Potassium myronate (sinigrin)}}{C_{10}H_{16}NS_2KO_9} \xrightarrow{H_2O} \underset{\text{Allyl isothiocyanate}}{CH_2CHCH_2NCS} + \underset{\text{Dextrose}}{C_6H_{12}O_6} + \underset{\text{Potassium acid sulfate}}{KHSO_4}$$

Different names such as myrosin, myrosinase, and sinigrinase have been suggested for the enzyme system responsible for the splitting of the thioglucosides, according to Mart and Halley (1932).

Winton and Winton (1945) pointed out that white mustard, although practically free from volatile oil, contains a glucoside, sinalbin, which, under enzymatic hydrolysis, splits into sinalbin mustard oil (*p*-hydroxybenzyl isothiocyanate), dextrose, and sinapin hydrogen sulfate by the following reaction:

$$\underset{\text{(sinalbin)}}{C_{30}H_{42}N_2S_2O_{15}} \xrightarrow{H_2O} \underset{\text{(sinalbin mustard)}}{C_6H_4(OH)CH_2NCS} + \underset{\text{(sinapin hydrogen sulfate)}}{C_{16}H_{24}NO_5HSO_4} + \underset{\text{(dextrose)}}{C_6H_{12}O_6}$$

Sinalbin mustard oil is not volatile and does not possess the same characteristics as those shown by allyl isothiocyanate.

B. Preservative Action of Spices and Spice Essential Oils

Bitting (1909) described the mechanism of action of mustard in the prevention of mold growth. Hoffmann and Evans (1911) observed that mustard, cloves, and cinnamon are about equal in their preserving efficiency, and that 0.5 g of mustard would preserve 100 g of applesauce for four months. Rippetoe and Wise (1912) found that, in addition to oils of mustard, cloves, and cinnamon, other spice oils, such as cardamom, cassia, coriander, cumin, mace, marjoram, pimento, and thyme, had some preservative action, whereas oils of celery seed and black pepper were without action. Oils of caraway, ginger, and nutmeg had a questionable effect. Hoffmann and Evans (1911), using applesauce as medium, found that

ground cinnamon, cloves, and mustard flour had high preservative properties, pimento and nutmeg had some, and ginger and pepper had none. They found mustard to be the most powerful. Bachmann (1916), however, concluded from her experiments that the quantities of spices normally used in the kitchen have little preservative action, but when used in large quantities they inhibit the growth of molds.

Grove (1918) observed that mustard, cloves, and cinnamon had the highest preserving action against spoilage in tomato ketchup, and the work of Lahache (1919) indicates that these spices were definitely stronger than even chemical preservatives.

Bitting (1920) stated that, in household practice, black and white mustard seeds are often added to cider and other homemade beverages in order to preserve them. In his studies on the growth of molds, he observed that mustard produced starvation and also disorganization of the protoplasm, as indicated by the stunted and distorted hyphae, the swollen and nonhyaline appearance of the protoplasm, and a tendency in the penicillium to form fruiting heads almost directly from the hyphae developed from the germinated spore. The hyphae formed matted tufts, difficult to separate, and with blunt, distorted fruiting heads. A peculiarity of the mustard was that the active principle causing the abnormal growth produced the effect when present in minute quantities, as in a 0.1% solution of ground mustard or 1.0% of seeds.

Campbell (1920) found that 0.1% of benzoate of soda did not preserve cider. The use of sulfites (sulfur dioxide or sulfurous acid) gave an undesirable taste to the product. Ground mustard used in conjunction with benzoate of soda, however, preserved cider, and amounts of less than 0.1% of either could be used to advantage. Four ounces each of ground mustard and benzoate of soda were recommended for the preservation of 50 gallons of cider.

Janke (1923) used mustard oil to suppress the growth of surface yeasts, which play an important role in the pickling industry. His experiments showed that a minimum of 0.006% of mustard oil was required to suppress the development of mycoderma, whereas for acetic acid bacteria, 0.008% by volume was required. He further found that 0.003% of mustard oil in 2.5% vinegar was sufficient to preserve tomatoes.

Various workers have examined the value of mustard oil in wine making. Thus, Moreau and Vinet (1924) have shown that 50 mg/liter sterilizes fermenting wine musts. Roos and Hughes (1924) found that as little as 1.5 mg/liter preserves finished wines from pathological fermentation. They state that this is the maximum that can reasonably be present, owing to its effect on flavor. Similar conclusions were reached by Delage (1923), but Ferre (1925) is of the opinion that 1 mg of mustard oil per liter is the maximum amount that can be allowed because of the effect on the flavor, and this is insufficient to protect the wine against secondary fermentation.

Myers (1926) found that mustard, cinnamon, and cloves, in that order, were fungicidal toward a yeast isolated from man.

Prasad and Joshi (1929), making inherent microbial population in fresh fruit a target for control, assigned a preservative role to cloves at a 2% level when used for pickling. The addition of 20% salt to pickles was reported to completely suppress the growth of spoilage organisms.

James (1931, 1938) examined the antiseptic action of mustard, cinnamon, and cloves against *Escherichia coli* and showed that antisepsis occurred only when large proportions of spices were used. Whereas mustard showed only slight inhibitive action, cloves and cinnamon, in sufficient quantities, had some antiseptic and even germicidal action. His experiments revealed that 480 mg of mustard exerted a germicidal efficiency against one to two million organisms of the coliform group in 15 ml of nutrient broth.

Corran and Edgar (1933) reported the following results of a comprehensive investigation on the potencies of certain spices and spice oils compared with chemical preservatives with respect to inhibition of yeast fermentation:

1. The effects of various spices and their essential oils on fermentation by ordinary yeast *(Saccharomyces cerevisiae)* were studied (Table VI). Of the spices studied, mustard flour was found to be the most efficacious, being followed in order of efficacy by cloves and cinnamon. Little or no preservative action could be detected with cardamom, cumin, coriander, caraway, celery seed, pimento, nutmeg, ginger, thyme, bay leaves, marjoram, savory, rosemary, black or cayenne peppers.
2. The herbs examined (caraway cumin, marjoram, rosemary, sage, savory, etc.), as well as black pepper, acted as if they contained yeast stimulants (probably the herbs themselves had a high initial yeast count).

TABLE VI PRESERVATION ACTION OF SPICES AGAINST YEAST FERMENTATION[a]
(Results after four days of incubation at room temperature, 0.5% yeast)

Spice	Concentration 1:100 (% loss of glucose)	Concentration 1:400 (% loss of glucose)
Brown mustard flour	0	18.5
Clove (ground)	0	60
Cinnamon (ground)	77	85
Ginger (ground)	83	85
Pimento (ground)	82	85
Nutmeg (ground)	84	85
Control	85	85

[a] From Corran and Edgar (1933).

3. Volatile oil of mustard was similarly found to be a stronger preservative than the essential oils of any of the other above-named spices or herbs. Cinnamon oil followed it in efficacy, and the oils of cloves, thyme, and bay leaves came next, being approximately equal in power. The remaining oils were of no practical use, although some slight action was manifested by caraway and cumin oils.

4. Mustard and its volatile oil were definitey stronger than either of the two chemical preservatives, and at concentrations where the chemical preservatives had no inhibiting effect, volatile oil of mustard still completely prevented all fermentation. Cloves had a potency of the same order as that of benzoic acid, and superior to that of sulfur dioxide. It was found that 0.05% of volatile oil of mustard was sufficient to prevent fermentation whereas 0.2% of benzoic acid and 0.1% of sulfur dioxide were necessary to achieve the same results (Table VII). These experiments further indicated that brown mustard flour was superior to benzoic acid or sulfur dioxide when the latter were added in the maximum amounts permitted under the law.

Another study of the relative toxicity of mustard oil, and related sulfur compounds, to certain fungi was made by Walker *et al.* (1937). They found that mustard oil had a marked toxic effect on molds, with *Aspergillus niger* showing the least sensitivity to it among the fungi studied. Sixty parts per million of mustard oil inhibited 59% of mold growth on the basis of the dried weight of mycelial mats when molds were grown on 25 ml of Czapek's solution.

Perazza and Prado (1937) observed that 25 to 50 mg of mustard oil per liter of grape juice prevented its fermentation if the juice was filtered and stored in tightly sealed containers. They also demonstrated that 1.5 to 2 mg of mustard oil per liter of grape juice prevented mold growth but did not stop fermentation.

TABLE VII COMPARATIVE PRESERVATIVE ACTION OF VOLATILE MUSTARD OIL AND CHEMICAL PRESERVATIVES[a]

(Results after two days of incubation at room temperature, using 0.5% yeast)

Chemical preservatives	Concentration (%)	Parts per million	Loss of glucose (%)
Volatile oil of mustard	0.05	500	0
Benzoic acid	0.2	2000	0
Sulfur dioxide	0.1	1000	0
Chloroform	0.1	1000	86
Toluene	0.1	1000	84
Phenol	1.0	10,000	0
Control		—	86

[a] From Corran and Edgar (1933).

Fabian *et al.* (1939), using a number of bacterial species as test organisms, concluded that there was considerable variation in the resistance of different organisms to the same spice and of the same organism to different spices. Their results showed that ground cinnamon and cloves were the only spices that were inhibitory in low concentration for bacteria. Ground peppercorn and allspice were inhibitory in 1% concentration, and mustard, mace, nutmeg, and ginger in 5% concentration. One-tenth percent of a 50% emulsion of oil of mustard gave some inhibition, while 1% of a 50% emulsion was completely inhibitory.

Foter (1940) made a comparative study of the bactericidal effects of allyl isothiocyanate, methyl isothiocyanate, and ethyl isothiocyanate. Methyl isothiocyanate was the most effective of the three compounds studied. He emphasized that the most complete or marked bactericidal effect resulted, in most cases, from concentrations of these compounds of 1 to 10^5 and 1 to 10^6. The organisms most resistant to these compounds were *Escherichia coli* and *Staphylococcus aureus,* while *Serratia marcescens, Bacillus subtilis,* and *Bacillus mycoides* were the least resistant.

Oztune (1940) found that 1,5% and 2.0% mustard exerted a definite preservative effect in fruit juices. He further indicated that, in Belgium, Azymol, a preparation containing allyl isothiocyanate, has been used in the preservation of fruit juices.

Akman (1942) included mustards among other preservatives that could be used for fruit juices. According to him, large quantities of mustard were used by the Romans in the preservation of juices from one season to another, as well as in the preparation of concentrated juices. Earlier philosophers considered mustard to be "wholesome for the body," but the reason for the effectiveness of mustard in its various uses remained unexplained until 1909.

The procedure for using mustard as a preservative in grape juice varies in different locales. The more common practices used in Turkey have been described by Tekeli (1943).

Blum and Fabian (1943) studied various spice oils and their components for controlling microbial surface growths occurring in fermented beverages such as beer and wine, as well as in products like olives, pickles, sauerkraut, and salted vegetables. They concluded that, of all the chemicals and spice oils tested, emulsified oil of mustard was the best for preventing scum formation on pickles and sauerkraut. It exerted a definite inhibitory effect on the growth of *Acetobacter aceti, Saccharomyces ellipsoideus, Saccharomyces cerevisiae,* and *Mycoderma vini.*

Webb and Tanner (1945) found spice oils (because of the greater concentration of the respective active principles present) to be more effective than whole or ground spices in preventing yeast growth in laboratory media. Oils of cinnamon, mustard, cloves, allspice, bay, wintergreen, and peppermint of 0.1% concentration retarded and in most cases completely inhibited the microbial growth. In

concentrations over 1%, mustard, cinnamon, and clove oils were germicidal to yeasts in spice oil–dextrose agar plates. Tested by the cup–plate method, oils of allspice and bay were also germicidal to yeasts. Anise and onion oils were classified as bacteriostatic.

Kosker *et al.* (1949) reviewed the history and the available literature on the use of mustard as a preservative and also reported the results of their experiments on the use of whole and ground mustard and of natural and synthetic mustard oil in the preservation of apple cider and grape juice. They reported that mustard was ineffective in checking mold growth even at a 10% level, although it delayed fermentation in apple cider for 24 hours at 0.5% concentration. The addition of 11 to 22 ppm of mustard oil or its equivalent in ground mustard to fresh apple cider was found to exert a favorable preservative effect in retarding fermentation (Kosker *et al.*, 1949).

Anderson *et al.* (1953) tested a number of essential oils for the inhibition of growth of acid food spoilage organisms (bacteria and yeasts) in dextrose broth. Mustard, garlic, onion, and cinnamon oils were the most effective. In acidified broth, the inhibitory action of most of the oils was increased, but for one strain of yeast the quantity of oil necessary to inhibit growth in the acidified broth was as great as or greater than that required in broth at pH 7.2.

Anand and Johar (1957) reported that cinnamon and cloves in small doses had an inhibitory action and did not permit the germination of mold spores. In the absence of 16 g of salt per 100 g of mango pulp, 0.3% cinnamon and 0.2% cloves were required to inhibit the mold (Table VIII); in the absence of salt, 3% cinnamon and 0.6% cloves were required to get the same effect. The effect of cloves as a preservative, like cinnamon, varied with the concentration of salt, as the salt played a complementary role. The effect of different proportions of sixteen condiments required to inhibit the growth of *Aspergillus niger* in salted mango pulp is given in Table IX.

The antiseptic effect of extracts of spices such as garlic (10% and 20%), horseradish (10% and 20%), onion skins (10% and 20%), and mustard (pulverized) on carrots stored in sand or in wood shavings was studied by Grimm (1959), who observed that the values for wholesome carrots at the end of 160 days in sand and 55 days in wood shavings were for onion 85.8 to 90%, for garlic 85%, for horseradish 94.9%, and for mustard 92%. In the control sets, the corresponding figures were 72.5% and 82.4%.

Subba Rao *et al.* (1963) reported that an emulsion containing active ingredients such as acetic acid, orange peel, mustard, and turmeric powders could prevent molds as well as yeasts in pickles containing low concentrations of salt.

The foregoing review indicates that the preservative action of some spices may be of practical importance, but it must be recognized that the quantities that can be used in foods are often strictly limited by flavor considerations.

TABLE VIII EFFECT OF DIFFERENT CONCENTRATIONS OF CINNAMON AND CLOVE ON THE CONTROL OF *ASPERGILLUS NIGER* IN MANGO PULP[a]

Quantity of spice (g/100 g pulp)	Growth of spores in the presence of salt[b] (g/100 g pulp)				
	0	4	8	12	16
		Cinnamon			
0.0	+	+	+	+	+
0.3	+	+	+	+	−
0.6	+	+	+	−	−
0.9	+	+	−	−	−
1.6	+	−	−	−	−
3.0	−	−	−	−	−
		Clove			
0.0	+	+	+	+	+
0.2	+	+	+	+	−
0.3	+	+	−	−	−
0.4	+	−	−	−	−
0.6	−	−	−	−	−

[a] From Anand and Johar (1957).

[b] +, growth positive; −, no growth.

C. Effect of Spice Essential Oils on Thermal Resistance of Microorganisms

The inhibiting effect of several essential oils on many life processes has been turned to our advantage by the use of these compounds as bactericidal and fungicidal agents. The effect of certain spice oils on the thermal resistance of microorganisms has also been investigated.

Baumgartner and Hersom (1956) suggested that essential oils of spices such as mustard, which are known to be toxic to microorganisms at normal growth temperatures, may have at processing temperatures an appreciable effect in reducing heat resistance even in low concentrations.

Kosker *et al.* (1951) demonstrated that 10 ppm of allyl isothiocyanate (volatile oil of mustard) added to buffer solutions and to apple and grape juices caused a marked reduction in the thermal resistance of *Aspergillus niger* and *Saccharomyces ellipsoideus*. The effect was less marked against Bacillus thermoorganisms than had allyl isothiocyanate.

Anderson *et al.* (1951) have shown that the addition of a dill spice oil composed of both garlic and dill seed oils at an acceptable flavoring level of 258 ppm and of a mustard oil at a level of 20 ppm considerably reduced the thermal death time of a test yeast suspended in an acidified brine. It appears from the results of these experiments that certain spice oils may have a considerable effect on the

TABLE IX EFFECT OF DIFFERENT PROPORTIONS OF CONDIMENTS REQUIRED TO INHIBIT THE GROWTH OF *ASPERGILLUS NIGER* IN SALTED MANGO PULP[a]

Condiment	Conventional proportions in pickle recipes (g/100 g mango slices)	Inhibitory levels for mold spores (g/100 g pulp)[b]
Aniseed	0.5–3.0	10.00
Asafetida (Hing)	0.1–0.2	0.50
Asafetida oil	Not used	0.04
Black pepper	0.0–3.0	10.00
Caraway (Zeera)	0.0–1.5	4.00
Chili	1.5–10.0	10.00
Cinnamon	Not used	0.30
Clove	Not used	0.20
Fenugreek (methi)	0.2–10.0	10.00
Garlic	Not used	0.50
Ginger (fresh)	0.0–10.0	15.00
Ginger (dry)	Not used	2.00
Kalaunji (onion seeds)	0.0–3.0	10.00
Mustard powder	0.0–4.0	10.00
Mustard oil	Covered with thin layer Growth positive in covered pulp	
Salt	5.0–25.0	26.00
Turmeric	0.3–3.0	6.00

[a] From Anand and Johar (1957).

[b] Mango pulp containing 15 g of salt per 100 g of pulp.

thermal stability of yeast cells, but their action is less marked against spores of bacteria. Within the limits of practical use, clove, cinnamon, dill, and garlic flavor oils were shown by Anderson *et al.* (1953) to have little effect on the spores of *Bacillus thermoacidurens* suspended in tomato juice. However, they found cinnamon and garlic oils to be effective in lowering the thermal resistance of a pickle spoilage yeast, the tests being carried out in acidified brine.

Bose and Roy (1959) found that mustard oils are effective in reducing heat resistance of *Bacillus subtillis* spores.

IV. ANTIMICROBIAL ACTIVITY

A. Antimicrobial Activity of Spices.

Several spices, particularly garlic, ajowan, black pepper, cinnamon, nutmeg, cloves, ginger, cumin, caraway, and asafetida, are being used extensively in the Indian dietary and in the Indian system of medicine for correcting a variety of

intestinal disorders (Krishnamurthy and Sreenivasamurthy, 1956). Garlic in particular has antibacterial properties and is widely used both in intestinal disorders and for a number of infectious diseases (Indian Pharmaceutical Codex, 1953). The antibacterial activity of garlic is due to the presence of allicin (Cavallito and Bailey, 1944), which in *in vitro* tests has been found to inhibit the growth of a number of microorganisms (Cavallito and Bailey, 1944; Raghunandan Rao *et al.*, 1946).

Cavallito and Bailey (1944) studied the antimicrobial properties of garlic and found that allicin, its active principle, was effective against both the gram-positive and the gram-negative organisms; it shows an activity equivalent to about 15 Oxford penicillin units per milligram, which is about 1% of the activity of penicillin. However, allicin is equally effective against the gram-negative organisms, which are practically unaffected by penicillin. They further observed that the antibacterial activity of allicin was unaffected by the presence of *p*-aminobenzoic acid. The LD_{50} for allicin in aqueous solutions was of the order of 60 mg/kg given intravenously and 120 mg/kg by subcutaneous administration.

The volatile oils found in various condiments and spices have been reported to be antimicrobial and to possess bacteriostatic properties. The resistance of microorganisms varies with the microorganism and the spice. The decreasing order of the germicidal activity of the spice components is allyl isothiocyanate (mustard, horseradish) and carvacrol (caraway seeds) (equal), cinnamic aldehyde and cinnamyl acetate (equal), eugenol (cinnamon, cloves), eugenol methyl ether (cinnamon, cloves), and eucalyptol (Anonymous, 1948).

Dold and Knapp (1948) tested the actions of twenty-seven different spices (by fifteen experimental methods) against *Escherichia coli, Salmonella typhosa, Shigella paradysenteriae, Staphylococcus* species, *Proteus vulgaris, Bacillus subtilis, Serratia marcescens,* and *Vibrio comma.* Only garlic was completely inhibitory to all but *B. subtilis.* Radish, horseradish, mustard, and marjoram inhibited only some of the organisms. The most active spices were from the Liliaceae family, followed by Myrtaceae, Cruciferae, and Labiatae. The antibacterial action was found to result from the action of the volatile oils (also containing sulfur compounds). A secondary antibacterial action from tannin and alkaloids was also noted. Some data have been presented on the stability of the action of oils of onion and garlic.

Ajowan contains thymol, which has been reported by Bose *et al.* (1949, 1950) to possess antiseptic properties.

Wright *et al.* (1954) observed that at higher levels several spices, particularly cinnamon, retarded gas evolution in simple yeast–sugar systems. The gas-promoting effect of spices was counteracted by mustard flour, which strongly inhibited yeast growth. The essential oil of caraway, cardamom, ginger, and nutmeg resulted in little or no accelerating activity on yeast fermentation, while essential oil of cinnamon retarded gas production. The gas-promoting factor was still present in spices after ether extraction, autoclaving, ethylene oxide treatment,

and prolonged boiling. It was not in ash or in charcoal-clarified filtrates from the boiled spices. The unidentified factor is relatively stable to heat and is extracted by ether. Litchfield and Wilcoxon (1949) reported an antibacterial spectrum of calamus.

Bhat *et al.* (1954) reported that asafetida had no bacteriostatic effect on some of the common intestinal microorganisms and that it altered the proportion of hydrogen and carbon dioxide formed by the gas formers in the intestines. Sreenivasamurthy and Sastry (1958) reported that asafetida oil, like an antibiotic, inhibits the growth of organisms, and, under the influence of the asafetida oil, *Streptococcus faecalis* assumed giant sizes, possibly owing to an interference with cell division and not growth. Both coliforms and anaerobes were much lower in number in the ceca of albino rats fed with asafetida oil than in those in the control group.

According to Boyle (1955), although there is considerable variation in the reported results, there is no doubt that spices and essential oils do possess antibacterial properties.

Capek (1956) found anethole to be responsible for the antimicrobial activity of anise and fennel volatile oils, as determined by tests with *Aspergillus niger*. Russian anise oil contained 87% anethole, and Czech fennel oil contained 61%.

Satoh (1952) reported an increase in the intestinal synthesis of thiamine on administration of garlic in human subjects. Hides Terado (1952) studied the *in vitro* synthesis of thiamine from garlic oil by *E. coli*. Watanabe (1953) and Yurugi (1954) carried out investigations on thiamine derivatives and found that allithiamine was formed by the reaction of thiamine and allicin. It was also reported that allithiamine was absorbed at a faster rate than thiamine in the intestines. Toxicity tests on mice indicated that the LD_{50} for allicin in aqueous solution was of the order of 60 mg/kg intravenously and 120 mg/kg by subcutaneous administration.

Subrahmanyan *et al.* (1957a,b) observed that, among the thirteen important spices studied (cardamom, ginger, mustard, chilies, cinnamon, cloves, aniseed, ajowan, cumin, onion, garlic, asafetida, and black pepper), garlic was the most potent spice, exhibiting high antibacterial action on *E. coli, Aerobacter aerogenes, Staphylococcus aureus,* and *Shigella sonnei* (Table X). The concentrations of garlic required to inhibit *Lactobacillus casei* and *Streptococcus faecalis* are much higher than those required to inhibit *E. coli, S. sonnei,* and *S. aureus*. Whereas the growth of *E. coli, S. sonnei,* and *S. aureus* is markedly inhibited at the 10-mg level, the growth of *S. faecalis* and *L. casei* is inhibited only at the 20-mg level. At the 5-mg level, only *S. aureus* and *S. sonnei* are inhibited (Tables X and XI). Subrahmanyan *et al.* (1958) also found that garlic reduced considerably the number of coliforms, anaerobes, and total count in the ceca of rats fed with either a stock diet or a poor South Indian diet containing a high percentage of redgram dhal. The results indicate that incorporation of garlic

TABLE X EFFECT OF SPICES ON THE GROWTH OF VARIOUS BACTERIA[a]

Spice	Antibacterial action[b] against:					
	E. coli	*A. aerogenes*	*L. casei*	*S. faecalis*	*S. aureus*	*S. sonnei*
Cardamom	+	+	+	+	+	+
Ginger	+	+	+	+	+	+
Mustard	+	+	+	+	+	+
Dried red chili	+	+	+	+	+	+
Cinnamon	+	+	+	+	+	+
Clove	+	+	+	+	±	±
Aniseed	+	+	++	+	+	+
Ajowan	+	+	++	+	+	+
Cumin	+	+	+	+	+	+
Onion	+	+	+	+	+	+
Garlic	−	−	+	+	−	−
Asafetida	+	+	+	+	±	±
Black pepper	+	+	+	+	+	+

[a] From Subrahmanyan *et al.* (1957b).

[b] +, growth; −, inhibition; ±, slight inhibition; ++, stimulation.

in the diet at moderate levels is likely to shift the balance of the microflora in the intestines in favor of lactic organisms, which generally have a positive effect on the absorption of minerals present in the diet.

Sreenivasamurthy *et al.* (1960) found that garlic extract inhibits the growth of *Torula utilis*. Garlic acts as a fungistatic agent at lower concentrations, whereas at higher concentrations, it acts as a fungicidal agent.

Mano Daiji (1962) observed that the soluble portion of the EtOH extract of garlic had an inhibitory action on bacterial growth, but the insoluble portion did not. No synergistic action of an alcoholic extract of garlic was seen with any of the three antibiotics—penicillin, streptomycin, and trichomycin.

Sreenivasamurthy and Krishnamurthy (1959) studied the effect of a water extract of aniseed on the growth of some bacteria and found that it stimulated the growth and acid production of *L. casei*.

Zinchentko (1959) studied the biological activity of onion skin pigments. The onion skins contained up to 2% of glucosides, the aglycons of which consisted of quercetin. Isolated glucosides and quercetin possessed no antibacterial properties against *S. aureus*, *E. coli*, and *B. subtilis*. Onion skins as a food by-product can, however, be utilized in the preparation of P vitamin by aqueous and alcoholic extraction.

Virtanen and Matikkala (1959) observed that fresh homogenized onions had a strong antimicrobial effect, 50 to 100 mg/ml completely inhibiting *S. aureus* growth in nutrient solution; inactivation of the enzymes in the whole onion by

TABLE XI RELATIVE SUSCEPTIBILITIES OF MICROORGANISMS TO GARLIC[a]

Organism	Concentration of garlic (mg/ml)	Number of organisms ($\times 10^4$) per milliter at the end of:			Acidity at the end of 48 hours of incubation (ml 0.05 *N* alkali per 10 ml broth)
		0 hour	6 hours	24 hours	
E. coli	20	17	22	300	2.1
	15	17	20	290	2.5
	10	17	21	375	3.0
	5	17	3,150	14,500	7.0
	0	17	3,600	16,800	7.8
S. faecalis	30	50	55	80	2.5
	20	50	59	100	2.9
	10	50	9,500	18,750	9.0
	5	50	16,000	24,500	10.5
	0	50	27,000	47,000	13.0
L. casei	30	20	250	350	3.0
	20	20	500	650	6.8
	10	20	1,550	34,500	16.8
	5	20	2,700	35,000	18.0
	0	20	5,400	41,000	19.0
S. sonnei	15	23	25	25	1.8
	10	23	27	30	2.0
	5	23	33	36	2.1
	0	23	1,700	5,000	4.2
S. aureus	15	55	92	100	0.8
	10	55	108	110	0.9
	5	55	195	250	1.4
	0	55	4,430	21,600	6.9

[a] From Subrahmanyan *et al.* (1958).

boiling or by freezing with CO_2 gave a very low antimicrobial effect, most of which is due to enzymatic reactions. $OS(Me)CH_2CHNH_2CO_2H$ and $OS(Pr)CH_2CHNH_2CO_2H$ were isolated and determined to 10% accuracy by chromatography of the MeOH extract on Amberlite IR 120 columns.

According to Maruzzella and Freundlich (1959), of the 195 extracts prepared from the seeds of thirty-nine spices, the Et_2O-and Me_2CO-soluble fractions had a greater microbial activity against *E. coli, S. aureus, S. marcescens, Mycobacterium smegmatis, Candida albicans, Erwinia caratovera,* and *Streptomyces venexuelae* than the EtOH-,BuOH-, and H_2O-soluble fractions. Sixteen of the seeds had no antimicrobial activity. Extracts of green cardamom, tonka, angostura, and celery were active against at least six of the seven test organisms as well as against twelve additional microorganisms.

Novak and Fisher (1964) studied the antimycolic activity of a number of fatty

acid derivatives including parsley seed oil acid against twenty-two pathogenic yeasts and molds. A number of these were fair or good against all or most of the yeast and molds, and almost all were fairly active against one of the organisms.

Petkov *et al.* (1969) screened several Bulgarian plants including thirteen spices for their antimicrobial action. They were aniseed, balm, coriander, fennel, garlic, horseradish, juniper, onion, parsley, rosemary, sage, savory, and sweet flag. The water extract of garlic strongly inhibited the growth of almost all the test microbes used in the experiments. The liquid extract of the leaves of sage inhibited the growth of almost all *Shigella dysenteriae* and *E. coli* test microbes. The extracts of horseradish showed a marked inhibitive influence on the growth of the golden staphylococcus and *x*-heniolytic streptococci. The authors believe that their investigation does not permit more concrete conclusions to be drawn about the nature of the substances possessing antimicrobial properties. Undoubtedly, these substances may have quite varied chemical properties. No appreciable importance should be attached to the essential oils contained in these spices, since most of the essential oils are lost during the preparation of their dry extracts.

Gal (1972) reported a simple method for the extraction of the antibiotic capsaicidin from mature capsicum seeds (crushed), followed by stabilization and alkaline treatment of the resulting aleurone suspension, etc. After one purification, 0.5 to 0.7% of active substance was obtained, which is water soluble and active in a dilution of 1:100,000 against *Saccharomyces ellipsoideus* T22.

Bullerman (1974) reported that cinnamon in bread inhibited mold growth as well as aflatoxin production.

B. Antimicrobial Activity of Spice Essential Oils

George and Pandalai (1958) studied the antibacterial activity of some essential oils, including cinnamon and clove oils, against *S. aureus, E. coli, E. typhosa,* and *B. dysenteriae*. It has been suggested that they may prove therapeutically useful in the treatment of the infections of the digestive tract in the form of their stable and nonirritant emulsions.

Bose *et al.* (1949, 1950) measured the bactericidal efficiency of several Indian essential oils by the R.W. test, etc. It was found to be generally high against gram-negative bacteria but very low against gram-positive organisms. The activity against the acid-fast organisms was practically nil. They also studied the relationship between the bactericidal action and the chemical structure of the compounds. The CHO group was found to be more active than the OH group. The presence of an α-B double bond in terpene aldehydes and alcohols was found to enhance the efficiency of the active group.

Sreenivasamurthy and Sastry (1958) studied the effect of asafetida oil on the intestinal microflora of rats fed this condiment for long periods. The results showed that asafetida produces an effect similar to that of antibiotics and that this property rests in the oil portion. Many of the therapeutic properties of asafetida

may be due to this antibiotic effect. However, more work is necessary on its effect on intestinal flora of human beings and on other intestinal pathogens.

Maruzzella *et al.* (1960) screened one hundred and ninety-three aromatic agents consisting of one hundred and fifteen essential oils and seventy-eight organic chemicals *in vitro* against three wood-destroying fungi using the filter paper disk method. Seventy-two percent of the essential oils and 73% of the organic chemicals produced zones of inhibition against at least one of the three test organisms. Prominent zones of inhibition were produced with oil of garlic (imported), origanum (rectified, water white), thyme (red), cassia, and styrax; and by hydratropic aldehyde, isovaleric acid, *n*-caproic acid, cinnamic aldehyde, and eugenol (Table XII). It is suggested that some of the aromatic agents screened might be of value in the wood-preserving industry.

Maruzzella *et al.* (1963) also studied the *in vitro* activity of one hundred and twenty-three essential oils against growing cultures of four phytopathogenic bacteria at concentrations from 1:1000 to 1:10,000. Seventy-eight essential oils produced inhibition of at least one test organism at concentrations up to 1:4000. The remaining forty-five essential oils produced no inhibition at 1:1000. Origanum red (40 to 45% phenols); clove leaf, Madagascar, redistilled; clove stem, Zanzibar; cinnamon, cassia, redistilled; clove, Zanzibar; wintergreen leaf, northern; cinnamon bark, Ceylon; and pimento leaf were found to be highly inhibitory against all test organisms. *Corynebacterium michiganense* was the most sensitive bacterium, and *Pseudomonas glycinea* was the most resistant (Table XIII).

C. Antimicrobial Activity of Volatile Constituents of Spices

Katayama and Nagai (1959a,b) studied the antibacterial activity of volatile components of nutmeg and coriander seed. Nutmeg, which is used as a condiment in manufacturing fish ham and fish sausage, was subjected to steam distillation, and the distillate was extracted with ether. The volatile compounds were separated into fatty acids, phenols, and terpenes.

The terpene fraction was refractionated, each fraction was submitted to chromatostripping, and the presence of α-pipene, α-terpineol, linalool, and geraniol was presumed.

Antibacterial activity of each fraction was tested with *Bacillus subtilis, Salmonella enteritidis, Staphylococcus aureus, Pseudomonas aeruginosa, Proteus morganii,* and *Escherichia coli.* All the fractions inhibited bacterial growth at dilutions lower than 20 to 200 times. Similar observations were made by the same authors about the antibacterial activity of chemical constituents of coriander seed.

Among a number of terpenes examined by Katayama and Nagai (1960), carvacrol, thymol, isoborneol, vanillin, and salicylaldehyde, in dilutions of 1:2000 or more, had antibacterial activities against *Bacillus subtilis, Escherichia coli,*

TABLE XII INHIBITORY ACTIVITY OF SPICE ESSENTIAL OILS ON SOME WOOD-DESTROYING FUNGI[a]

Essential oil	*L. trabea*	*P. versicolor*	*L. lepideus*	Essential oil	*L. trabea*	*P. versicolor*	*L. lepideus*
Allspice, N.F.	20	8	10	Garlic, imported	25	30	50
Anise, U.S.P.	12	5	18	Ginger	0	0	0
Balsam, Peru	6	0	0	Lovage	3	2	4
Basil, sweet	10	10	0	Marjoram, sweet	11	11	6
Bay, N.F.	15	10	26	Nutmeg, East Indian, U.S.P.	11	7	10
Bay or laurel leaf, distilled	5	0	2	Origanum, rectified, water white	35	8	50
Calamus	6	4	1				
Caraway, N.F.	0	0	0				
Cassia, U.S.P.	31	24	15	Pepper, black	0	0	0
Celery seed	0	8	4	Peppermint, rectified, U.S.P.	24	10	5
Cinnamon bark, Ceylon	15	15	10	Peppermint, natural	14	20	17
Cinnamon leaf, Ceylon	15	0	25	Pimento from berries	18	5	11
Clove, U.S.P.	20	6	15	Pimento leaf	23	5	12
Clove leaf, Madagascar, redistilled	20	10	10	Rosemary, N.F.	0	0	0
				Rosemary, acetylated	2	0	0
Clove leaf, rectified	20	6	15	Sage, clary	10	2	2
Clove stem, Zanzibar, redistilled	20	4	13	Spearmint, N.F.	5	2	5
				Thyme, red, N.F.	40	25	15
Coriander, U.S.P.	20	20	12				
Dill seed	7	0	0				
Dill weed	6	5	4				
Fennel, U.S.P.	18	11	4				

[a] From Maruzzella *et al.* (1960).

TABLE XIII INHIBITORY CONCENTRATIONS OF SPICE ESSENTIAL OILS FOR PHYTOPATHOGENIC BACTERIA[a]

Spice essential oil	*E. carotovora*	*C. michiganense*	*P. glycinea*	*P. striafaciens*
Allspice	1:1000	1:2000	+[b]	1:2000
Angelica root	+	+	+	+
Angelica seed	+	1:1000	+	+
Anise	+	1:2000	+	1:2000
Basil, sweet imported	+	+	+	+
Bay, West Indian	1:1000	1:1000	+	1:1000
Bay or laurel leaf, distilled	+	1:1000	+	+
Calamus, imported	+	1:2000	+	+
Caraway, rectified	+	+	+	+
Cardamom	+	+	+	+
Celery seed, domestic	+	+	+	+
Cinnamon bark, Ceylon	1:1000	1:1000	1:1000	1:1000
Cinnamon leaf, Ceylon	+	1:4000	+	1:1000
Cinnamon, cassia, redistilled	1:4000	1:2000	1:2000	1:2000
Citronella, Ceylon	+	1:2000	+	1:1000
Citronella, Formosan	+	1:1000	+	1:1000
Clove, Zanzibar	1:1000	1:2000	1:1000	1:4000
Clove leaf, Madagascar, redistilled	1:1000	1:4000	1:1000	1:4000
Clove stem, Zanzibar, redistilled	1:1000	1:4000	1:1000	1:4000
Coriander	+	1:1000	+	1:1000
Cumin seed	+	+	+	+
Dill seed	+	1:1000	+	+
Dill weed	+	1:1000	+	+
Fennel, sweet	+	1:4000	+	1:4000
Juniper tar	+	1:4000	+	+
Lovage root, imported	+	1:1000	+	+
Mace, distilled	+	+	+	+
Marjoram, sweet	+	+	+	+
Nutmeg	+	+	+	+
Origanum red, 20–25% phenols	1:1000	+	+	+
Origanum red, 40–45% phenols	1:2000	1:4000	1:1000	1:4000
Origanum white, 20–25% phenols	1:1000	1:2000	+	1:4000
Origanum white, 40–45% phenols	1:1000	1:1000	+	1:1000
Parsley seed, distilled imported	+	1:4000	+	+
Peppermint, American, Mohawk	+	1:1000	+	1:1000
Peppermint, redistilled	+	1:4000	+	1:2000
Peppermint, triple-distilled	+	1:1000	+	1:1000
Pimento leaf	1:1000	1:1000	1:000	1:1000
Rosemary	+	+	+	+
Sage, Dalmatian	+	+	+	+
Sage, clary, imported	+	1:1000	+	+
Sage, Spanish	+	+	+	+
Tarragon	+	+	+	+
Thyme red, 40–45% phenols	1:1000	1:1000	+	1:1000
Thyme white, 20–25% phenols	1:2000	1:2000	+	1:1000
Thyme white, 40–45% phenols	1:4000	1:2000	+	1:4000

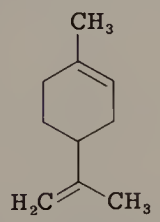
[a] From Maruzzella *et al.* (1963).

[b] Signifies no inhibition at 1:1000.

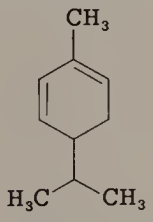
Salmonella enteritidis, Staphylococcus aureus, Proteus morganii, and *Pseudomonas aeroginosa* (Table XIV).

D. Significance of Chemical Structure and Antibacterial Activity of Terpenes

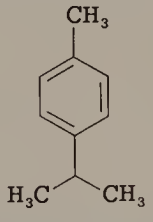
Katayama and Nagai (1960) also studied the chemical significance of the chemical structure and antibacterial activity of terpenes. Compounds that showed inhibitory action on the growth of the test bacteria in dilutions of 2000 or over were eugenol, carvacrol, isoborneol, thymol, vanillin, and salicylaldehyde. As is shown here, there was no difference in antibacterial activity according to the number of double bonds in a cyclohexane ring (Katayama and Nagai, 1960).



1-Methyl-4-isopropenyl-1-cyclohexene (*d*-limonene)

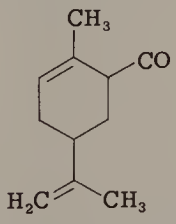


1-Methyl-4-isopropyl-1,5-cyclohexadiene (*α*-phellandrene)



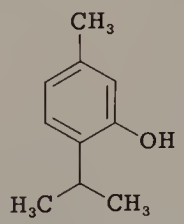
1-Methyl-4-isopropylbenzene (*p*-cymene)

Further, there was no change in the antibacterial activity on introduction of a ketone radical into the foregoing compound.

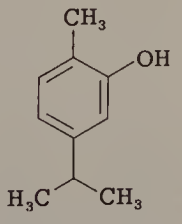


1-Methyl-4-isopropenyl-6-cyclohexene-2-on (carvone)

Introduction of a hydroxyl resulted in increased antibacterial activity to show growth inhibition in dilutions of 2000 or over.



3-Methyl-6-isopropylphenol (thymol)



2-Methyl-5-isopropylphenol (carvacrol)

TABLE XIV ANTIBACTERIAL ACTIVITY OF TERPENES, VOLATILE ALCOHOLS, AND ALDEHYDES[a]

Compound	Bacteria					
	B. subtilis	*E. coli*	*S. enteritidis*	*S. aureus*	*P. morganii*	*P. aeruginosa*
	Dilutions[b]					
Linalyl acetate	20	10	20	20	20	10
Terpenyl acetate	100	—	—	100	10	—
n-Hexyl alcohol	100	100	100	20	100	100
n-Octyl alcohol	200	200	200	200	200	200
Furfuryl alcohol	10	—	10	20	20	100
Benzyl alcohol	20	20	20	20	20	200
α-Terpineol	20	20	20	20	20	20
Citronellol	200	20	100	100	100	20
Geraniol	1000	200	200	200	200	200
Linalool	10	20	20	10	10	20
Eugenol	<2000	<2000	<2000	<2000	<2000	<2000
Thymol	1000	<2000	2000	1000	1000	<2000
o-Cresol	100	100	100	100	1000	1000
m-Cresol	100	100	100	100	100	1000
p-Cresol	100	100	100	20	100	100
Carvacrol	2000	200	1000	2000	<2000	<2000
Isoborneol	2000	—	1000	1000	1000	1000
Vanillin	2000	2000	2000	2000	<2000	200
Isovanillin	100	100	100	100	100	200
Salycylic aldehyde	2000	<2000	<2000	<2000	2000	1000
Furfural	20	20	20	20	20	100
Cinnamic aldehyde	200	20	100	200	100	20
Anisaldehyde	20	100	200	20	1000	1000
Citronellal	100	10	100	100	20	20
Citral	200	100	100	100	100	20
Perillaldehyde	100	20	20	20	20	10
Carvone	100	20	20	20	20	20
Pseudoionone	200	20	20	200	200	1000
Camphene	10	10	10	—	—	20
Fenchon	20	10	20	20	20	20
Benzaldehyde	20	100	100	20	100	100
Acetaldehyde	20	20	20	20	20	20
Safrol	100	—	100	100	100	100
Isosafrol	200	10	1000	200	200	100
1,8-Cineol	—	20	10	10	10	10

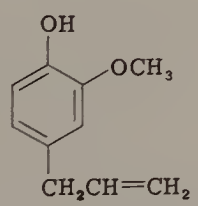
(*continued*)

TABLE XIV (*Continued*)

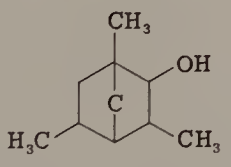
Compound	Bacteria					
	B. subtilis	*E. coli*	*S. enteritidis*	*S. aureus*	*P. morganii*	*P. aeruginosa*
Ascaridol	10	10	10	10	10	20
α-Pinene	10	10	10	10	10	10
β-Pinene	100	20	20	20	100	100
Terpinolene	20	20	100	20	20	200
d-Limonene	200	10	10	20	10	200
α-Phellandrene	10	20	20	10	10	10
p-Cymene	—	—	—	—	—	10
Lemon oil	100	—	10	10	10	—

[a] From Katayama and Nagai (1960).
[b] Maximum dilution at which bacterial growth was inhibited.

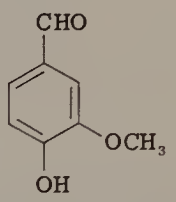
Eugenol, isoborneol, vanillin, and salicylaldehyde introduced with hydroxyl into the ring also showed antibacterial activity in dilutions of 2000 or over. It is interesting that all these compounds, possessing a hydroxyl directly bonded to a cyclohexane or benzene ring, show antibacterial activity in dilutions of over 2000. It is presumed that addition of the condiments containing such terpenes to fish ham and fish sausage will have an antiseptic and antifungal effect.



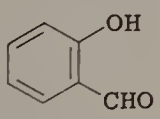
1-Hydroxy-2-methoxy-4-allylbenene (eugenol)



2-Hydroxycamphene (isoborneol)



4-Hydroxy-3-methoxy-benzaldehyde (vanillin)



O-Hydroxy-benzaldehyde (salicylaldehyde)

Chakraborty *et al.* (1961) studied the antifungal and antibacterial activity of the following coumarins by the agar cup method: umbelliferon, ayapin, herniarin, murrayan, marmin, psoralene, isopsoralene, xanthetoxin, and marmesin. They also discussed the relation between structure and antibacterial activity.

Stanley (1963) has made a systematic review of the recent developments in the

chemistry of coumarins in plants, including Umbelliferae, and their physiological activity, etc.

V. PHYSIOLOGICAL AND MEDICINAL ASPECTS

Spices are food components that affect many functional processes, as demonstrated by Glatzel (1968a,b) and other workers. Spices intensify salivary flow and the secretion of amylasis, neuraminic acid, and hexosamines. They aid in cleansing of the oral cavity from food adhesion and bacteria, and they may help to check infections and caries and to protect the mucous membrane against thermic, mechanical, and chemical irritation. The secretion of saliva rich in amylase facilitates the digestion of starch in the stomach, and in this way it makes meals that are rich in carbohydrates more digestible. Spices possibly activate the adrenocortical function and thus help to strengthen resistance and increase physical and psychical capacity. Stroke volume, blood pressure, and stroke frequency can be diminished or augmented by means of spices taken in a specific way. The significance of these possibilities is evident for the sick individual whose heart should be spared stress and for the athlete whose heart will be capable of higher performance. Spices inhibit thrombus formation and accelerate thrombolysis. All these important physiological and medicinal aspects of spices and condiments have been well discussed by Glatzel (1968b) and are only briefly reviewed here.

A. Stimulation of Salivary Flow and Amylasis

Salivary quantity and amylase activity are interesting, especially with regard to their digestive and physiological aspects. Neuraminic acid and hexosamines represent the measure of viscosity that can be determined only approximately by physical means. The content of neuraminic acid is important in protecting against infections (Blumberger and Glatzel, 1965; Glatzel, 1968a,b).

After each meal, spiced or unspiced, basically the same process takes place: Salivary quantity and contained substances increase immediately after the beginning of eating; during food intake they reach their maximum; and immediately after the end of ingestion they decrease rapidly (Fig. 1). In many cases, the resting values have already been reached 30 seconds after the end of the meal. Evidently, the height and duration of the increase are independent of the level of the resting value.

Addition of spices induces a higher increase of salivary quantity and contained substances than is found with unspiced basic food.

It is possible to arrange spices and condiments in the following groups according to their intensity. First there are citric acid, acetic acid, and tartaric acid. These acid flavoring agents are followed by the sharp and biting (burning)

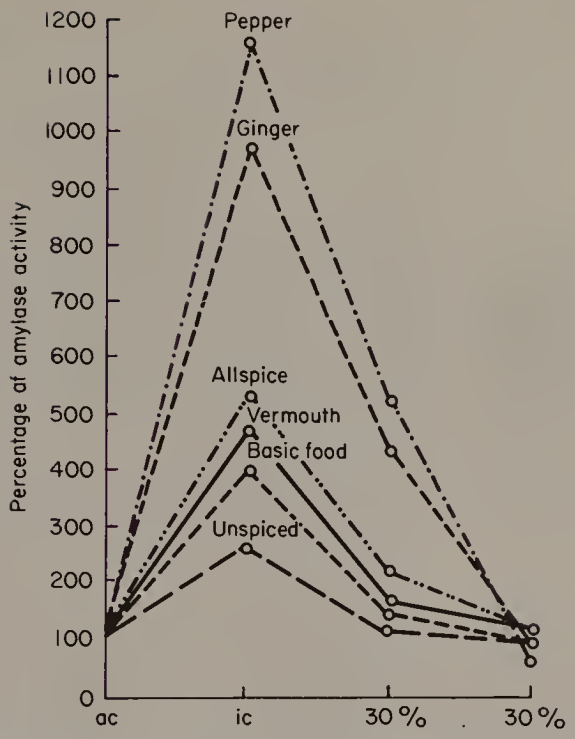


FIG. 1. Amylase activity (a.c.) per milliliter after intake of unspiced and spiced test meals. From Blumberger and Glatzel (1963).

spices—chili, curry powder, ginger, capsicum, pepper, and mustard. Sugar is within the same order. The blunt and aromatic allspice (Jamaica pepper) is apart from the others. In the last place are the bitter condiments—aloes, gentian, hops, bigarade *(Citrus amara),* rhubarb, and vermouth.

These groups may be characterized in the following way:

1. *Acid flavoring agents:* Large salivary quantities. High amylase activity, concentration in hexosamines unchanged compared to the resting values.
2. *Chili group:* Smaller salivary quantities than in the acid group. Exception: great quantities after chili! Amylase activity smaller than in the acid group. Exception: high amylase activity after curry powder and mustard. Concentration in hexosamines little changed compared to the resting values.
3. *Allspice:* Salivary quantity, amylase activity, and hexosamine concentration smaller than in the chili group.

Glatzel (1968a) observed that, in the testing state, the salivary values of different test subjects ranged in different orders (Table XV). The individual differences in the intensity of response to the same spice stimulus ranged in similarly different orders. One individual always reacted strongly, another much less

TABLE XV MEAN RESTING-STATE VALUES OF THE SALIVARY SECRETION OF TEN TEST SUBJECTS[a]
($X + S$ from 4 to 20 single values)

Test subject	Salivary secretion (ml/min)	Amylase (mg glucose/ml)	Neuraminic acid (μg acetyl-neuraminic acid/ml)	Hexosamine (μg glucosamin-hydrochloride/ml)
1	0.98 ± 0.17	1640 ± 624	33 ± 5	125 ± 19
2	0.35 ± 0.11	2177 ± 431	65 ± 12	232 ± 66
3	0.24 ± 0.14	957 ± 277	86 ± 12	207 ± 57
4	0.38 ± 0.09	1569 ± 449	61 ± 10	191 ± 21
5	0.52 ± 0.03	2434 ± 808	48 ± 12	138 ± 19
6	0.49 ± 0.27	408 ± 81	47 ± 16	168 ± 48
7	0.39 ± 0.06	1024 ± 438	126 ± 7	257 ± 28
8	0.52 ± 0.12	1327 ± 229	40 ± 9	144 ± 39
9	0.77 ± 0.12	504 ± 112	70 ± 10	188 ± 27
10	0.47 ± 0.12	2269 ± 481	60 ± 8	208 ± 35
	0.51 ± 0.14 (CV 27%)[b]	1431 ± 376 (CV 26%)	64 ± 11 (CV 18%)	185 ± 34 (CV 19%)

[a] From Glatzel (1968a).
[b] CV, coefficient of variability.

so. Also, the position of a spice within its group is not always the same for all persons. One individual may respond more strongly to pepper than to chili; another may react more strongly after chili than after pepper. But, as a rule, the homogeneity of the groups is not disturbed.

The effect of spices on saliva is closely associated with their immediate contact with the sensory organs of the mucous membrane of the mouth, pharynx, and nose. This contact can be prevented by dispensing the spices in a capsule soluble in the stomach. In this way, any specific spice effect is lacking, and the spiced meal has no more effect on the salivary gland function than the unspiced food (Fig. 2).

Blumberger and Glatzel (1963) reported on the effects of paprika and chili on the excretion of saliva, etc. Salivary excretion increased 50% after paprika and 200% after chili, as compared with the rate for rice without condiments. Despite the increase in salivary volume, there was also an increase in amylase activity—1.5-fold after paprika, and 4.5-fold after chili. The mucin content (neuramintic acids and hexosamine) after rice alone increased to three-fourths the value for normal saliva; after paprika the decrease was slightly less, but there was a 1- to 5-fold increase after chili. On the basis of increased flow and salivary activity after rice without condiments, the amylase activity increased over the normal

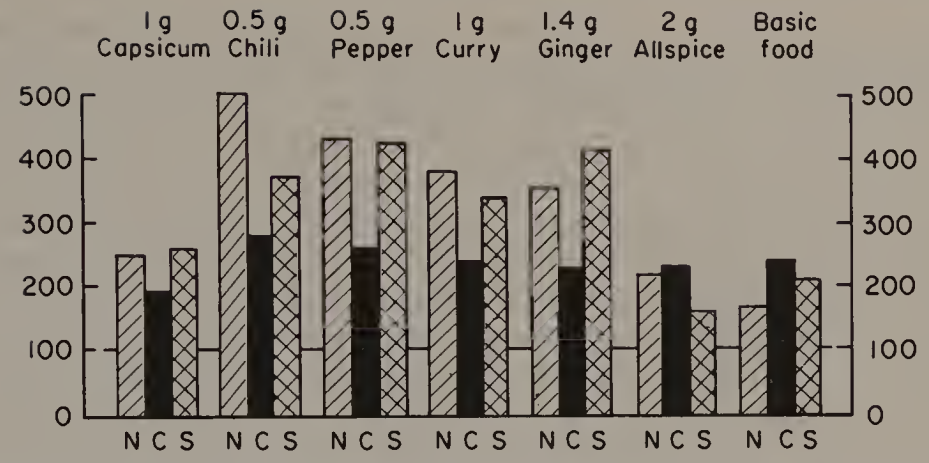


FIG. 2. Amylase activity per milliliter after intake of rice with addition of spice given as normal food intake (N), in capsules (C), and as sham feeding (S). Changes in percentage of the resting state values. From Glatzel (1968a).

resting value 5-fold after paprika, and 24-fold after chili. The stimulatory effect exerted by paparika disappeared quickly after the meal was finished.

Glatzel (1968a) studied both salivary secretion and amylase activity. Various spices were given in diets, and the salivary secretion rate, amylase activity, and contents of neuraminic acid and hexosamine were determined. Citron, curry powder, red pepper, mustard, and sugar (in order of descending activity) increased amylase activity. Neuraminic acid and hexosamine concentrations remained unchanged. Circulation intensity was determined in the fingers and forehead. Red pepper increased the circulation in the hand and decreased the circulation in the forehead within 30 to 45 minutes. Butter, curd, and rice increased heart volume most effectively. Lard had the least effect. Butter also increased the beat frequency. Grape sugars, curds, and meat were also quite effective. Heart stroke volume and stroke frequency decreased after mustard intake. Red pepper decreased the fibrinolytic activity and the number of thrombocytes 20 minutes after intake. In three weeks on a diet rich in red pepper, the excretion of free cortisol in the urine increased 200% with 40 g/week and 300% with 80 g/week. In one case, plasma cortisol levels increased 4-fold and urinary excretion increased only 120%.

B. Stimulation of Gastric Secretion

Physicians previously warned patients suffering from peptic ulcer to avoid spices and highly seasoned foods without having sufficient scientific evidence in support of the effect of different spices on gastric mucosa and the healing time of peptic ulcers. A review of the available literature reveals that the earlier studies (prior to 1955) were somewhat incomplete and sometimes conflicting. For in-

stance, Heupke (1932) showed by actual experiments on healthy human subjects that aniseed oil (10 guttas in 300 cc of saline) stimulated gastric acid secretion, but caraway seed oil (10 guttas), nutmeg (0.5 g), mustard oil (1 gutta), and pepper (0.5g) either had no effect or, in fact, decreased gastric secretion. Basing his conclusions empirically on clinical observations, Heupke (1932) advised the elimination of all spices or spice mixtures in acute gastrointestinal diseases, but permitted the use of cinnamon, bay leaf, vanilla, and nutmeg in the ulcer regime, prohibiting curry powder, clove, garlic, paprika, mustard, pimento, and onion.

Of all the spices studied so far, paprika has probably received the greatest attention. Varga (1938) showed that this spice caused a mild increase in gastric acidity in normal subjects, hyposecretors, and hypersecretors and thus confirmed the earlier observations of Beresey (1934), who, using three types of paprika, found that all three types had the same effect when administered for 30 minutes as an 11 to 30% paprika extract.

By means of tube feedings in human subjects, Frank (1942) demonstrated that coriander, garlic, marjoram, dill, sage, savory, rosemary, celery, thyme, and caraway had no significant effect on the secretion of acid by the stomach (1.0 g in 100 cc of water for 5 minutes). This was confirmed later by Kim (1943) and Harth (1943). Damrau and Ferguson (1949), using x ray and fluoroscopy, found that garlic relaxed the gastrointestinal tract and delayed gastric emptying.

Rabinowitsch (1950) demonstrated in Pavlov dogs that mustard, cinnamon, and cloves were mild gastric secretory stimulants; equivocal results were obtained with paprika, pepper, caraway, and nutmeg. On the other hand, Hollander (1948) observed an increase in gastric secretions with mustard oil but found that it consisted of a serous transudate with no increase in acid.

Sanchez-Palomera (1951) showed that cinnamon, cloves, paprika, and pepper (in 1-g quantities) had no effect on acid secretion and that mustard definitely inhibited gastric acidity in human beings. Mustard, and to a lesser extent paprika, pepper, and cinnamon, caused edema of the mucosa of dogs. Celery salt, cloves, nutmeg, and sage had no effect. The intenstinal mucosa was found to be less resistant to these agents than the gastric mucosa. The author suggested that the defense mechanism of gastric mucosa was superior to that present in other regions of the gastrointestinal tract. A concept of defensive mechanism—the gastric mucosa barrier—is presented and discussed.

With a view to elucidating the continuity of the traditional prohibition of spices in the treatment of peptic ulcer disease, Schneider *et al.* (1956) reported systematic studies on the effect of spice ingestion on (1) the clinical symptomatology and rate of ulcer healing, (2) the clinical symptomatology of inactive peptic ulcer patients not on a routine ulcer regimen, (3) the gastroscopic appearance of the stomach, and (4) the gastric secretion of pepsin. Their main conclusions were as follows: (1) Cinnamon, allspice, mace, thyme, sage, paprika, and caraway seed

administered to peptic ulcer patients, under treatment with an ulcer diet, interval feedings, antacids, and antispasmodics, did not appear to alter the healing time of the ulcer crater when given in relatively large amounts with food three times daily over a period of two weeks to five months. (2) No untoward symptoms such as heartburn, indigestion, belching, or pain were observed if these spices were ingested with food. (3) Symptoms such as heartburn, aftertaste, belching, or epigastric distress often occurred if the spices were taken on an empty stomach. This applied to all spices studied. (4) Black pepper and cloves produced epigastric symptoms in patients with inactive duodenal ulcers who ingested the spices with meals, but who were not maintained on the usual ulcer regimen. (5) Black pepper, chili pepper, cloves, mustard seed, and probably nutmeg may be considered as gastric irritants. (6) Black pepper and chili pepper, which produced symptoms, also induced the most marked hyperemia and edema of the gastric mucosa observed gastroscopically. Thyme and mustard acid produced gastric erythema. (7) The uropepsin excretion was not altered by spice ingestion.

Zaidi and Mukherji (1958) studied the significance of the mucus barrier in the prevention of histamine-induced ulceration. Mucus was produced in guinea pigs by using capsicum as an irritant. The greater the production of mucus, the less was the degree of ulceration. Mucus also protected mucosa and submucosa from inflammatory reaction. The higher the mucin contents, the lower was the free acidity. It has been suggested that mucin neutralized the action of acid gastric juice and also adhered to the surface of the mucosa, forming a barrier against peptic ulceration.

Capsicum in small doses for a short period did not produce any mucosal hemorrhage or ulceration or even depletion of epithelial cells of the gastric mucosa of guinea pigs. The most important gastric response was the production of massive amounts of mucus.

Spices and condiments provoked the production of mucus (Sanchez-Palomera, 1951; Zaidi and Mukherji, 1958). Hollander *et al.* (1947) found that a 1% emulsion of mustard oil in water was a poor stimulus for gastric mucus secretion. Hollander and Lauber (1948) noted that 5% eugenol, a major component of clove oil, was effective. Sonnenblick *et al.* (1950) observed that, after repeated topical application of eugenol in high concentration to the mucosa of the Heidenhain pouch, the mucosa contained desquamated epithelial cells including columnar, parietal, and mucus neck cells in the early stages of application, but after six or seven cycles of stimulation, the stage of mucus barrier normally protecting the underlying epithelium was completely destroyed; however, it rapidly recovered. Lauber and Hollander (1950) showed that the intragastric instillation of eugenol in low concentration (0.2 g/kg) was safe for dogs even when repeated. With these observations in mind, alkali salts of eugenol were prepared so that, when administered into the stomach, they would split and liberate very small amounts of

eugenol, just enough to stimulate mucus secretion and form a mucus barrier without causing desquamation. The liberated alkali would be expected to neutralize the excess free acid.

Zaidi *et al.* (1958) reported on the effect of mucus stimulation on histamine-induced peptic ulceration as follows:

1. Experimental acute peptic ulceration was produced with massive doses of histamine in guinea pigs protected by an antihistaminic, promethazine hydrochloride.
2. Eugenol in small doses (0.5% emulsion) lowered free acidity and peptic activity and gave a significant mucus response, which prevented ulceration.
3. Studies with sodium, barium, and calcium eugenates in the prevention of ulceration have been made. These compounds, when administered in the stomach, liberated eugenol, which stimulated mucus secretion to form a mucus barrier against ulceration, and the alkali neutralized the free acidity. Calcium eugenate appears to be more active in the prevention of ulceration than other compounds used.
4. The significance of mucus stimulants in the prevention of acute peptic ulceration has been discussed.

Mukherji *et al.* (1961) investigated the effect of *Curcuma longa* on the gastric secretion in rabbits. *Curcuma* test meal did not show any change in the volume, free acid, or peptic activity, but appreciably increased the mucin contents of the gastric juice. It has been suggested that the beneficial effect of *Curcuma longa* as a therapeutic agent in gastric disorders may be due to its mucus stimulatory effect.

Analysis of gastric juice of normal rabbits showed high acid values as compared with that of other laboratory animals. This high acidity may serve as a useful measure for testing drugs for their power to neutralize or suppress acidity (Mukherji *et al.* 1961).

Hollander (1948) noted an increase in gastric secretion with mustard oil, but found that it consisted of a serous transudate with no increase in acid.

Using x ray and fluoroscopy, Damrau and Ferguson (1949) found that garlic relaxed the gastrointestinal tract and delayed gastric emptying. Glatzel and Hackenberg (1966) reported an x-ray investigation on the action of capsicum and mustard on the motor and secretary organs and discussed their dietetic importance.

C. Effect on Blood and Circulatory System

1. Effect on Blood Cholesterol

Tempel (1962) studied the effects of two water-soluble fractions of fresh garlic and two synthetic polysulfides resembling those in garlic oil on the percentage of

aortic surface affected by atheromatous changes in rabbits fed cholesterol. The water-soluble fractions were given five times a week in amounts equivalent to 10 g/day for the fresh bulbs; the polysulfides, 0.2 ml/day, five times a week. Only the synthetic polysulfides significantly reduced atheromatous changes in the aorta. Their protective action appears to be independent of the total cholesterol in blood and organs and is limited to the aorta; the increase in free cholesterol was less in all treated rabbits for the first week than for the controls.

Aiyar *et al.* (1964) reported that, in general, the effect of turmeric extract on endogenous levels of cholesterol in the liver and serum of the rat was not appreciable. However, this effect was pronounced in counteracting the increment in liver cholesterol induced by feeding cholesterol, provided the feed ratio of the turmeric derivative to cholesterol was 1:10; this effect was not seen when the ratio was 1:50. The pattern of results was similar with the total alcoholic extract of turmeric or its lipid-free fraction (crude curcumin) with the growing rat or the adult rat, fed with peanut oil or its hydrogenated product. The cholesterol levels reached in the liver of rats fed cholesterol were, however, higher with the unsaturated oil in the diet than with saturated fat. Contrary to previous reports, an inexplicable result repeatedly noted was that cholesterol feeding did not perceptibly enhance the serum levels or the fecal excretion of sterols. The implications of the results and their relevance to human physiology are briefly discussed (Aiyar *et al.*, 1964).

The alcoholic extract of ginger containing the resinous fraction had a direct stimulating effect on the heart (Ally, 1961).

2. Blood Sugar

Karaev *et al.* (1958) observed that the extracts of *Cichorium intybus, Zactuea sativa, Coriandrum sativum, Aloe arborescens,* and *Schizandra chinensis* markedly improved the utilization of glucose in rabbits, as shown by glucose tolerance tests. The possible existence of an active component in these extracts is suggested, and it is recommended that these preparations be tested in human subjects with diabetes mellitus.

Brahamachari and Augusti (1961) compared the orally active hypoglycemic extracts of *Allium cepa* with tolbutamide. Srinivasan (1972) has indicated the beneficial effect of curcumin in the management of diabetes.

3. Blood Clotting

Szirmai (1961) found that extracts of paprika and pepper added to clotting systems *in vitro* shortened prothrombin time, recalcification time, and clotting time with factors V and VI. Thrombin activation was accelerated, and the heparin level lowered. The hypercoagulative effect of these spices appears to be manifested at several steps in the clotting process.

Song *et al.* (1963) isolated a blood anticoagulant (I) from *Allium sativum* and

found it to be toxic to both mice and rabbits. The LD_{50} in mice was 222 mg/kg. Intravenous injection of 70 mg of (I) into rabbits caused death with muscular hyperactivity. (I) has a hypocalcemic effect both *in vitro* and *in vivo*.

4. Effect on Pulse Pressure, Blood Pressure, and Stroke Volume

Pulse pressure, blood pressure, and stroke volume have long been considered to be proved criteria to judge the capacity of the heart. They can be influenced effectively by certain spices. According to Zawahry (1964), the alcoholic extracts (20% in Tyrode's solution) of marc of seeds from *Nigella sativa* (black cumin) showed a lowering of blood pressure in dogs and inhibited the contraction of rabbit intestine.

Glatzel and Rattenmaier (1965) reported that the effect of the addition of spice to food was interesting with regard to the clinical significance of the stroke volume and also with regard to the mechanism of the effect of different foods on stroke volume. Figure 3 shows mean value curves of fifteen analogous experiments on five test subjects. The test subjects ate cooked rice without anything added, cooked rice with an addition of 1.4 g of chili, and cooked rice with an addition of 32 g of mustard. The effect of the two spices was surprising.

In contrast to the response after the unspiced test meals, the stroke volume decreased greatly immediately after the end of food intake. By 10 to 20 minutes after ingestion, it had returned to its initial level. Thereafter, the effects of chili-spiced and mustard-spiced rice meals were quite different. After the chili meal, the stroke volume over a period of time attained higher values than are observed after the unspiced rice meal. After the mustard-spiced meal, the stroke volume was below the level found after the unspiced food, and it hardly changed compared with the resting-state values.

The spice-induced changes in stroke frequency, systolic blood pressure, and pulse pressure were considerably smaller than the stroke volume changes observed after intake of the unspiced food. But, on the whole, their courses were parallel.

Of course, the circulatory activity of other spices may have to be examined. It will be necessary to find out whether spice effects depend on the composition of the basic food. In any case, in principle, it is possible to effect, by means of spices, the required changes—changes that cannot be produced by any of the known medicaments.

5. Prophylaxis and Therapy of Thrombosis

Glatzel (1968a) fed an unspiced and a copiously chili-spiced rice meal and examined the blood before and 20 minutes after intake. Thrombolastogram, factor II, and factor VII showed no food-induced changes. The thrombocytes behaved quite differently. The decrease of 1.4% in the fasting-state value after an unspiced test meal was not statistically significant. After adding chili drug to the basic food, a decrease of 7% in the fasting-state value was found. This is not a

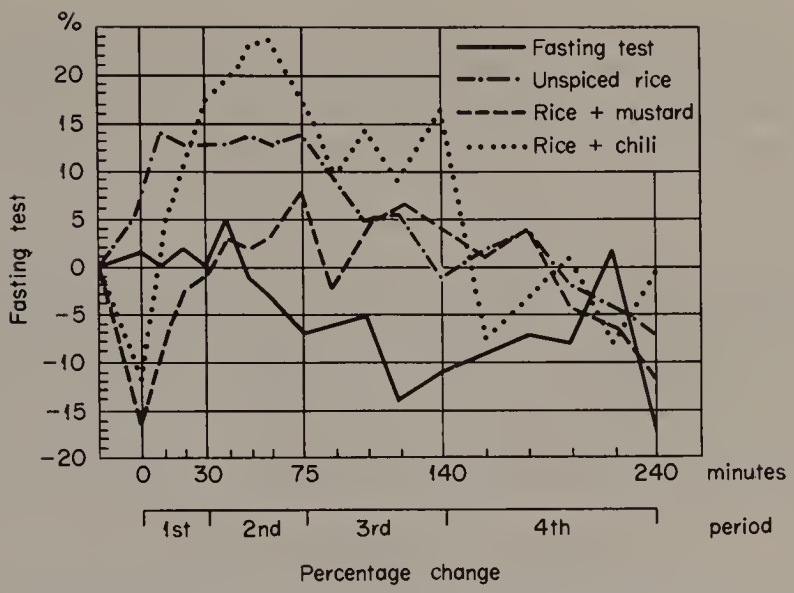


FIG. 3. Percentage changes in the stroke volume after intake of unspiced and spiced test meals, related to the initial value. Mean values of fifteen analogous experiments in five test subjects. From Glatzel and Rettenmaier (1965).

great difference, but the two values are statistically significant. Chili influences the thrombosis process by decreasing the thrombocyte number—that is, by rendering thrombus formation difficult. It favors thrombolysis by checking the postcenal inhibition of the fibrinolytic activity. Evidently, the fibrinolytic activity decreases after each food intake. On the other hand, chili inhibits the clotting disposition, and it does not lose its effect.

D. Spasmolytic and Diuretic Activity

Extracts of seeds of caraway, anise, and sweet fennel administered with the aid of a tube directly into the stomach of rabbits manifested diuretic properties. Similar diuretic effects could be elicited by mixing such extracts with the usual ration of the animals (Skovronskii, 1952).

Kaczmarek *et al.* (1961) reported that methanolic extraction of dry onion scales gave a flavone complex with diuretic properties. Tests on dogs showed increased extraction of NaCl. The total amount of flavone glycosides was 6.74%. They were separated into quercitin (4.81%), spireoside (1.13%), and other heterosides (0.8%).

Saturated solutions of oil of ginger in water showed powerful spasmolytic action, being more potent than zerumbone. Barium-induced spasms were similarly influenced by these drugs (Sirsi, 1962).

Kaczmarek *et al.* (1962) reported that the spasmolytic activity of five components of essential oil of *Petroselinum sativum* was lower than that of papaverine HCl by a factor of 7.0 to 102.6. The activity was governed primarily by the

presence of OCH_2? groups. The effect of OMe groups was small. Only the purified mixture of apiin and luteolin 7-apioglucoside had a slight spasmolytic activity, when tested on dogs. Both apiole and a crude flavone-containing preparation lowered diuretic activity and NaCl excretion.

E. Role in Rheumatism

Sreenivasamurthy *et al.* (1962a) assessed the value of a garlic preparation by following the changes in mucoprotein levels of the blood in rheumatoid arthritis patients. Of the forty-five patients, thirty-two experienced good relief, indicating that the garlic preparation was useful in the treatment of rheumatism.

Bhargava and Haskar (1962b) also reported that the pale-green oil obtained from ajowan seed was useful for external application in cases of rheumatism.

F. Ecbolic or Antifertility Properties

Noding *et al.* (1950) examined four alcoholic extracts of cinnamon with respect to their estrogenic effect. Ovariectomized mice and rats were used as experimental animals. Two extracts gave a positive reaction with the vaginal smear method.

Saha and Kasinathan (1961a) made a critical review of the available standard literature on ancient Indian medicinal plants. All indigenous plants, numbering 277 (including spices), said to be used for or believed to possess emmenagogic and/or abortifacient properties have been tabulated. The spices mentioned are sweet flag, onion, garlic, chives, dill seed, celery, tarragon, caper, cassia, cinnamon, saffron, cumin, turmeric, cardamom, asafetida, fennel seed, juniper, bay or laurel leaves, mint, mace, black cumin, sweet basil, origanum, black pepper, pomegranate, sage, ajowan, and fenugreek. The rediscovery of the modern therapeutic properties of some traditional medicinal plants indicates that there may be some scientific truth in the above statement, but prior to 1960 no scientific work seems to have been reported in support thereof. Gujral *et al.* (1960) and Saha and Kasinathan (1961a) screened only a few of these plants with modern scientific methods with regard to their effect on the isolated uterus of laboratory animals and determined their oxytocic values in quantitative terms against a standard dose of therapeutic oxytocin. The oxytocic value of a given plant serves as an index of its probably ecbolic, including abortifacient and emmenagogic properties. Thus, Saha and Kasinathan (1961b) have shown that onion bulb has very few oxytocic properties (2.9 mg of onion bulk = 0.003 I.U. of oxytocin), and garlic has almost no oxytocic properties (a 31- to 50-mg bulb will produce the same amount of uterine contraction as is produced by only 0.003 I.U. of oxytocin). Likewise, dried pomegranate *(Punica granatum-Anardana)* is now reported to reduce the fertility of female rats and guinea pigs, when administered orally.

Frank (1961) reported the ingestion of saffron in excess (5 g) to induce artificial abortion in a woman and described the resultant saffron poisoning in her.

In studies on the estrogenic activity of pomegranate seed oil, Saraf and Nigam (1964) reported that ovariectomized mice injected with 0.4 ml of pomegranate seed oil twice daily for 2 days showed a response, as measured by cornification of vaginal cells, equal to that produced by 0.1 of estradiol, similarly administered. Immature rats injected with 0.5 ml of the oil daily for 10 days and then killed showed accelerated uterine development equal to that induced by a similar course of ten estradiol injections. Ten 250-mg injections of the unsaponifiable oil fraction had decided estrogenic effects on rabbits; injections of the fatty acid fraction had more.

According to Sharaf and Goma (1965), aniseed *(Pimpinella anisum)* oil and *Glycyrrhiza glabra* extracts showed estrogenic activity when tested on the vagina and uterus of mice, rats, and rabbits. They also blocked the effect of testosterone on the seminal vesicles and that of progesterone on the vagina.

Dac (1966), in reporting on an oral contraceptive, found that the title mixture taken in twenty-two daily doses, during abstention from intercourse, prevents conception for at least one year. There are no side effects. The action is to prevent a fertile ovum from adhering to the endometrium. The powder constituents for a course are 4 drams of the active principle *Emberlia sibis,* 4 drams of *Piper longum,* 2 drams of purified asafetida, and 4 drams of purified borax to give twenty-two doses.

Rainova and Penova (1967) investigated the well-known tonic effect of parsley on uterine musculature. Previously, the active principle of parsley was considered to be apiol, the main component of the essential oil of parsley. It was found, however, that the aqueous extracts of parsley leaves containing only small amounts of apiol, as well as the aqueous extracts of parsley leaves and roots from which the apiol was completely removed, possess a noticeable uterine effect. The data obtained warrant further studies to elucidate the nature of the active principle in parsley.

Zolotovich *et al.* (1967) tested nonalal, citronellol, citral, *p*-anisaldehyde, menthone, carbone of enchone, thujone, and camphor at 1, 10, or 100 g/ml for cytotoxic effects on HeLa cells. Citral had the most pronounced cytotoxic effect, whereas that of citronellol was weak. None of the other compounds were active.

Spaziano *et al.* (1968) reported the synthesis of some 9-*p*-(bis-2-chloroethylamino)phenyl-6-substituted purines as potential cytotoxic agents.

G. Anthelmintic Properties

Subrahmanyan (1942) observed that the powdered rhizome of sweet flag possesses insecticidal properties useful against bedbugs, moths, lice, etc. Scarpa (1950) found that the seeds of fenugreek possessed less anthelmintic power than

chenopodium oil against the most common nematodes. Mukherji and Govind (1959) reported that, as a contact poison, the petroleum ether extract of *Acorus calamus* rhizome was less toxic than DDT, against *Musca nebulo*, by a factor of 17. As an ingested poison, the ether extract was toxic to the larvae of *Bombyx mori*. Bhatnagar *et al.* (1974) studied the insecticidal activity of garlic oil. Su (1977) studied that of black pepper against rice weevels.

H. Use in Leprosy

Reports have been presented by Sreenivasamurthy *et al.* (1962b) on five human cases with acute *Lepromatous neuritis* in one or more nerves, who were treated with garlic by oral and inunction routes. There was considerable improvement in all the cases, with subjective relief of pain and subsidence of nerve inflammation. During the treatment with garlic, none of the patients was given any other routine treatment or steroids. Therefore, there was reason to claim that the clinical improvements were due to the effects of garlic. Garlic has been found to be an anti-inflammatory, analgesic, and antiphlogistic drug, and beneficial effects were expected in the painful inflammatory condition of acute neuritis in lepromatous leprosy. No side effect was noted in any of these cases, nor was any intolerance reported. The administration of the drug is easy and suitable for out-patient treatment. According to these authors, it is not clear whether this curative property of garlic is due to its antibiotic principle—allicin—or to allithiamine formed as a result of allicin's having combined with thiamine, or to the thiamine status of the patient, since thiamine is important in carbohydrate metabolism of the central nervous system, and its deficiency causes impairment in the function of the central nervous system. Thus, more intensive work is necessary to understand the mode of action of garlic in the treatment of *Lepromatus neuritis*.

I. Spices as Tonics

The medicine of ancient times consisted of many spices used as "tonics." It is difficult to define physiologically the term tonic. It is understood to be a medicament that "fortifies" the tonus, or the vitality of the organism—its reactivity, its capacity, and its power of resistance. For this reason, modern medicine connected the conception of tonus with the cortex of the suprarenal gland. Since lay medicine attributed to the capsicums special tonicizing capabilities, Glatzel (1968b) examined the relation between capsicums and adrenocortical function. The measure of the function intensity was the content in plasma 17-hydroxycorticoids and their urinary excretion. Total 17-hydroxycorticoids and urinary 17-ketosteroids were not influenced by the addition of capsicum, but the 17-plasma hydroxycorticoids attained under capsicum-spiced food in mean 120,

respectively, 400%, the free urinary 17-hydroxycorticoids in mean 175, respectively, 155% of the unspiced food levels. However, the difference of 120% is statistically not significant.

No definite statement can be made regarding these results. They do not give more than the impression that food with copious additions of capsicums could induce an increase in the plasma 17-hydroxycorticoids as well as an increase in the urinary excretion of free 17-hydroxycorticoids. This could be considered a sign of an activated adrenocortical function. Further studies are needed on this aspect of other spices.

J. Role in Dermatology

Gastineau (1941) discussed dermatitis caused by nail polish, mercury and new mercury compounds, cinnamon, chocolate, Brazil nuts, lacquer, etc.

In external application, garlic juice is used as a rubefacient in skin diseases and also as ear drops (Krishnamurthy and Sreenivasamurthy, 1956).

Aoi (1960) studied the effect of Japanese curry powder on rabbit skin function. There appears to be only a slight inflammatory element in curry powder. Therefore, its use daily in ordinary quantities is not harmful.

Another Japanese worker (Yamamoto, 1960) has shown that the ingestion of ginger induced exacerbation of eruptions in patients with acute inflammatory skin diseases and acceleration of skin functions in healthy persons. It also induced the manifestation of disposition to dermal inflammation.

K. Antitumor Effect of Spice Essential Oils

Fitzpatrick (1954) observed that, of 310 plant extracts tested, 72 inhibited *Mycobacterium tuberculosis* at a dilution of 1:80 or higher. Of the 27 plants tested in herbals as therapeutics, only 9 plant extracts, including those of cloves and garlic, inhibited *M. tuberculosis* at a dilution of 1:80 or higher. Maruzzella and Brenner (1963) tested 50 essential oils against sarcoma 180 in mice to ascertain their antitumor effect. Any oil showing 25% or more inhibition, based on average diameters of tumors, was considered to possess antitumor activity (Table XVI). Sassafras oil produced 32% inhibition of tumor growth. The remaining 49 oils were considered inactive against sarcoma 180 in the test system. Of these 49 essential oils, 5 reduced average tumor diameter 2 to 19%, 3 caused no reduction in average tumor diameter, while the essential oils of caraway, parsley, and peppermint reduced tumor diameter by 24% each. None of the essential oils were found to stimulate tumor growth.

Kimura Yanagi and Yamamoto Kotaro (1964) studied the cytological effect of crude extracts of garlic and other related species on tumors. Of the plants tested, the extract from garlic exerted the most marked antimitotic effect on tumor cells.

TABLE XVI EFFECT OF ESSENTIAL OILS ON TUMOR GROWTH[a]

Essential oil	Dilution of essential oil	Number of animals (S/T)[b]	AWC (T/C) (g)[c]	Inhibition (%)
Allspice, N.F.	1:2	0/5	—	—
	1:4	2/5	—	—
	1:8	14/15	+0.5/3.5	6
Anise, U.S.P.	1:2	2/5	—	—
	1:4	14/15	+0.0/2.0	0
Bay, West Indian, N.F.	1:2	0/5	—	—
	1:4	12/15	−1.0/2.0	13
Caraway	Undiluted	0/5	—	—
	1:2	15/25	−1.0/2.5	24
	1:4	15/15	+3.0/3.5	16
Celery seed, domestic	1:2	19/20	−1.0/3.0	8
Cinnamon bark,	1:2	0/5	—	—
American, N.F.	1:4	0/5	—	—
	1:8	19/20	+1.5/2.5	4
Clove leaf, Madagascar,	1:2	0/5	—	—
redistilled	1:4	1/5	—	—
	1:8	1/5	—	—
	1:16	15/15	+1.0/2.5	17
Dill weed	Undiluted	0/5	—	—
	1:2	15/15	+0.5/3.0	12
Origanum	Undiluted	0/5	—	—
	1:2	1/5	—	—
	1:4	0/5	—	—
	1:8	14/15	+0.5/1.5	6
Origanum, rectified	Undiluted	0/5	—	—
	1:2	0/5	—	—
	1:4	15/15	+1.0/1.5	9
Parsley seed, distilled	1:2	3/5	−1.0/4.0	24
imported	1:2	2/5	—	—
	1:4	15/15	−0.5/2.5	9
Peppermint, U.S.P.	1:2	14/20	−2.0/3.0	24
Peppermint, triple	1:2	1/5	—	—
distilled	1:4	15/15	+0.0/2.0	5
Pimento leaf	1:2	0/5	—	—
	1:4	13/14	−2.0/2.0	9
Rosemary, N.F.	Undiluted	2/5	—	—
	1:2	18/20	−0.5/3.5	17
Sage	Undiluted	0/5	—	—
	1:2	0/5	—	—
	1:4	1/5	—	—
	1:8	1/5	—	—
	1:16	20/20	+3.5/4.5	6
Savory select	1:2	1/5	—	—
	1:4	15/15	+0.0/3.0	6

(*continued*)

TABLE XVI (*Continued*)

Essential oil	Dilution of essential oil	Number of animals (S/T)[b]	AWC (T/C) (g)[c]	Inhibition (%)
Spearmint	1:2	18/20	−0.5/2.0	13
Sweet fennel, U.S.P.	Undiluted	1/5	—	—
	1:2	15/15	−2.0/2.0	8
Thyme, red	1:2	1/5	+1.0/3.0	—
	1:4	15/15	+1.0/3.0	3

[a] From Maruzzella and Brenner (1963).

[b] S/T, survivors over total in number of treated mice, no deaths in control groups.

[c] AWC, average weight change in surviving mice; T/C, treated over controls; plus unless indicated by minus.

Cytologically, the effects of this plant were much like those induced by colchicine, producing blockage of metaphase cells and scattering, as well as abnormal condensation of metaphase chromosomes. Extracts from *Allium victorialis* and *A. cepa* (onions) produced effects similar to those of garlic, although their action was not very strong.

Papanstassiou *et al.* (1967) synthesized a number of enamine mustards of the general formula $ZCH{:}CRN(CH_2CH_2X)_2$ (where Z and R are an electron-withdrawing group, or R may be H, and X is Cl, F, or OSO_2Me) as possible antitumor or carcinolytic agents. Martinez *et al.* (1968) synthesized the fluoro mustards of uracil (R = Cl or a sulfonyloxy group) and tested for antitumor activity. Negative results were obtained with an increase in toxicity, and no antitumor activity occurred with the replacement of Cl in the Et group with F.

L. Pharmacological Aspects

Chang (1940) studied the pharmacological action of cinnamon. Sodium cinnamate produced vasodilation in frogs and profused frog hind legs. White blood cells, especially the eosinophils, increased after subcutaneous injection of sodium cinnamate and cinnamaldehyde.

Leclerc (1948) reported the antispasmodic and other pharmacological properties of fenugreek and *Lotus corniculatus* in relation to their chemical constituents. Subrahmanyan *et al.* (1954) reviewed the important pharmacological and physiological aspects of asafetida. Epshtein *et al.* (1959) studied the biochemical changes in nervous tissue caused by the action of the low-volatility compounds of onion, garlic, and horseradish. In trials with brain tissue, a decrease in the activity of sulfhydrases was found.

The valuable component of ajowan is the thymol contained in the essential oils. Methods for obtaining this material in forms suitable for medicinal applications were investigated by Kasimova (1957). Ally (1961) studied the phar-

macological action of *Zingiber officinale*. Its alcoholic extract containing the resinous fraction stimulated the vasomotor and respiratory centers of anesthetized cats. It also had a direct stimulating effect on the heart.

Tsuno *et al.* (1960) reported that garlic extract, allicin, alliin, cysteine, and methionine had the effect of promoting the action of mouse liver rhodanese. As for the amino acids that do not contain sulfur, no promoting effect was perceived.

Hata and Kozawa (1961) studied the various pharmacognostical aspects of unbelliferous plants, and Truitt *et al.* (1961) studied the pharmacology of myristicin and the psychopharmacology of nutmeg. The pharmacological effects of myristicin were examined in man and in laboratory animals and compared with those of whole nutmeg powder from which myristicin was obtained. In man, myristicin produced a mild cerebral stimulation but did not reproduce the entire activities of nutmeg powder. The effects of nutmeg powder in one subject were vasomotor instability, tachycardia, hypothermia, absence of saliva, contracted pupils, some emotion lability, a feeling of isolation, and instability to carry on intellectual processes.

Hu *et al.* (1964) reported the strong action of *Illicium majus* (star anise) on excitation of the central nervous system in cats, rats, and mice. According to Preininger and Vrublovsky (1965), a patient experienced relatively beneficial effects from ingestion of 3 to 4 teaspoonfuls of poppyseed. The authors reported the occurrence of several alkaloids in poppyseed but in amounts too small to explain their effect on the patient.

Petkov (1966) conducted pharmacological and clinical studies on garlic, as well as a literature survey. The addition of the extract of garlic in cholesterol feeding experiments on rabbits (52 to 82 days) influenced, to a degree, severe forms of experimental percholesteremia: 923.80 to 558.53 mg%. Pathological atheromatous aortic plaque changes were less marked, with the aortic intima smoother in its entire length than in groups not receiving garlic. In acute blood pressure studies in cats, intravenous injections of 0.05 g/kg (corresponding to 0.20 g of garlic) exerted a hypotensive effect (50 mm). In chronic studies in dogs with experimentally induced hypertension, garlic reduced their systolic arterial pressure. In clinical studies on 114 patients having hypertension and atherosclerosis, garlic caused a marked improvement in the systolic (8 to 33 mm) and diastolic (4 to 20 mm) arterial tension. In workers suffering from chronic lead poisoning, garlic had a beneficial prophylactic action; the erythrocyte concentration improved, and the urinary porphyrin content went from (109 ± 6.2)% to (15 ± 2.2)%. In *in vitro* experiments on rabbit intestinal loop tissue, concentrations of 1:1000 to 1:2000 were inhibitory. Garlic had a definite bactericidal effect on pathogenic microorganisms. In growth experiments on piglets, garlic increased body weight approximately 12% higher than that in controls. It is believed that the pharmacological effects of garlic are due not to the readily volatile polysulfides but to stable, glycoside-like sulfur compound.

M. Other Medicinal Aspects

Kohman (1947) reported that the chemical components of onion vapors possess wound-healing qualities.

Mukherji (1954) stated that various spices and aromatics are used in the Indian system of medicine to correct intestinal disorders resulting from faulty diets. Some of these are garlic, black pepper, ginger, ajowan, and asafetida.

Ramprasad and Sirsi (1956) found sodium curcuminate and sodium salt of the pigment curcumin isolated from turmeric to be an active choleratic, inducing nearly 100% increase of bile production in anesthetized dogs, in doses nontoxic to the animal. The essential oil and some of its fractional distillates also induced choleresis, but to a lesser extent than the pigment.

Cowan *et al.* (1967) studied the antithyroid activity of onion volatiles. Allyl monosulfide administered by gastric intubation into rats was well tolerated even at 350 ml, but allyl disulfide, methyl disulfide, and allyl alcohol were toxic at 100, 50, and 30 ml, respectively. Allyl monosulfide had no antithyroid activity. Consumption of *Allium* species, of which the compounds tested were major volatile constituents, may contribute to the prevalence of goiter in endemic areas.

Czernicki and Weiser (1962) studied the effect of pure carotenoids, especially those of paprika, on the color of yolk.

Ananev (1959) observed that H_2O_2 (2 to 3%) had a virucidal action on tick-borne encephalitis virus. The inactivation of the virus by H_2O_2 can be considerably increased by the presence of peroxidase (horseradish extract).

The other medicinal properties of different spices such as cinnamon have been reported and reviewed by Stanley (1929a); cloves by Stanley (1929b) and Molnar (1942); asafetida by Subrahmanyan *et al.* (1954), Subrahmanyan and Srinivasan (1955), and Patwardhan and Sastry (1957); saffron by Zofia (1948) and Sastry *et al.* (1955a); and garlic by Krishnamurthy and Sreenivasamurthy (1956), Pruthi *et al.* (1959a), and Sreenivasamurthy and Krishnamurthy (1959). Standard reference works also cover the various medical and pharmacological properties of important spices: Council of Scientific and Industrial Research, Publication Directorate (1949–1976), Kirtikar and Basu (1933), Dastur (1951), Nadkarni (1954), R. N. Chopra *et al.* (1958), Glatzel (1968b), and national pharmaceutical codices of different countries.

Yoshimura and Arai (1958) in their studies with rats fed on diets containing thiamine or 0 to 0.5 g of leek showed that a gain in weight or thiamine accumulation in the liver occurred only when thiamine was given. Administration of leek did not increase thiamine excretion, indicating a lack of bacterial synthesis of thiamine in the intestine.

The essential oil obtained by steam distillation from the crude oil of *Nigella sativa* (cumin black) was found to be active in protecting guinea pigs against histamine-induced bronchispasm (Mahfouz and El Dakhakhny, 1960). The activ-

ity was in the acidic part of the oil containing a noncarbonyl and a carbonyl fraction. The carbonyl fraction is composed of a single constituent, nigellon ($C_{18}H_{22}O_4$).

According to Perry *et al.* (1965), fifteen exogenous phenolic and indole amines were detected in normal human urine after subjects were maintained on "plant-free" diets for 72 hours and then fed large amounts of several fruits, beverages, and flavoring agents. Synephrine, *p*-hydroxybenzylamine, and 3-methoxy-4-hydroxybenzylamine were found after the ingestion of orange, mustard, and vanilla extracts, respectively.

Prasad *et al.* (1966) reported studies on the anti-inflammatory activity of some indigenous drugs (including allisatin isolated from garlic) in albino rats. Allisatin (200 mg per 100 g of body weight per day) reduced food volume, necrosis of the feet, and tenderness following formalin injection.

Garlic has been claimed to be effective for the treatment of arthritis in the Ayurvedic system of medicine (Kirtikar and Basu, 1933; Nadkarni, 1954; R. N. Chopra *et al.*, 1958). There being no experimental evidence to corroborate this claim, Prasad *et al.* (1966) conducted studies on the anti-inflammatory activity of garlic and other drugs. The pharmaceutical preparation "allisatin" (concentrated preparation of fresh garlic) was suspended in water and administered orally to albino rats once a day in doses of 200 mg per 100 g of body weight per day. Results revealed that allisatin had a slight anti-inflammatory activity against formalin arthritis, but none against granuloma pouch. Allisatin also appeared to suppress the delayed periarticular changes more than the acute inflammatory reactions. It had no action on the adrenal gland. An interesting finding was that none of the rats fed with allisatin developed gastric ulcer or hemorrhage, whereas an incidence of 25% was observed in the controls. This activity needs further elucidation.

Petrusenko (1967) studied the treatment of arsenical periodontitis. The therapeutic effect of topically applied (into the tooth canal) unithiol (2,3-dimercapto-1-propanesulfonate) in ninety patients with arsenical periodontitis was statistically more significant than the effects of camphorated phenol or clove oil, or the effects in controls treated with water. The effect was determined by the mean time (in days) that elapsed between application and disappearance of the main reaction to the percussion of the tooth. The respective times for the four groups were 1.26, 6.28, 6.33, and 5.53 days.

Kharkov scientists have devoted ten years to studying the effect of onion on the human organism. This research has resulted in new onion preparations, such as allilcepum (Kharchenko, 1971). It exerts a favorable effect on the digestive system and improves the work of the heart, especially a weakened one. Even when diluted one to a million, allilcepum dilates the peripheral vessels of the heart. Experimental data on the effect of the agent have been confirmed by tests in Kharkov's clinics. It has proved to be effective in the treatment of hypertonia in the sclerotic state and intestine atonia (Kharchenko, 1971).

After two months of treatment with a dry onion preparation, the cholesterol level in blood was lowered considerably in a group of rabbits in which experimental atherosclerosis had been produced by pharmacologists. Lecithin content increased (Kharchenko, 1971).

Fresh onion juice (extracted from red onions) may be used for treating suppurative, infected wounds; it kills diphtheria and tuberculosis bacilli; and, owing to its volatile fractions, it is helpful in cases of angina, the grippe, and suppurative processes in the lungs. The allilglycerum drug made of onion is effective for treating trichomonas colpitis (Kharchenko, 1971). Thus, onion is beneficial to everybody, including people of advanced age, as an effective agent for preventing atherosclerosis, and to patients with a high blood pressure and violations of the secretory function of intestines. Shankaracharya and Natarajan (1977) have reviewed the role of spices in health.

VI. USE IN PERFUMERY AND COSMETICS

The use of fragrances for ornamental, ritual, and medicinal purposes dates back almost to the beginning of civilization. The fumes from burning aromatic plants were used to drive away the evil spirits associated with diseases. This belief in the hygienic value of aromatic substances continued for many centuries. However, the wisdom or folly of such beliefs could not be substantiated until the latter part of the nineteenth century. It was during this time that the relationship between microorganisms and diseases was established and the first scientific approach was undertaken in the control of microbial growth with odoriferous plant products. Essential oils were among the first aromatic principles to be investigated. They were demonstrated to possess germicidal activity, as will be briefly reviewed below.

Thompson (1927) mentions the extreme antiseptic powers of volatile plant products. Those who handled and compounded perfumes were thought to remain immune to diseases of the respiratory tract, and the inhabitants of the flower-growing districts of France appeared to be less prone to tuberculosis. Numerous similar accounts have been reported by Sagarin (1945), Maruzzella and Henry (1958), and Ellis (1960), attributing the remarkable curative powers to essential oils and aromatic principles.

Not all spice oils are suitable for perfumery. Some of the important spice oils used in different types of perfumes are the oils of cardamom, cumin, celery, chive, juniper, nutmeg, and nutmeg butter (Pomini, 1952; Anonymous, 1969). Ginger oil is used in the preparation of essences for pharmaceutical purposes. The oils of cinnamon, dill seed, fennel seed, and nutmeg and nutmeg butter are used for scenting soaps, dental preparations, hair lotions, etc. The persistent odor of celery seed oil is much valued both as a fixative and as an ingredient of novel perfumes (Anonymous, 1969).

Suzuki-Shinobu Sohonsha Co., Ltd. (1963) have patented a cosmetic aerosol preparation for skin that stimulates increased blood circulation to the skin and provides for the removal of impurities by inducing perspiration. This preparation, which contains scordinine from chives (*Allium schoenoprasum* L.), is obtained by the addition of an oil–water emulsion, followed by the immediate addition of propellents and exclusion of air from the container.

The literature that has accumulated in this area is voluminous and has been reviewed by Cade (1954), who lists more than one hundred references and describes the many methods employed in studying this subject. Maruzzella (1962) has presented a review of the more recent developments in the field, which can be summarized as follows:

1. Essential oils with esters as the main constituents were more active, followed by hydrocarbons, oxides, phenols, aldehydes, ketones, and alcohols.
2. The common skin bacterium *Staphylococcus aureus* was inhibited by 23% of the essential oils and by 10% of the vapors. The pathogenic yeastlike fungus *Candida albicans,* which attacks the mucous membranes and sometimes the skin, was inhibited by more than 70% of the essential oils in contact and in the vaporous state.
3. Since perfumes are associated commonly with the vaporous state, the antimicrobial properties of perfume oil vapors were studied by Maruzella *et al.* (1959a,b), who showed that 87% of the perfume oils were active against bacteria and 97% against fungi. In the vaporous state, 23% of the perfume oils killed bacteria and 45% were lethal to fungi. Thus, perfume oil vapors possess about one-third the germicidal power of the perfume oils themselves. *Staphlococcus aureus* was killed by 85% of the perfume oils and 15% of the vapors, and *Candida albicans* by 97% of the perfume oils and 45% of the vapors.
4. Some perfume oils at 1:1000 have inhibited the growth of *S. aureus* within an hour of contact. As a matter of fact, many of the experiments indicate that at 1:5000 to 1:10,000 and beyond, certain perfume oils and perfumery chemicals will inhibit a variety of dermatophytes (such as *Trichophyton mentagrophytes, Sporotrichum schenkii,* and *Pityrosporum ovale*). Certainly, such concentrations fall well within the limits of their use in scenting such products as soaps, creams, shampoos, lotions, ointments, powders, sprays, and the medicaments. Of the hundreds of products applied to the skin and hair of the human body for cosmetic and hygienic purposes, many are made fragrant in order to increase their ornamental value and to mask unpleasant odors. If the concentrations are at levels at which the aromatic principles could be demonstrated experimentally to be germicidal, then such toilet articles and similar preparations would destroy microbes in the areas applies. Thus, the selection of a fragrance from the germicidal point of view would be a more rational approach to healthful living.

5. The correlation between the chemical structure of aromatic substances and their antimicrobial activity is difficult to establish, since essential oils and perfume oils are extremely heterogeneous in composition. The comparison of activity and structure becomes meaningful when isolated and purified components are tested. Likewise, the mode of action by which microorganisms are killed by aromatic substances is not precisely known, although Maruzzella (1962) has briefly reviewed some possible modes of action. Some clues are given also by the work of Gal'perin and Dunaeva (1954, 1955), who found that some essential oils affect dehydrogenase activity in paramecia and helminths. Manly *et al.* (1959) have also presented evidence to indicate that some perfumery chemicals inhibit salivary glycolysis. Another interesting manner in which aromatic principles might afford protection against microbial invaders has been reviewed (Maruzzella, 1962).

Unfortunately, work on bacterial or fungal enzyme systems is also lacking and needs detailed investigation.

6. Essential oils, perfume oils and perfumery chemicals in contact or in the vaporous state kill fungi more efficiently than they kill bacteria.

7. Essential oils and perfume oils in contact with microorganisms possess greater germicidal powers than they do in their vaporous state.

8. Perfumery chemical vapors possess the following order of decreasing germicidal activity: acids, aldehydes, alcohols, ketones, esters, ethers, miscellaneous vapors, acetals, and lactones.

Maruzzella (1960) also studied the antifungal properties of the vapors of 136 essential oils *in vitro* against growing cultures of five fungi. The data thus obtained showed that, of 136 oils, the vapors of 114 oils possessed some antifungal properties against at least one of the five test organisms. The vapors of the following essential oils showed the greatest activity against the test organisms: cumin, garlic, eucalyptus (single-distilled, redistilled, rectified), tarragon, estragon, sweet birth (southern), spearmint, and caraway (Table XVII). Of the 114 oil vapors tested, 84 vapors produced zones of inhibition against all the test organisms. The vapors of the following essential oils showed the most prominent zones of inhibition: cumin, garlic, eucalyptus (80 to 85%, single-distilled), eucalyptus (70 to 75%, redistilled), eucalyptus (rectified), eucalyptus (80 to 85%, redistilled), tarragon, estragon, sweet birth (southern), spearmint, and caraway (Table XVII). Further inspection of the data reveals that, of the 136 essential oil vapors tested, 96 (70%) were active against *F. oxysporum f. lycopersici,* 99 (72%) against *S. cereviseae,* 101 (74%) against *C. albicans,* 101 (74%) against *R. nigricans,* and 105 (77%) against *Phoma betae.* This indicated that *F. oxysporum f. lycopersici* was the least vulnerable to essential oil vapors, and *Phoma betae* was the most vulnerable. It was further observed that fungi appeared to be

TABLE XVII INHIBITORY ACTIVITY OF ESSENTIAL OIL VAPORS ON FUNGI[a]

Oil vapor	Diameter of zone of inhibition (mm)				
	F. oxysporum f. lycopersici	*C. albicans*	*R. nigricans*	*P. betae*	*S. cereviseae*
Anise, U.S.P.	29	30	25	90	26
Basil, sweet	40	40	90	90	35
Bay, N.F.	39	26	30	50	30
Calamus	0	0	0	15	0
Caraway, N.F.	90	40	90	90	25
Cardamom, N.F.	0	22	30	45	10
Cassia, U.S.P.	35	40	20	16	36
Clove, U.S.P.	37	24	37	45	37
Clove leaf, Madagascar, redistilled	50	35	40	55	20
Clove leaf, rectified	27	23	30	38	25
Clove stem, Zanzibar, redistilled	40	30	44	64	30
Coriander, U.S.P.	40	45	90	90	40
Cumin	90	90	90	90	90
Dill seed	90	32	50	90	30
Fennel, U.S.P.	27	28	26	90	30
Garlic, imported	90	90	90	90	90
Laurel leaf	30	40	90	90	30
Mace	35	40	40	70	28
Marjoram, sweet	20	27	65	37	28
Nutmeg, East Indian, U.S.P.	30	40	60	70	30
Nutmeg, West Indian, U.S.P.	0	28	90	90	40
Pepper, black	0	0	5	0	0
Peppermint, rectified, U.S.P.	19	30	43	37	33
Peppermint, natural	19	40	55	50	35
Rosemary, N.F.	20	28	60	90	28
Rosemary, acetylated	18	23	90	40	25
Sage, clary	0	25	35	40	0
Sage, Dalmatian	30	40	90	90	30
Sage, Spanish	33	35	90	90	30
Savory select	55	40	90	90	25
Tarragon	90	90	90	90	25
Thyme, red, N.F.	40	40	60	50	35
Thyme, white, N.F.	48	40	72	55	40

[a] From Maruzzella (1960).

almost twice as vulnerable to essential oils vapors as did bacteria and that the essential oils and perfume oils in contact with microorganisms possess greater germicidal power than they do in their vaporous state (Maruzzella *et al.*, 1959a,b).

Of the 22 essential oil vapors that had no antifungal activity, the following are the spice essential oils: angelica root, balsam (Peru), celery seed, ginger, juniper (twice-rectified), lovage, and parsley seed, Strangely enough, sandalwood oil and vetiver (Haiti) oil had no antifungal activity (Maruzzela, 1960).

Some evidence has been presented by Maruzzella (1962) to suggest that the use of certain fragrances in toilet articles or medicaments applied externally to the surfaces of the body might afford some degree of protection against microbial invaders.

Pomini (1962) has reviewed different aspects of juniper, including its use in perfumery and cosmetics. Langenau (1968) reported on the correlation of objective and subjective methods as applied to the perfumery and cosmetics industries.

VII. OTHER USES OF SPICES

Murav'ev (1946) reported that "coriander tincture" was similar to "mint tincture" in its gastric effects. Adamanis and Kaczmarek (1955) suggested the use of coriander flakes as cattle feed.

Cipriani (1959) stated that capsicum compounds such as capsanthin ($C_{40}H_{58}O_3$), capsorubin ($C_{40}H_0O_4$), and capsaicin ($C_{18}H_{27}NO_3$) are added to fuels for internal combustion engines.

Pruthi *et al.* (1960a) reported the utilization of ginger and spent (exhausted) ginger (after the recovery of oleoresin) in the preparation of a number of useful products.

The ground kernel of the tamarind seed (after the removal of the seed coat) may be used in the manufacture of paper and cardboard (Anonymous, 1962a). The chief use for tamarind seeds is in the manufacture of sizing powder. It is widely used in sizing jute yarn and some cotton yarn. It is only half as costly as starch.

Rao (1959) and Narayanamurthy and Rao (1961) reported the preparation of a gum or paper adhesive from tamarind kernel powder. Tamarind kernel powder may also be used as a creaming agent for rubber latex, as a soil stabilizer, and as a pectin substitute (Lewis and Neelakantan, 1964).

While reviewing the chemistry, biochemistry, and technology of tamarind, Lewis and Neelakantan (1964) reported an integrated process for the production of pectin tartrates, and ethanol from tamarind pulp and a process for the extraction of tartrates with acidified ethanol and subsequent extraction of pectin. However, factors such as (1) the high cost of raw material, (2) the low price of

imported tartrates, (3) the limited demand for pectin, and (4) the limited demand for fermented products have stood in the way of commercial exploitation of these processes. A process for making tamarind juice concentrate has been developed (Lewis *et al.*, 1970).

Curcumin (turmeric extract) in conjunction with bixin (annatto extract in oil) is used for coloring of margaine. Ratios of curcumin to bixin of 1:33 up to 3:1 give satisfactory coloring to butter, margarine, cheese, and other fatty or oily foods (Todd, 1964). Saxena *et al.* (1964) have developed a turmeric–oxalic acid reagent for the routine analysis of boron in soil and water.

Hooker Chemical Corporation (1967) reported the use of saffron oil as one of the flame retardants for elastomers, to obtain good flame retardancy without the loss of physical properties.

The American Spice Trade Association has developed several recipes for flavorful low-sodium diets for out-patient service, since most of the spices are low in sodium.

Fazli and Hardman (1968) examined fenugreek seed as a commercial source of the steroidal sapogenin (diosgenin).

The medicinal and antibacterial properties of garlic have been extensively studied and reviewed by Granroth (1970). Amonkar and Reeves (1970) have demonstrated the larvicidal properties of garlic oil against at least four species of mosquitoes in *Culex* and *Aedes* genera. Greenstock (1970) has shown that garlic oil also destroys aphids, white cabbage butterfly caterpillars, and Colorado beetle larvae. Amonkar and Banerji (1971) have observed the antagonistic properties of diallyl disulfide and diallyl trisulfides against several pests of economic importance such as the potato tuber moth, the red cotton bug, the red palm weevil, houseflies, and mosquitoes. They have isolated, identified, and even duplicated synthetically the active ingredients within garlic oil that are responsible for killing the wormlike larvae of certain mosquitoes and agricultural pests. The larvicidal principles of garlic have been identified as diallyl disulfide and diallyl trisulfide. Both natural and synthetic samples of these larvicides are fatal at 5 ppm to *Culex pipiens quinquefasiatas* Say. This opens a new vista for the possible widespread production and application of the powerful ingredients of garlic oil or its active principle, whether natural or synthetic, as insecticides and pesticides. Arora and Pandey (1977) have reported the application of essential oils and their isolates for blue mold decay in *Citrus* reticulate, while Bjeldanes and Kim (1978) have studied the sedative activity of celery oil constituents. Bullerman *et al.* (1977) reported on the inhibition of growth and aflatoxin production of cinnamon and clove oils, cinnamic aldehyde, and eugenol. Guenther (1966) reported on the use of angelica and caraway in the liquor industry.

3

Chemical Aspects—Analytical Techniques

I. GENERAL METHODS OF ANALYSIS

In addition to their individual flavoring components (volatile oil, oleoresin, etc.), all spices contain at least some of the usual components common to most plants and tannins. These components are usually determined by the general methods employed for the proximate analysis of plant products, such as moisture, fat, crude fiber, ash, starch, protein, and tannin. The detailed procedures for these as well as other special techniques are given in standard books on methods of analysis, such as *Food Analysis* by Woodman (1941) and books by Joslyn (1970), Jacobs (1958), the American Spice Trade Association (ASTA) (1968), and the Association of Official Analytical Chemists (AOAC) (1975); they are also covered in national standards and specifications by the Indian Standards Institution (1976) and international (ISO) standards for test methods relating to sampling, determination of extraneous matter (refraction), weight per liter, moisture, total ash, water-soluble ash, acid-insoluble ash, calcium, arsenic, lead, crude fiber, nonvolatile ether extract, volatile oil, alcohol-soluble extract, cold-water-soluble extract, starch, etc. Of these, methods for moisture, volatile oil, determination of filth, heavy metals, and pesticide residues have received comparatively greater attention and are reviewed only briefly here.

A. Moisture

On the basis of collaborative studies, Sindall (1920) recommended the distillation method for the determination of water in spices (cloves, allspice, and pep-

per). May (1926) successfully determined moisture in Madagascar cloves by the distillation method, using petroleum white spirit (d = 0.8135 and b.p. 165° to 195°C). Joslyn (1970) has described in his interesting monograph the various physical, chemical, electrical, and other instrumental methods for the determination of moisture in food products, including spices.

Mossel *et al.* (1951) compared the results obtained by the entrainment distillation method (using isooctane), with vacuum desiccation at 75°C over P_2O_5 and 1 mm Hg pressure, and with the same desiccant and vacuum at room temperature. The results, in general, were in agreement, with the added advantage that the distillation method was faster and had greater applicability, since samples rich in essential oils and lipids caused no difficulty.

Josef (1956) determined the moisture and volatile oil in sixteen spices by titrimetric and gravimetric methods.

The American Spice Trade Association (ASTA) (1968) and the Association of Official Analytical Chemists (AOAC) (1975) have adopted the Dean and Stark method of entrainment distillation using toluene. Rauscher and Korn (1961) used the Dean and Stark apparatus for water determination in food products and compared it with an improved apparatus. Relatively rapid and accurate determinations of moisture could be made with the improved apparatus even when very little moisture was present in the product.

Thung (1964) determined moisture in dried vegetables after lyophilization by drying them in evacuated desiccators, at about 0.1 mm Hg over desiccant, at 50°C, 60°C, 70°C, or room temperature. The results at 50°C were considered to be close to the correct moisture content. They were in good agreement with values obtained by titration with Karl Fischer reagent after extraction with $HCONH_2$ and thus suggested the suitability of the Fischer method for the determination of moisture in dired vegetables. Determination of an end point of drying at 70°C under vacuum after lyophilization was not possible, presumably because of thermal decomposition. Studies were conducted with leek, onion, and other vegetables.

A collaborative study was conducted by Rader (1966) to compare a near-infrared spectrophotometric method and a Karl Fischer volumetric method with the official vacuum oven method. Results by the near-infrared method were as accurate as and more rapid and specific than the vacuum oven method. Results by the Fischer method were not as satisfactory. Garlic, green onions, and toasted onions were used in the study.

Palotas *et al.* (1966) reported that drying paprika powder for 4 to 5 hours at 105°C gave incorrect values for moisture content. More suitable values were obtained by the trichloroethylene distillation procedure of Cocking and Middleton. No caramelization took place. Correct values were also obtained by means of the acetyl chloride titration method of Smith and Bryant.

Previously, moisture in spices and condiments was determined mostly by the

Dean and Stark method, which generally takes 2 to 2½hours for each estimation. For rapid quality control, it is desirable to use more modern, simple, quick, and fairly foolproof methods. Pruthi and Lakshmishankar (1968a,b) reported the rapid determination of moisture with a moisture meter, after its necessary standardization and calibration for individual spices. The systematic method for the calibration of the Marconi moisture meter and the construction and use of the calibration chart have been described. In addition, data on the moisture content of 685 samples of Indian chilies of different grades have been presented and discussed. Calibration charts for ten spices—black pepper, cardamom, coriander, chilies, curry powder, fennel seed, ginger, light berries, pinheads, and turmeric—have been constructed, presented, and discussed (Pruthi and Lakshmishankar, 1968a,b).

B. Volatile Oils

Schut (1925) observed that the then official Dutch method for the assay of oil of cloves by steam distillation and weighing of the ether extract of the distillate was cumbersome and unreliable. He therefore suggested two other methods: (1) the colorimetric method based on the red color produced by concentrated H_2SO_4 in a very diluted solution of the ether extract of the drug; and (2) volatilization of the essential oil by heating the ether extract at 100° to 105°C to constant weight. Method 2 is less exact then method 1, but is also applicable to anise seed and cinnamon bark.

Following a discussion on five different procedures for the determination of volatile oil content, Markwell and Walker (1928) suggested a sixth method that is rapid and yields results closely approximating those obtained by the U.S. Pharmacopocia (U.S.P.) method and moreover is sufficiently accurate for cloves.

In 1928 Clevenger introduced an apparatus for the laboratory estimation of the volatile oil content of plants and plant products. The method, which employed the steam distillation of the relatively water-insoluble oil components, was intended to overcome the deficiencies of the so-called "volatile ether-soluble extract" method as a measure of essential oil content (Clevenger, 1935).

Kofler and Herrensschwand (1935) described a simple procedure, according to which the essential oil is driven over and determined volumetrically. The entire operation required only 1 hour. This receiving and measuring apparatus presupposes a distillation apparatus of the type suggested by previous investigators. It requires no more than 60 ml of distillate for exhaustion of the drug. With certain drugs, such as peppermint and sage, the yield of oil is less from the powdered drug than from the whole drug.

Winton and Winton (1945) had described previous attempts to establish an analytical method for the steam distillation of volatile oil from spices, but the inconveniences of these methods and their lack of sufficient accuracy or preci-

sion precluded their acceptance. The Clevenger design was suitable for routine laboratory use and gave results considerably more reliable than those obtained with previous steam distillation procedures: the quantity of oil obtained could be read directly in the trap, and the uncontaminated oil was then available for further study of its chemical and physical properties.

Although still highly empirical as an estimate of the quality of the particular plant material, the Clevenger method as applied to spices was recognized as a distinct improvement over the "volatile ether extract" method, and a series of investigations were undertaken by Clevenger (1928, 1933, 1942) and the ASTA to evaluate the trap design and details of the procedure. The method was adopted as tentative by the ASTA in 1934 and also appeared in the U.S.P. and the National Formulary (N.F.) for a number of years.

A second series of investigations was begun in 1949 by N. A. Carson to modify the tentative method so as to improve the reliability of the results. These studies resulted in several changes, which were included in the modified procedure adopted by the Association of Official Analytical Chemists (1975).

Van Os (1950) suggested that, for the determination of the volatile oil in coriander seeds, greater accuracy may be obtained by preheating the steam used for the steam distillation to 120°C. A correction must be applied, because the fatty acids also distill over. The method appears somewhat tedious and more time consuming. A series of papers by Carson (1950, 1953, 1954, 1955) cover several modifications in the steam distillation method, including (1) replacement of the cold finger by a West condenser, to prevent leakage of vapor; (2) elimination of the practice of using ether to wash oil from the trap; (3) use of ground-glass connections; (4) heating with a Glas-Col heating mantle; and (5) centrifugation of allspice oil to consolidate the lighter- and heavier-than-water fractions.

Hadorn (1954) and Misra and Krishnarao (1961) suggested an oxidation method for the determination of essential oils in spices, and Hadorn *et al.* (1954) discussed the various existing methods—gravimetric, volumetric, and oxidimetric—for the determination of volatile oil in spices. The sources of errors are traced. The proposed new method consists in placing the sample in a closed flush with steam at 105°C in an oven. The volatile oils are absorbed and oxidized by a mixture of known amounts of bichromate + sulfuric acid and titrated.

During 1954–1956 the ASTA began an analytical research program in collaboration with the U. S. Department of Agriculture, at the Eastern Regional Research Laboratory (Lee and Ogg, 1956). One of the initial projects was a study of the determination of volatile oils in spices by Lee and Ogg (1956), who improved Clevenger's steam distillation method by use of a magnetic stirrer, etc., which made the method quicker and more accurate. Steindelowna (1957) suggested an easy and practical method for the estimation of essential oil by dehydration. A specially prepared Cu–CuO mixture is used as catalyst. The estimation is carried

out in tetrachloroethane solution in the Deryng apparatus (estimation of essential oils in Polish Pharmacopoeia III). Results obtained differ by 1.26% from the theoretical value. Interference by geraniol is negligible.

Kerzner and Ershova (1961) have given the details of the improved method for the determination of allyl oil. The results agree well with those of the gravimetric determination. The method is more economical and less time-consuming than the gravimetric one.

Hadorn (1962) conducted a comparative examination of the methods for the determination of ethereal oils in condiments. The standard deviations shown by collaborative analyses of black pepper and clove indicate appreciable differences in the results of four different laboratories. On the whole, the deviations shown by results obtained by the diffusion method of Hadron *et al.* (1954) are greater than those shown by the Flick distillation (AOAC) method. Comparatively low standard deviations for the Hadorn method are, however, reported by a laboratory in which this method has been in use for six years—± 0.08 for black pepper and ± 0.45 for cloves (thirteen samples of each).

Mazur (1964) described a control method for determination of volatile oil in vanilla in which a saturated solution of NaCl and petroleum ether are used.

A Russian worker, Malysheva (1965), has described a method for the determination of essential oil in mustard seeds, which involves fermentation, binding of the free essential oil with NH_3, and $AgNO_3$ titration. To simplify and shorten the process, the proteins are precipitated with $Pb(OAS)_2$ and Na_2SO_4 after fermentation.

Rosebrook and Barney (1968) made a critical study of the variables in the AOAC and ASTA methods for determining volatile isothiocyanates in mustard flour and seed, which led to the development of a new method that is useful for both mustard flour and seed. The proposed method dispenses with the long reaction times and the digestion of the silver sulfide precipitate now required by the AOAC method. When mustard seed is analyzed, the ground seed is boiled in water for 3 minutes to solubilize the sinigrin, and yellow mustard flour is added as a source of enzyme. When mustard flour is analyzed, the boiling procedure is omitted. Results are more reliable by the proposed method than by the AOAC or ASTA method, and analysis can be completed in a single day.

Rosebrook *et al.* (1968b) also examined the variables that affect the determination of steam-volatile oil in cassia by the official ASTA method in a preliminary study. On the basis of the results of this study, an improved method was devised, which was tested in a statistical program involving four types of cassia, two types of traps, two distillation rates, two sample weights, and two distillation times. Improvements in the method allow complete analysis in 3 to 5 hours, with an increase in the yield of steam-volatile oil and improved reliability. Abraham *et al.* (1973) have reported oxidimetric determination of volatile oil in asafetida.

C. Light and Heavy Filth

Eisenberg (1949) reported a method for separating filth from prepared mustard. He also described two semimechanical procedures—sifting and picking, and gravity flotation—for sorting and separating insect-infested, moldy, and decomposed berries from normal ones.

Sica and Nanz (1950) reported the identification of insect fragments in spices and other food materials, utilizing a differential stain. Azure-1 (1%) in methyl alcohol was used in the acetone flotation method for the determination of insect fragments in foods. It stained food particles preferentially, leaving the insect fragments unchanged.

When the usual AOAC method for the determination of filth in ground cinnamon was employed by Schwartzman (1955) to finely ground samples, a gel-like material was formed; this resulted in poor recoveries and made the filter papers difficult to examine. A method is described in which sodium oxalate–alcohol is used to prevent the formation of gel by the pectin-like substances in cinnamon and to cause flocculation and precipitation of the cinnamon particles. In experiments on insect fragments the recovery was 100%, and for rodent hair it was 80%.

The Association of Official Analytical Chemists (1975) covers the detailed method for the determination of light and heavy filth in spices.

Roaf and Brickey (1968) developed a new method for the extraction of extraneous materials from ground cinnamon. Hydrochloric acid is used to disperse gel formations, wet sieving is used to eliminate the dispersed gel, and a percolator replaces a trap flask to separate the extraneous material from cinnamon tissue. Collaborative results were good, and the method is recommended for adoption.

Thrasher and Brickey (1968) developed a rapid extraction method for the separation of light filth from paprika. The paprika was defatted with a surfactant, wet-sieved, and floated with *n*-heptane from 60% alcohol and a Tween 80–Versene mixture. Collaborative results by this method gave 8.7% increased recovery of insect fragments, and 16.1% for rodent hairs. The method is recommended for adoption.

D. Heavy Metals

The Association of Official Analytical Chemists (1975) gives details of dithiazone method for the determination of lead and also methods for the determination of copper, iron, arsenic, etc. Dytkowska (1964) determined the heavy metal content of certain plant materials, including spices. The following average values were reported for lead, copper, zinc, and iron in caraway seeds: 0.01%, 0.0058%, 0.011%, and 0.044%, respectively. Chauhan and Ray (1964) reported a microchemical (paper chromatographic) method for the determination of strontium in some plant materials. Garcilase (1961) reported the microdetermination of cobalt in native Venezuelan foods, including red pepper.

E. Pesticide, Insecticide, and Fungicide Residues

Egan and Laws (1962) presented a review of the developments in analytical methods for the determination of pesticide residues in foods, particularly the rapid instrumental techniques.

According to Starr *et al.* (1963), the analysis of peppermint hay and peppermint oil after treatment of the crops with DDT, aldrin, dieldrin, or Dibrom indicates that each of these pesticides persisted through the processing of the hay. Both field and laboratory distillation results indicate that the amount of residue found in peppermint oil depends, in part, on the severity of the distillation—that is, the amount of steam used. Up to 60 ppm of dieldrin was found in the oil recovered in a small still when an excessive amount of steam was used. Oil recovered with the conventional distillation procedures contained less than 1 ppm of dieldrin. Peppermint grown in aldrin-treated soils contains more dieldren than aldrin. Gas chromatography has been successfully applied to all samples, with sensitivities as low as 0.01 ppm attainable.

Murphy *et al.* (1965) presented a colorimetric procedure for the determination of microquantities of Bidrin in crop samples. Residues as low as 0.20 ppm can be accurately determined from samples as large as 125 g. An average of twelve samples per day can be analyzed by this procedure. A tolerance limit of 5 ppm has been established under the Federal Food Drug and Cosmetic Act for residues of the herbicide in garlic, onion, and mustard (Anonymous, 1965).

Moye (1968) made a comparison of a phosphorimetric procedure for parathion on celery with the Averell–Norris and electron-capture methods. A rapid and sensitive procedure to determine parathion on celery has been described. Shankaracharya *et al.* (1974a) have discussed analytical methodology in pesticide research. Ahmad *et al.* (1976) discussed the effect of oleoresins on residual toxicity of pyrethrin.

F. Solvent Residues

Todd (1960) reported a technique for evaluating residual solvents in spice oleoresins at levels below 50 ppm with an accuracy of ± 12 ppm. Each solvent (for example, methyl chloride, ethylene dichloride, trichloroethylene, hexane, methanol, isopropanol, and acetone) was studied individually and in combination with other solvents. Labruyere *et al.* (1968) described a gas chromatographic headspace technique for the determination of solvents in spice oleoresins in the region of the official United States tolerance limits. Acetone was found to be a natural constituent of black pepper.

G. Flavor (Aroma and Taste) Evaluation

Sensory evaluation has emerged as an important tool in assessing the quality or flavor of foods. Advances in sensory physiology and evaluation methodology

have broadened and deepened our knowledge and experience in this important segment of research in food science and technology. No piece of laboratory equipment can match the ability of the human senses of taste, smell, touch, and sight to gauge subtle differences in food flavor, texture, and appearance. Whatever we do to improve the quality of a food product, or modify processing conditions, warehousing, transportation, and retailer handling practices, we must ultimately rely on human judgment by either expert tasters or trained taste panels for analysis. Where objective chemical or physical tests have been developed and used successfully, one can be sure that they were first calibrated against human judgments. Unfortunately, a major defect still remains in the precise measurement of sensory responses. The majority of the methods in use for measuring intensity are arbitrary and qualitative. Descriptive terms in common use permit only subjective interpretation (Kramer, 1959). As a result, experimental designs for measuring responses to intensity are very limited (Sinsheimer, 1959) and, generally speaking, have been used only in establishing threshold values (Harrison and Elder, 1950).

The flavor of foods is due to a combination of odor, basic taste, and usually one or more of the following: taste sensation, texture, temperature, and psychological factors (appearance, color, etc.). Among these flavor contributors, taste sensation, basic taste, and odor depend greatly on the nature and concentration of chemicals present in the food under study as well as their structural variations. This terminology is used to differentiate between a *true* or *basic taste*—that is, sweet, bitter, sour, or salty—and a *specific sensation* related to taste—that is, a cooling, burning, pungent, or biting effect, as in different spices. Chemicals exerting a taste sensation are the components that add to flavor of the complex of chemicals irritating the chemical sense receptors as described by Moncrieff (1951). An example of a chemical having a taste sensation is (−)-menthol, which is an essential part of all mint flavors and a contributing factor to this complex of taste sensation, odor, and taste. Other important examples are piperine in black and white pepper, capsaicin and dihydrocapsaicin in chilies, zingerone, shagaol, and gingerol in ginger, eugenol in cloves, and isoeugenol in nutmeg and various essential oils

Spices can be divided into the following five categories, according to the type of flavor and color they impart to foods (Clark, 1970):

1. *Hot and pungent:* cayenne pepper, ginger, various types of paprika, and, most important of all, pepper.
2. *Aromatic:* bay, cinnamon, clove, mace, nutmeg, and pimento (allspice).
3. *Pungent:* garlic, horseradish, leek, onion.
4. *Herbaceous:* anise, basil, caraway, cubeb, cumin, dill, laurel leaf, marjoram, rosemary, sage, terragon, thyme.

5. Those spices that are used mainly *for coloring,* although some of these would certainly impart their own particular flavor—for example, paprika, saffron, and turmeric.

Of the five senses, odor perception is perhaps the most subtle, and certainly the least understood. In reviewing the literature, one is impressed by the significant fact that in reports on organoleptic evaluations and measurements of odor an important factor is absent: a precise knowledge and understanding of the mechanism of odor perception. Most books (Crocker, 1945; Moncrieff, 1951; Caul, 1957; Little, 1958; Kuehner, 1964; Amerine *et al.,* 1965; Gould, 1966), the review by Hornstein and Teranishi (1967), and the Proceedings of the International Symposium on Sensory Evaluation of Foods organized by the Swedish Institute for Food Preservation Research (1968) are filled with philosophical discussions, hypotheses, and theories, trying to explain this complex function. Unfortunately, we do not have adequate data from which to draw meaningful conclusions.

The available information on sensory evaluation of spices is thus rather scanty, but will be briefly reviewed here.

Flavor evaluation is done by both subjective and objective methods.

1. Subjective Methods of Sensory Evaluations

Cartwright and Nanz (1948) and Nanz *et al.* (1950) used a taste panel technique for the evaluation of the flavor of natural spices, including capsicum, caraway, cassia, cloves, coriander, ginger, mace, mustard, nutmeg, paprika, pepper (black and white), and sage, in comparison with spice substitutes in representative test foods including white sauce, oil white sauce, pumpkin, eggnog, and cream sauce, which were found to be useful aids in flavor evaluation. In general, spice substitutes were poorer in odor and taste than were natural spices. Separation of flavor-bearing components of natural spices from their original loci appeared to alter and reduce the acceptability of the spice flavor.

Nanz and Cartwright (1951) reported the comparative evaluation of spices with respect to flavor retention of natural spices and of commercially available spice oils and spice extractives after boiling and baking. The flavoring materials were added to white sauces and foods such as pumpkin and cream sauce, which were boiled before evaluation. The superiority of the natural spices over spice oils and oleoresin was established.

Pruthi *et al.* (1959f) and Singh *et al.* (1959b,c) reported the effect of nitrogen packaging and storage time and temperature on the flavor, color, and texture of garlic powder, and Misra and Pruthi (1962, 1963b) reported the results of both physical and chemical evaluation of the flavor and color of Indian curry powder.

Peryam (1958), Tilgner and Barylko-Pikielnal (1959), Tilgner (1962a), and Mathur *et al.* (1971) found the dilution index method an aid in quantifying odor and flavor responses. The use of the index permits the establishment of anchor points for descriptive terms used in rating odor and flavor of foods and beverages. Precise and comparable data are obtained that reflect within psychophysiological limitations the intensity of sensory qualities in a less arbitrary way than with conventional methods. Tilgner (1962a) described dilution index methodology, and odor dilution index values of nine selected spices, in common use in the Polish meat industry, have been presented (Table XVIII) and discussed. It is interesting to note the extreme range of variation in values (1:4000 to 1:110,000) among the nine spices studied and also the wide variation among samples of the same spice (1:4000 to 1:100,000 for juniper). On the basis of these dilution index values, he also observed that spices underwent a 5 to 30% loss of aroma during three months' storage in ordinary paper containers. Other tests showed that storage in glass bottles resulted in much less loss of aroma.

The dilution flavor profile method of sensory evaluation, introduced a number of years ago (Cairncross and Sjöström, 1950; Caul, 1957), has considerable value but requires great skill, education in odor and flavor sensations, keen interest, and considerable intelligence on the part of the judge. In addition to these requirements, Caul (1960) has added one more factor—*honesty*. She emphasized that descriptive methods of this kind are based on good communication and, above all, on an honest analysis of the sensations perceived.

Tilgner (1962b) later reviewed the "anchored sensory evaluation" tests for foods. Quality scoring of food products by sensory means is generally done by reference to subjective standards derived from the training and experience of the grader. Accuracy in quality scoring could be much greater if objective standards were available to the grader for ready reference in restandardization. In fact, it should be the aim of sensory grading systems to provide objective, easily reproduced, and unvarying reference standards for this purpose. They would certainly improve the accuracy and consistency of graders' scores by permitting convenient restandardization of quality scales.

A number of workers have expressed the need for anchored sensory evaluation tests (Tilgner, 1962b). Such tests have been especially valuable in establishing the threshold of individual quality grades. For example, they can be used to establish the point at which a product is considered "medium," "good," or "excellent" in flavor. In fact, grade limits anchored in terms of physiochemical attributes can be considered a sine qua non of modern quality control. If such anchored tests should be established for sensory qualities, we shall have made great strides in achieving better quality control of foods in general and of spices in particular.

According to Neale (1963), the spices that bring out the best flavor in foods are coriander seeds for pultry, potatoes, and peas; cumin for rice and peas; ginger for poultry and sweet potatoes; and black pepper, turmeric, red chilies, and dehy-

TABLE XVIII DILUTION INDEXES FOR SELECTED SPICES[a]

Product	Odor dilution index
Allspice	1:25,000–1:47,000
Black pepper	1:35,000–1:60,000
Caraway	1:30,000–1:75,000
Cardamom	1:100,000–1:110,000
Coriander	1:40,000–1:80,000
Ginger	1:35,000
Juniper	1:4,000–1:100,000
Nutmeg	1:15,000–1:75,000
Sweet bay	1:75,000

[a] From Tilgner (1962a).

drated onion and garlic for all meats, poultry, and vegetables. Cloves and black cardamom seeds help in blending different flavors.

The quality of the spice mixture is also dependent on the dilution index (Tilgner, 1962a) of the mixture and the individual components when used in vegetable and mutton mince curries. Data in Table XIX show that the dilution of each spice in the threshold dilutions of spice mixtures and curries is much greater than their respective dilution index (Mathur *et al.*, 1971). The dilution index of curried vegetable is reduced to one-eighth for odor and one-fourth for taste, whereas the dilution index of mutton mince curry is reduced to two-sevenths for odor and one-twentieth for taste.

Mathur *et al.* (1971) have further presented data (Table XX) that show threshold dilutions from the dilution index of the ingredients of spice mixtures, curried vegetables, and mutton mince. These are calculated on the basis of the additive effect on threshold values as reported by Guadagni *et al.* (1963b) for various flavors. It is seen that the actual dilution indices are much less than these values. This shows that some of the spices possess the property of enhancing the flavor of other ingredients or have a mutual synergistic effect, as reported by Nawar and Fargerson (1962) and Mathur *et al.* (1971).

Hamann (1966) has described and discussed definitions of flavor, factors affecting food acceptance, basic principles involved in the physiology of flavor perception, threshold concentrations for taste and odor, flavor evaluation, flavoring materials (including spices), progressive use of synthetics, use of mixtures of spice essential oils in flavoring of foods, flavor precursors, flavor potentialities, and flavor regulations. Stone *et al.* (1965) reported techniques for sensory evaluation of food odors.

2. *Objective Methods of Sensory Evaluations*

Langenau (1968) has discussed the correlation of objective and subjective methods as applied to the perfumery and cosmetics industries and has concluded

TABLE XIX DILUTION INDEX AND DILUTIONS OF INGREDIENTS AT THRESHOLD DILUTIONS OF CURRIED VEGETABLES AND MUTTON MINCE[a]

Sample	Dilution index (× 10)		Dilution at threshold dilutions of spice mixture (× 10^4)		Dilution at threshold dilution of curried vegetable (× 10^4)		Dilution at threshold dilution of mutton mince curry (× 10^4)	
	Odor	Taste	Odor	Taste	Odor	Taste	Odor	Taste
Coriander	200	100	8.0	4.0	4.0	2.0	1.04	1.04
Cumin	200	100	12.0	6.0	6.0	3.0	1.56	1.56
Ginger	200	100	8.0	4.0	4.0	2.0	1.04	1.04
Black pepper	500	100	2.0	1.0	1.0	0.5	0.26	0.26
Dehydrated onion	200	5000	61.0	30.5	30.5	15.8	7.90	7.90
Dehydrated garlic	5	100	0.6	0.3	0.3	0.15	0.04	0.04
Turmeric	500	200	1.6	0.8	0.8	0.4	0.10	0.10
Chili	500	200	3.2	1.6	0.8	0.4	0.21	0.21
Clove	200	50	2.0	1.0	1.0	0.5	0.26	0.26
Black cardamom	50	20	2.0	1.0	1.0	0.5	0.26	0.26
Spice mixture	100	50	—	—	50.0	25.0	13.00	13.00
Vegetable curry without spice mixture	8000	2000	—	—	950.0	475.0	—	—
Vegetable curry with spice mixture	1000	500	—	—	—	—	—	—
Mutton mince curry without spice mixture	700	4000	—	—	—	—	187.00	187.00
Mutton mince curry with spice mixture	200	200	—	—	—	—	—	—

[a] From Mathur *et al.* (1971).

by stating that at the present state of our knowledge the correlation of odor with molecular structure is of very limited value. Similar work is needed for spices.

The advantages afforded by the corroborative use of objective analytical methods such as gas–liquid chromatography (GLC) and thin-layer chromatography (TLC), infrared and ultraviolet spectrography, and the classical analytical techniques, together with subjective evaluations of odor and flavor, are illustrated by descriptions of their application to the evaluation of cinnamic aldehyde, the establishment of analytical limits, the differentiation among various synthetic and natural types of geraniol, the influence of optical rotation on the odor of carvone, the determination of the geographical source of essential oils, the sclareol content of clary sage, etc. (Langenaur, 1968).

Although such techniques are very useful, they are nevertheless subject to many limitations. Instances are cited in which the human nose is a far more sensitive detector than any instrument.

With regard to perfume compounds, objective–subjective methods are useful primarily for duplication. The esthetic value of perfumes can be judged only by subjective means such as panel evaluations. Because of the many limitations, pointed out by Langenau (1968), objective–subjective correlations should be attempted only after exhaustive study of the many factors involved in each individual problem.

Schwimmer and Weston (1961) reported the enzymatic development of pyruvic acid in onion as a measure of its pungency.

Schwimmer and Guadagni (1962) observed that a highly significant correlation ($r = -0.97$) exists between the amount of enzymatically developed pyruvic acid present in the juice of comminuted onion and the olfactory threshold concentration of the juice. The correlation indicates that determination of pyruvic acid in freshly prepared onion juice constitutes a fairly reliable, simple, and convenient method of estimating at least one aspect of onion flavor. Later, Schwimmer *et al.* (1964) studied the relation between pyruvate odor strength of eighteen samples of reconstituted dehydrated commercial onion powders. They concluded that the estimation of total pyruvic acid may be of value in assessing at least certain aspects of the flavor quality of dehydrated onions.

Chang (1966) discussed flavor characterization, methods of isolating volatile flavor, and the application of new techniques such as gas–liquid phase chromatography (GLPC), infrared spectrometry, and mass spectrometry.

Gold and Wilson (1963a,b) developed a coloimetric ester test as a measure of

TABLE XX RATIO OF ACTUAL AND CALCULATED THRESHOLD DILUTIONS FOR CURRY POWDER AND CURRIED FOODS[a]

Material	Characteristic	Calculated threshold dilutions	$\frac{\text{Actual value}}{\text{calculated value}} \times 100$
Curry powder	Odor	145	69.0
	Taste	224	22.3
Curried vegetable (on the basis of curry powder)	Odor	1800	62.0
	Taste	676	74.0
Curried vegetable (on the basis of individual spices)	Odor	3333	30.0
	Taste	3597	13.9
Curried mutton mince (on the basis of curry powder)	Odor	727	27.5
	Taste	727	27.5
Mutton mince (on the basis of individual spices)	Odor	2198	9.1
	Taste	2198	9.1

[a] From Mathur *et al.* (1971).

total flavor in celery. Farber (1949, 1957), Pruthi *et al.* (1959d), and Singh *et al.* (1959b) reported the volatile reducing substance method for the estimation of volatile flavor in garlic and onion. However, this technique has some limitations, particularly for samples stored at high temperatures. Sigmund and Guadagni (1968) studied the kinetics of the enzyme development of pyruvic acid and odor in frozen onions treated with *L*-cysteine sulfoxide lyase. Tassan and Russell (1975) presented a paper covering the sensory properties of cumin constituents, how they are affected by heat treatment, and the contribution of four aldehydes to the main impact character of cumin odor. Todd Jr. *et al.* (1977) determined the pungency in capsicums by GLC.

H. Determination of Aflatoxins

A number of methods for the quantitative estimation of aflatoxins have been reported for various agricultural commodities. Suzuki *et al.* (1973) showed the necessity of modifying these methods in order (1) to prevent the occlusion of alfatoxins in fixed and/or volatile oils and (2) to remove naturally occurring interfering substances, especially in black pepper. They also analyzed eighty samples of various spices by their modified method and found aflatoxins in only seven samples: three of black pepper, one of celery seed, and three of nutmeg.

II. SPECIFIC ANALYTICAL TECHNIQUES

A. Instrumental Techniques

During the past few decades, the domain of chemical analysis has expanded considerably and made astounding advances, including many specialized techniques to supplement chemical analysis. This was mostly because of the impact of instrumentation techniques such as microscopy, polarography, colorimetry, ultraviolet, infrared, x ray, and nuclear magnetic resonance spectrography, mass spectrograms, paper and adsorption chromatography, TLC, and GLPC, which are now being used on an increasing scale in the analysis of foods, including spices. The popularity these specialized techniques enjoy stems from a number of features held in high esteem, particularly by the analytical chemist. Only small amounts of material are required for investigation, and often the sample may conveniently be recovered for further study. The methods are relatively simple, and significant information is obtained within a very short time. The experimental data may be traced automatically in the form of a chart giving a permanent record of the analysis. Often the curves so produced are sufficiently rich in detail to permit fine structure determinations and the detection of subtle differences between compounds. They represent, indeed, characteristic molecular fingerprints. Some of the important specialized techniques are briefly discussed below.

1. Microscopy

Esses and Schelton (1956), in a series of six interesting papers, described the procedures for microscopic examination of about thirty-five spices and condiments used in flour confectionery, nicely illustrated by photomicrographs. They have also discussed the botanical and microscopic aspects along with the common adulterants used in each spice powder.

The characteristics of cinnamon, cassia, nutmeg, mace, ginger, and cloves and methods for their preparation for microscopic examination have been outlined with photomicrographic illustrations (Anonymous, 1956).

2. Polarographic Techniques

Hanc and Santavy (1946, 1947) described a simple polarographic method based on the reducibility of piperine in pepper on a mercury drop electrode. Bitter (1950) studied the polarography of some spice essential oils and determined various aldehydes by using 50 to 100 mg% solutions of some essential oils in 96% alcohol suitably diluted and mixed with LiCl or NH_4Cl solutions. Markman and Fazylova (1961) studied the polarographic behavior of vanillin in H_2O, EtOH, and aqueous EtOH solutions. For analysis of aqueous solutions, LiCl and NH_4Cl can be used as supporting electrolytes. For EtOH solutions NH_4Cl and for alcoholic solutions Et_4NI should be used as the supporting electrolyte. Czionkowska *et al.* (1966) carried out polarographic determinations of copper, lead, and zinc in dried mints, rosemary leaves, fruits of coriander and fennel, and roots of pimpinella. The average copper content for the aboveground parts of spices ranged from 0.00104 to 0.00136%; for underground parts the average was 0.00066%. The average lead values were from traces to 0.000477%, with no significant differences being found in the different genera. Great differences were observed for zinc, the average amounts ranging from 0.00292 to 0.01049%.

3. Colorimetric Methods

Nigam and Kumari (1963) have reviewed the application of colorimetric methods for the analysis of essential oils, including spice essential oils. Karawya and Wahba (1967) developed a colorimetric method for the determination of eugenol in essential oils of allspice, bay, clove, etc. The method is based on the measurement of the intensity of blue color produced by the reaction of eugenol with 2,6-dichloroquinone chlorimide in isopropyl alcohol and a phosphate buffer at pH 8.6. The following factors, which may affect the color stability and sensitivity, were studied: pH, time, temperature, concentration, wavelength, and color interference of some compounds naturally present in the essential oils (that is, furfural, cineole, pinene, phellandrene, caryophyllene, and limonene). The following conclusions were reached: (1) Phenolic and nonphenolic constituents accompanying eugenol in natural products did not interfere with the color reaction. (2) Concentrations of eugenol ranging from 20 to 200 μg obeyed Beer's law.

(3) The colorimetric method is feasible for determining eugenol in oils of clove, allspice, and bay, and the results are reproducible with an error of −0.75 to +1.3%. (4) The differences between the results obtained by the colorimetric method and that of Egyptian Pharmacopoeia (5%, 6.6%, and 25.33% for oils of clove, allspice, and bay, respectively) were caused by the presence of other phenolic and nonphenolic constituents, which were alkali-soluble and interfered in the latter but not in the former method. (5) The proposed colorimetric method required as little as 20 to 300 μg of eugenol contained in the oil sample, whereas the Egyptian Pharmacopoeia method required 10 ml of oil.

Kaul and Nigam (1966) applied colorimetric methods to the estimation of limonene in caraway and dill seed oils, of eugenol in cinnamon leaf oil by the production of blue color with a 2% alcoholic $FeCl_3$ solution, of geraniol in oil of citronella by using the same color reaction as for limonene, and of 1,8-cineol in the oil of *Alpinia galanga* by using Sehorn's reagent.

Koparieva and Potapova (1966) reported a photometric method for the determination of essential oil in garlic and onion. Pyruvic acid formed in the enzymatic decomposition of 3-(allylsulfinyl)alanine contained in the essential oil was determined as its 2,4-dinitrophenylhydrazone.

Kapur *et al.* (1957) described a colorimetric method for determination of vanillin, which is based on the fact that thiobarbituric acid reacts readily with vanillin to give a characteristic yellow complex that is stable and partially soluble in water. As little as 0.2 μg of vanillin per milliliter in 92% phosphoric acid (B.P.) gives a color reaction with 0.025 *M* thiobarbituric acid in *M* phosphoric acid. The yellow complex is characterized by a single absorption peak at 434 mμ, irrespective of the concentration of phosphoric acid. The method is applicable to several other aldehydes also.

Chernysheva (1966) described a colorimetric method for the determination of linalool in coriander oil. Tsatsaronic and Kehayoglou (1964) examined green pepper colorimetrically.

Kokoski *et al.* (1958) reported the characteristic fluorescence of each of 133 powdered drugs (including some spices and herbs) with *N* NaOH, *N* NaOH in MeOH, *N* HCl, HNO_3, and H_2SO_4 separately diluted with an equal volume of water, when examined under ultraviolet radiation.

4. Nephelometry

Gurvich and Nepardze (1966) reported the nephelometric determination of the essential oil content of small coriander. Twelve samples contained 0.83 to 1.01% of oil, and in comparison with the Ginsberg method, a quadratic derivation of ±0.008% was found.

5. Spectrophotometric Methods

Betts (1965a) determined spectrophotometrically the carvone content of the developing fruits of *Apium graveolens* and *Carum carvi* over three seasons.

Carvone in the fruits attained a level of 11 to 20 mg per 100 caraway fruits, or 4 to 9 mg per 100 dill fruits after three or four weeks of pollination. Variations in the carvone content of the essential oils distilled from these fruits were probably due to the variable quantities of limonene.

According to Polyakov and Falina (1962), the spectrophotometric determination of citral in oxidized coriander oil was more precise than that by a polarographic or an oxime formation method.

Gaind *et al.* (1965) developed a spectrophotometric method for the determination of alliin in some *Allium* species. Beer's law is obeyed for 5 to 20 μg of alliin per milliliter. White garlic, red garlic, and *Allium* species contain 0.76 μg, 0.57 μg, and 0.07 μg of alliin, respectively; 4 μg of alliin can be detected by this method.

Malla *et al.* (1962) reported the spectrophotometric determination of menthone and menthol in the oil of *Mentha arvensis*. Menthone dinitrophenylhydrazone was prepared by heating 2 g in CCl_4 with dinitrophenylhydrazine. The reaction was chromatographed and eluted, and the absorbance was determined. The menthol was first converted to menthone. The accuracy of the method is reported to be $\pm 0.4\%$. Mafredini *et al.* (1962) studied the ultraviolet spectrophotometry of some mint essential oils. The composition of *Mentha arvensis, M. viridis*, and *M. piperita* essential oils was obtained from ultraviolet spectra (in 3% EtOH solution) and for *M. arvensis* confirmed through vapor-phase chromatography. The concentrations of menthol, menthone, pulegone, and carvone were established and the data used successfully in the analysis of mixtures.

Pomerantz *et al.* (1957) developed a rapid spectrophotometric technique for quality evaluation of vanilla extracts. The rapid scanning spectrophotometer makes possible the detection of adulteration by a simple and rapid method, and appears to offer significant promise for quality control. Fitelson (1960) gave details of a modification of the old method of the AOAC for the determination of resins in vanilla extracts, with final measurements made either gravimetrically or spectrophotometrically. It was later adopted by the AOAC after a collaborative study. Smith (1965) described the details of an ultraviolet spectrophotometric method for vanillin based on the difference in absorbance between alkaline and neutral solutions of vanilla. By the use of suitable equations, both vanillin and *p*-hydroxybenzaldehyde can be determined. The method was more accurate than the AOAC colorimetric method or the alkaline ultraviolet method. Neuzil *et al.* (1967) made a spectrophotometric study of the color reaction of vanillin with Millon's reagent and the reaction mechanism. The color reaction of Millon's reagent with vanillin is dependent on concentration and time. The absorbance at 530 μm is directly proportional to the concentration of vanillin at a fixed concentration of the reagent. Triaxis graphs are given, showing the variation of absorbance as a function of the concentration of each reactant and as a function of Millon's reagent concentration and time. By various chromatographic techniques,

the violet product was determined to be a mercury chelate of 5-nitrosovanillin. The yellow coloration was found to be due to 5-nitrovanillin. The mechanism of formation of these substances is related to the nitrosation and nitration of phenols. The reaction with Millon's reagent is a result of these competing processes, with nitrosation being stabilized by the formation of chelate.

6. Column Chromatography

Karawya and Wahba (1962a) conducted the analysis of oil of peppermint by aluminum oxide–silicic acid double-column chromatography, eluting it with light petroleum ether, followed by volumetric and colorimetric determination of constituents. According to Karting *et al.* (1965), ether extraction of the roots and fruit of *Foeniculum vulgare* and resolution on an alumina column yielded five fractions, which were examined by TLC and infrared spectroscopy. The roots yielded stigamasterol, 7-hydroxycoumarin (umbelliferone), and an unidentified fraction, which, on acid hydrolysis, yielded a crystalline derivative, m.p. 60° to 80°C. The compound is provisionally classified as a triterpene glycoside (Karting *et al.*, 1965). During a recent analysis of lovage root oil, Lawrence *et al.* (1969b) encountered four interesting artifacts originating from preliminary column chromatography. On the basis of column (alumina) chromatography, they assumed that the artifacts—mesityl oxide, diacetone alcohol, phorone, and semiphorone—were obtained by the dimerization and trimerization of the acetone used as an eluent. Because of their polarity, they were preferentially absorbed on the alumina and were not eluted until methanol was used as an eluent. When Carbowax 20M was used as the stationary phase in the above analysis, semiphorone dehydrated to phorone. For this reason, they used SE-30 for all GLC separations.

7. Paper Chromatographic Techniques

Paper chromatography of a great number of terpenes with 15% EtOAc–*n*-hexane as the developing solvent showed that hydrocarbons had R_f values above 0.6, esters at 0.5 to 0.6, and alcohols below 0.50. Carbonyl compounds could be identified as the 2,4-dinitrophenylhydrazones (Katayama, 1960).

Buzas and Sqendrei (1964) developed an effective method for identifying more than one hundred crude drugs. The powdered drug (0.1 g) is extracted with 100 ml of BuOH saturated with 1% aqueous citric acid by macerating for 1 hour with occasional shaking. The filtrate is submitted to capillary analysis such as ring paper chromatography, with a defatted wick, and allowed to develop for 30 to 45 minutes. After drying, the pattern is observed with light, and colors of the various zones (up to six) radiating from the center are noted. The method is illustrated with twenty-seven drugs, including spices.

Beyrich (1964a) studied the behavior of the most important furocoumarins of the psoralene and angelicin types on paper chromatography by using the system

formamide or $HCONMe_2$ and heptane–benzene as solvent. The relations between structure and chromatographic behavior were discussed. The compounds were detected by their fluorescence and by means of the diazo reaction. In order to determine to which group an unknown compound belongs, the Emerson reaction is recommended, which is possible only for the angelicin class. It is possible to identify all the isomeric furocumarins investigated by their chromatographic behavior and their specific color reactions.

By means of adsorption chromatography and paper chromatography Nigam and Purohit (1961) identified pinene, dabinine, dipentene, cardinene, α-caryophyllene, and terpineol in the essential oil of *Murraya koenigii*.

Vilkki (1954) conducted chromatographic studies on the formation of pyruvic acid in onion juice.

Yoshimura (1958) used paper chromatographic techniques to separate allicin homolog of the formula RS(O)SR from diallyl sulfide homolog (RSSR) and from allyl mercaptan homolog (RSH), where R is an alkyl group. Color identification of allicin homologs was achieved with a solution of 0.9 g of iodine, 1.5 g of KI, and 2.5 g of NaN_3 in 150 ml of water. Yoshimura *et al.* (1958) reported that all the varieties of leek studied contained alliin and its methyl and propyl homologs. This was shown by paper chromatographic, paper electrophoretic, and enzymatic (determination of pyruvic acid and NH_3) methods.

Hemingway *et al.* (1961) examined eighty-six mustard seed samples for their mustard oils by paper chromatography. The presence of allyl isothiocyanate (I) only, 3-butenyl isothiocyanate (II) only, or a mixture of I and II in the seeds was correlated with geographical distribution. The results, and the fact that seeds of *Brassica nigra* contain I, and those of *B. compestris* contain II, indicate that *B. juncea* is a natural hybrid between these two species. This interesting observation needs further investigation.

Paris and Godon (1961) studied thirteen essential oils (naturally distilled), including parsley oil, present in medicinal preparations. The chromatographic methods based on waterproofed papers and glass plates were rapid, simple, and accurate enough even when very small quantities of essential oils were used. They may be used for studying the metabolism of essential oils in plants and identifying the oils used in pharmacy.

Svendsen (1960) reported that paper chromatographic and paper electrophoretic methods were the best means for the determination of morphine in opium. In both methods, spots may be determined directly by densitometry or indirectly by elution of the chromatogram or pherogram and colorimetric determination of the eluate; the indirect method is better. Paper chromatography was superior to electrophoresis under the conditions of the experiment used, since better separations were obtained. A gravimetric method with the precipitate formed with fluorodinitrobenzene following column chromatography gave good results, as did the International Pharmacopoeial method. Paris *et al.* (1961) determined

morphine by paper chromatography and paper electrophoresis in fifty papaver species cultured in France, containing 0.5 to 0.6% morphine. Izmailov *et al.* (1963) separated mixtures of narcotine, thebaine, papaverine, and other alkaloids from poppy capsules by chromatography on paper treated with buffer solutions having pH 4.0 or 6.6. The accuracy of the method was 1.6% for narcotine, 1.15% for thebaine, and 1.12% for papaverine.

Herndon *et al.* (1963) reported that the combination of uniform solvent delivery, centrifugal force, and strip groove modification in the chromatograph apparatus and the use of a tandem technique made horizontal paper chromatography useful in the study of dye separations, including saffron. Clear, reproducible separations can be effected in just 10 minutes. Paris and Rousselet (1958) studied the characterization of dyes of vegetable origin, including turmeric and saffron, by paper chromatography.

Beyrich (1964b) isolated the furocoumarins—bergapten, m.p. 193° to 195°C; xanthotoxin, m.p. 146° to 148°C; and imperatorin. m.p. 99° to 100°C—from parsnips. Mixtures of bergapten (3.2 to 22.4 μg), xanthotoxin (3.5 to 34.2 μg), and imperatorin (3.5 to 18.0 μg) were separated by chromatography. The standard deviations of the above determinations of bergapten, xanthotoxin, and imperatorin were $\pm$12.9%, $\pm$5.2%, and $\pm$13.9%, respectively. The molar absorptivities were 1.44×10^4, 1.2×10^4, and 1.29×10^4, respectively. The method for quantitative determination in celery, parsley, and parsnip has been described. The seasonal variations in parsnips were determined: bergapten, 4 to 44.4 μg, average 24.0; xanthotoxin, 16.6 to 55.2 μg, average 34.8; and imperatorin 17.9 to 46.2 μg, average 31.8, respectively. The average deviations were $\pm$7.9%, $\pm$4.6%, and $\pm$6.1%, respectively.

Woggon and Kohler (1960) described a method for the determination of vanillin and ethyl vanillin in foods by first separating the two aldehydes in a suitable extract by paper chromatography and then determining each in semicarbazide hydrochloride solution with the oscillopolarograph. Tests on model mixtures and commercial products gave reproducible results, an average error of 10% being found on quantities of 2 μg of vanillin or ethyl vanillin per milliliter. The method is simple and rapid and hence suitable for routine determinations. Fitelson (1962b) reported collaborative studies on a qualitative test for vanilla resins, which showed that, depending on the characteristic fluorescent pattern of vanilla resins after development by paper chromatography, Fitelson's (1960) test can provide satisfactory confirmatory information. The AOAC quantitative resin method gives reliable results with authentic samples. By paper chromatographic separation, Kleineri (1963) detected *p*-hydroxybenzaldehyde in alcoholic extracts of Bourbon and Tahiti vanilla stalks. Vanillin and *p*-hydroxybenzaldehyde appeared in the seed body as well as in the fruit pulp. Eleven physical and chemical quality control tests for vanillin were described. In vanilla fruit, the AOAC method and the fractocolorimetric method were in closer agreement,

although the latter method recorded somewhat low values (Kleineri, 1963). Additional qualitative tests for vanilla resins and supplemental two-dimensional paper chromatographic detection of foreign plant matter in vanilla extracts have also been described (Anonymous, 1962b). Stoll and Barnier (1964) used ascending paper chromatography to separate hydroxy methyl furfural and various vanillins. Results are correlated by spectrophotometry. Synodinos *et al.* (1964) separated components of vanilla extracts by chromatography on Whatman No. 1 paper by using BuOH saturated with 2% NH_4OH as developing solvent. The spots were made visible by treatment with *p*-nitroaniline. The R_f values were 0.92. Thin-layer chromatography on 2: 1 silicic acid–Celite containing 5% each of MgO and starch yielded the following R_f values: vanillin 0.58, ethyl vanillin 0.76, *p*-hydroxybenzaldehyde 0.65, piperonal 0.98, coumarin 0.95, p-$(HO)C_6H_4CO_2H$ 0.35, and vanillic acid 0.28. Fitelson (1965) adapted a number of methods for vanilla extracts for use in vanilla powders, including lead number, vanillin, ethyl vanillin, and organic acids by paper chromatography, and resin. Methods are given for the preparation of solutions for analysis of powders with little or no starch and of powders with starch. Results on samples compared well with results on the original extracts or concentrates. The paper chromatographic method for foreign plant material was not suitable for powders. Fitelson (1967) conducted a second collaborative study on two ultraviolet absorption methods for vanillin and a paper chromatographic method for the detection of vanillin and ethyl vanillin when present together. All three methods gave adequate and reproducible results. Both the modified ultraviolet method and the paper chromatographic procedures are recommended for adoption as official. Guenther *et al.* (1959, 1961, 1963, 1965, 1967, 1969, 1971, 1973, 1975, 1977) have reviewed the literature on different analytical techniques for vanillin, etc., including paper chromatographic techniques.

8. Thin-Layer Chromatography

Since the work of Kirchner *et al.* (1951), Kirchner and Miller (1952), and Miller and Kirchner (1952, 1953), considerable attention has been directed toward thin-layer chromatography and its use in the analysis of essential oils (including spice essential oils) and their constituents. Because of its simplicity, the technique lends itself to a rapid screening of essential oils for semiquantitative composition studies. It also provides an excellent means for the detection of adulterants. In this latter area, the pharmaceutical industry has been active, especially in the field of umbelliferous and labiatious oils. A number of reviews have appeared in the literature on the use of TLC in essential oil analysis. Of particular interest are the chapters by Stahl (1962) and Kirchner (1967) in their comprehensive texts covering the whole range of TLC and also the excellent reviews by Lawrence (1968), by Guenther *et al.* (1959, 1961, 1963, 1965, 1966, 1967, 1969, 1971, 1973, 1975, 1977), and by Guenther (1966).

Essential oils are very complex, possessing as many as fifty to three hundred constituents, with their percentages covering many orders of magnitude. There are, however, a small number of major constituents that allow a means of monitoring the oils for purity and detection of adulteration by TLC.

The fundamental questions involved in TLC of essential oils are (1) the type of layer to be used, (2) the solvent system, and (3) the eluent. The choice of a suitable solvent system is the most difficult problem. Lawrence (1968) has listed the following top ten solvent systems in order of their popularity: (1) benzene; (2) benzene–ethyl acetate (95:5); (3) hexane–ethyl acetate (85:15); (4) hexane; (5) benzene–ethyl acetate (90:10); (6) chloroform–benzene (1:1); (7) chloroform; (8) hexane–ethyl acetate (95:5); (9) benzene–ethyl acetate (85:15); and (10) hexane–ethyl acetate (90:10).

For the detection of compounds on the plates, the use of ultraviolet light can be of some advantage either with or without a fluorescent spray or impregnation of the layer, but not all the compounds show up. However, general or specific spray reagents are employed for the purpose. The reagents shown in Table XXI are those most widely used for the detection of terpenoid compounds; a reference to any text on chromatography will give the procedures for the preparation of these reagents.

Bhramaramba and Sidhu (1963) used TLC to study the nonphenolic portion of *Cinnamomum zeylanicum* leaf oil of Indian origin. They identified benzyl benzoate, methyl eugenol, *p*-cymene, and linalool and found three unknown compounds using silica gel layers with benzene as the eluent and formalin in sulfuric acid as a detecting reagent. In order to determine the botanical origin of some *Cinnamomum* species, Betts (1965b) used chromatographic separations on a thin layer of magnesium silicate. He was able to differentiate between the eugenol- and noneugenol-containing species using the Folin–Ciocalteu reagent.

Voelker *et al.* (1967) proposed a procedure for the determination of the geographical origin of different types of commercial cinnamons and cassia. The technique was designed primarily for quality control; however, unknown mixtures of cinnamon and cassia can be determined to within ±20% of one type in the mixture. Tanaka and Ono (1965) used TLC to remove the interfering constituents from red camphor oil in their safrole determination by infrared spectroscopy. In a study of the hydrocarbons in the oil of *Lauris nobilis* (laurel), Chow *et al.* (1965) used silver nitrate-impregnated layers to separate γ- and δ-cadinene and β-selinene from the sesquiterpene fraction of the oil.

Schultz and Mohrmann (1965a) studied fourteen solvent systems and found that carbon tetrachloride–methanol–water (20:10:1) and (21:10:2) were the best eluents for the analysis of garlic *(Allium sativum)*, by means of which nine and fourteen oil constituents, respectively, were separated. They found that diallyl disulfide was the main constituent of the oil.

TABLE XXI CHROMATOGRAPHIC SPRAY REAGENTS USED TO DETECT TERPENOID COMPOUNDS[a]

Reagent	Uses
Sulfuric acid	All organic compounds
Ammonium hydrogen sulfate	All organic compounds
Antimony trichloride	General
Antimony pentachloride	General
Antimony trichloride-pentachloride	General
Phosphomolybdic acid	General
Chlorosulfonic acid	General
Vanillin-sulfuric acid	General
p-Dimethylaminobenzaldehyde-sulfuric acid	General
Diphenylpicryl hydrazyl	Unsaturated compounds
Fluoroscein-bromine	Unsaturated compounds
Iodine	Unsaturated compounds
Potassium permanganate	Unsaturated compounds
2,4-Dinitrophenyl hydrazine	Carbonyl compounds
Stable diazonium salts (i.e., Fast Blue Salt B)	Phenols
Acid-phloroglucinol	Phenols
Folin-Ciocalteu reagent	Phenols
Ferric chloride	Phenols

[a] From Lawrence (1968).

Wulff and Stahl (1960) used TLC for the identification of the geographical origin of a number of *Acorus calamus* oils.

Using benzene, benzene–5% ethyl acetate, and hexane as solvent systems, Tyihak and Vagujfalvi (1962) investigated the extracted or distilled oils of *Artemisia absinthium, A. dracunculus* (tarragon), *Foeniculum vulgare, Mentha crispa, M. longifolia,* and *M. piperita.* Jaspersen-Schib (1961) also used the technique to examine a number of volatile oils for purity and identification of constituents. The same author was able to show the presence of adulterants in some of the oils investigated. In the third of a series of papers on the chromatographic analysis of essential oils, Pertsev and Pivnenko (1962), using 15% ethyl acetate in petroleum ether as an eluent, carried out the microanalysis of coriander, lavender, and nutmeg oils, identifying the major constituents by comparing their R_f values with those of standard compounds. Pertsev (1963) reviewed the use of chromatography in the analysis of pharmaceutically important essential oils. Paris and Godon (1963) used the techniques of waterproof paper as well as TLC on silica gel to determine the constituents of twenty-one different oils as their mercury derivatives. Vagujfalvi and Tyihak (1963) studied the thin-layer chromatographic separation of ten different essential oils and, from their results, specific constituent properties were determined.

Horhammer *et al.* (1964b) used TLC as a method of analyzing the pharmaceutically significant umbelliferous oils. Using benzene as a solvent, they separated the major constituents of anise and fennel oils; with 5% ehtyl acetate in benzene, the major constituents of coriander, caraway, parsley, and dill were also identified. Horhammer *et al.* (1960) used TLC to detect adulteration of *Pimpinella saxifrage* by *Heracleum sphondylium,* and in a second paper Horhammer *et al.* (1963) used alumina layers to separate the constituents of *Angelica sylvestris*. Atal and Shah (1964) used TLC in a qualitative study of the umbelliferous fruits and their adulterants on silica gel layers with benzene and 5% ethyl acetate in benzene as eluents. The fruits they studied were caraway, coriander, fennel, cumin, and ajowan; however, no components were identified. The technique could not be applied to ajowan. The oils of coriander, fennel, cumin, and dill were studied chromatographically by El-Hamidi and Ahmed (1966) to find out the seasonal variations in the compositions of these essential oils of Egyptian origin. They found that there was a positive correlation between the growth of the plant and oil composition and that there were wide variations in the magnitude of constituents present.

El-Deeb *et al.* (1962b) used 15% ethyl acetate in hexane to identify limonene and carvone in caraway oil on silicic acid layers. This separation was compared with the official method of the Egyptian Pharmacopoeia, and the results showed a favorable agreement. Preuss (1964) used TLC to study the photochemical reaction that can occur in the aging of caraway oil constituents. In this study, five possible hydroperoxides were postulated as being formed by the autoxidation of limonene, three of which were detected by TLC using 13% diethyl ether in hexane as a solvent system.

Karawya and Wahba (1962d) used TLC to identify α-pinene, anethole, and anisaldehyde in anise oil. Anise seed oil was studied chromatographically by Zacsko-Szasz and Szasz (1965) using benzene–chloroform (1:1) as the eluent on silica gel layers. They found that, during storage of the oil, the percentage of 4-allylanisole decreased, whereas the percentage of anisaldehyde and methyl chavicol increased.

Brieskorn and Zimmerma (1965) isolated squalene, anethole, and pristane (2,6,10,14-tetramethylpentadecane) from the unsaponifiable fraction of a hexane extract of anise fruits by chromatography on a silica gel column and elution with petroleum ether. The identification of pristane was proved by gas–liquid chromatography on two columns with different polarity, by mixed chromatograms with authentic substance, and by infrared spectrophotometry, density, refractive index, etc. Zacsko-Szasz and Szasz (1965) outlined the TLC method and gave the R_f values and typical chromatograms of pure compounds (anethole, anisaldehyde, anise ketone, chavicol methyl ether, anisic acid, and 4,4′-dimethoxystilbene) and of samples of aniseed oil. Zacsko-Szasz and Szasz (1966) reported that the main component in aniseed oil is anethole and that the quality of the oil

depends on the presence or absence of anethole derivatives. They are either oxidized forms of anethole, such as anisaldehyde, anise ketone, and anisic acid (4-methoxybenzoic acid), or anethole derivatives, such as photoanethole (4,4′-dimethoxystilbene). All these compounds can be satisfactorily separated by TLC on activated kieselghur with C_6H_6-$CHCl_3$ (1 : 1) as a developing solvent. Various chromogenic agents were used to observe the spots, and the color obtained with these reagents served as identification. Anise ketone, anisic acid, and photoanethole were not present in properly handled aniseed oils. The presence of anethole derivatives therefore indicates degradation, and TLC can serve as a method for quality control of aniseed oils.

Deryng *et al.* (1966) used TLC to study the constitution of coriander oil and caraway oil. By using a 5:1 mixture of benzene and ethyl acetate, they separated a number of constituents. For detection purposes, they found that 1% *p*-dimethylaminobenzaldehyde in 75% sulfuric acid was the most useful reagent tried. Sidhu and Bharmaramba (1963) studied the chromatography of cinnamon leaf oil.

Continuing their study of the volatile oils described in the Polish Pharmacopoeia, Deryng *et al.* (1967) identified α-pinene, limonene, anethole, and anisaldehyde in anise oil, and anethole, fenchone, anisaldehyde, and a terpene hydrocarbon in fennel oil. In another chromatographic study of fennel oil, Osisiogu (1967) found that Nigerian fennel oil contained about 86% anethole, but no fenchone could be found. Ishibashi *et al.* (1967) used both TLC and GLC to identify eleven constituents in European fennel oil. Using TLC, Betts (1964) identified the extracts of unknown umbelliferous fruit mixtures with benzene–chloroform (1 : 1) as a solvent system. He was able to determine 20% of *Anethum sowa* (Indian dill) in *Anethum graveolens* (North American or European dill), but he was not able to distinguish caraway from dill or anise from fennel. In their preliminary investigation of Egyptian cumin oil, El-Hamidi and Richter (1967) used TLC to identify cuminaldehyde, cumin alcohol, perillaldehyde, and an unknown ketone, which they postulated as being cryptone.

Lassanyi (1965) used TLC to determine semiquantitatively the linalool content of coriander oil. The method was found to be quite favorable within a certain range of linalool concentrations. In a study of the composition of the essential oil of *Coriandrum sativum* (coriander), Schratz and Qadry (1966) identified geraniol, linalool, geranyl acetate, decanal, thymol, and *trans*-tridecen-2-al. In another paper, Reisch *et al.* (1966) synthesized *trans*-tridecen-2-al and used TLC to compare the synthetic unsaturated aldehyde with the naturally isolated compound. Continuing their study, Schratz and Qadri (1966) used TLC to compare the oil composition of *Coriandrum sativum* var. *macrocarpum* of Indian and German varieties during ontogenesis. They found that, although the Indian and German corianders were morphologically dissimilar, they were similar in composition and behavior, except that thymol could be found only in the Indian variety.

The oils of angelica, anise, calamus, caraway, coriander, fennel, juniper, pep-

permint, sage, thyme, and mustard were studied by Zielinska-Sowicka and Szepczynska (1965), using TLC as a means of general identification of components present in the above oils that are described in the Polish Pharmacopoeia. The method is recommended because of its simplicity and the high resolution obtained. Thin-layer chromatography was also applied to a number of essential oils by Nigam *et al.* (1965b), who showed that this technique could have great use in taxonomic and phytogenetic problems of essential oil-bearing plants. Kaul and Nigam (1966) analyzed caraway and dill oils by both column and thin-layer chromatography.

Horhammer *et al.* (1964b) used benzene as an eluent to identify safrole, anethole, anisaldehyde, farnesol, and terpineol in star anise oil. Using GLC and TLC, Ishibashi *et al.* (1967) identified fourteen components in Chinese star anise oil, one of which was *cis-p*-menthene-3,6-diol, which they postulated as probably being formed by the autoxidation of α-phellandrene.

In a series of papers on mint oil, Ito *et al.* (1953, 1954) analyzed a number of oils of Japanese origin. Reitsema (1954) and Reitsema *et al.* (1957, 1961) used the technique of Kirchner *et al.* (1951), Kirchner and Miller (1952), and Miller and Kirchner (1953) to characterize many essential oils, especially the oils of the *Mentha* species, and in this study most of the major constituents were identified. With the aid of both TLC and column chromatography, Pertsev and Pivnenko (1961, 1962) identified the following constituents in the pharmaceutically important oil of peppermint: menthol, menthyl acetate, cineol, menthone, and limonene. They used a number of specific detecting reagents and also pure substances as standard compounds to assist them in the identification. Using ethyl acetate–cyclohexane (15:85), Karawya and Wahba (1961–1962) identified menthol, menthone, menthyl acetate, and terpene hydrocarbons in peppermint oil. Jaspersen-Schib (1961) used TLC to examine and standardize procedures for the analysis of the volatile oils of the *Mentha* species, and from the results he was able to show how the oils from one species could be found as adulterants in the oil of another species. Battaile (1960) studied the chromatography of peppermint oil terpenes on silicic acid chromatoplates with AcOEt in hexane as developing solvent and separated known components of the oil. A sequence of examinations of the plates allowed the detection and recovery of several compounds in commerical peppermint oil and leaf extracts. The method is so sensitive that routine examination of the constituents of the oil from the pair of leaves at a single node of the plant was possible. Avakova *et al.* (1963) used TLC in conjunction with GLC and column chromatography to study the composition of a low menthol-containing peppermint oil. Rumanian peppermint oil was chromatographically analyzed for constituents by Rothbacher *et al.* (1964), using ethyl acetate–benzene (5:95) as the eluent. In a second paper, Rothbacher (1965) identified menthol, cineol, limonene, menthone, menthyl acetate, and menthofu-

ran in the same oil. Using the technique of coupling GLC and TLC, Nigam and Levi (1964) were able to determine a classical distinction between *Mentha arvensis* and *M. piperita*. This technique was proposed as a method of determining *M. arvensis* as an adulterant in the oil of *M. piperita* by the methofuran content of the mixture due to the former. The same authors also showed that trace amounts of menthofuran could be found in the volatile oils of *M. cardiaca, M. spicata, M. pulegium, M. citrata, M. sylvestris,* and *M. rotundifolia*. Nigam *et al.* (1963b) used coupled GLC and TLC to identify piperitone and piperitone oxide in *M. piperita* and *M. arvenis*. In a later paper, Nigam *et al.* (1965b) showed the different chromatographic patterns of *M. cardica, M. spicata,* and *M. virdis* using vanillin–sulfuric acid as a detection reagent.

Juhasz *et al.* (1965a,b) used TLC to separate the components of the essential oils of *Lavandula officinalis, L. vera,* and *M. piperita* from steam distillation residues.

Using a solvent system of 2% ethyl acetate in hexane, Tatar (1964) qualitatively studied the volatile oil of paprika on silica gel layers. Heusser (1964) has described a procedure for accurate determination of capsaicin in capsicum and morphine in powdered opium. The technique is rapid enough for an experienced operator to perform eight to ten morphine determinations in 2 days.

Klohr-Meinhardt (1958a,b) utilized TLC in a study of the synthesis of volatile oils in parsley and lovage. This technique was also useful in a study of the synthesis of volatile oils in a heteroplastic graft in the same unbellifers. Stahl and Jork (1964) used TLC in their studies on the essential oils of twenty-eight varieties of *Petroselenium hortense* (European parsley). They monitored the oils on the basis of their myristicin, apiole, and 1-allyl-2,3,4,5-tetramethoxybenzene contents and found that the results were independent of geographical origin.

Chopra (1965) used TLC to separate and identify caryophyllene oxide, cryptone, dihydrocarveol, and piperonal in black pepper oil, using mixtures of heptane and diethyl ether as the solvent systems.

Jaspersen-Schib and Flueck (1962) compared the use of TLC and paper chromatography in the analysis of the essential oils of *Rosmarinus officinalis, Lavandula officinalis,* and *L. latifolia*. In a more general paper on the TLC of volatile oils, the same authors (1961) identified borneol, cineol, limonene, bornyl acetate, and pinene in rosemary oil. The compositions of rosemary oil and rosemary wine were chromatographically determined by Graf and Hoppe (1964), and a number of constituents were identified. In a phytochemical study of the oil of *R. officinalis* of Egyptian origin, Saleh and Mohamed (1965) identified borneol, bornyl acetate, camphor, cineol, and α-pinene. Karawya and Wahba (1962b) used ethyl acetate–hexane (15 : 85) as the solvent system to determine chromatographically the presence of camphene, α-pinene, bornyl acetate, camphor, and borneol in rosemary oil. They found that this method compared fa-

vorably with the existing ones described in the Egyptian and British Pharmacopoeias. The new method was then recommended as a general technique for the analysis, particularly because it required only small samples of oils.

Using GLC and TLC, Brieskorn and Wenger (1960) identified a number of constituents in the volatile oil of *Salvia officinalis*. They found borneol, camphor, bornyl acetate, thujone, linalyl acetate, and pinene in the oil by comparing their R_f values with those of standard compounds and also by specific color reactions with the vanillin–sulfuric acid reagent. Both red and white Egyptian basil oils were analyzed chromatographically by Horhammer *et al.* (1964a), and some differences in the chemical composition of the two oils were found.

Phokas (1966) used TLC as a method of determining adulterants in the essential oil of *Melissa officinalis*. Some of the potential adulterants were *Dracocephalum moldavicum* and *Cymbopogon winterianus*, and these could be determined by the technique described.

Using 5% mixtures of chloroform and ethyl acetate in hexane or heptane as solvent systems, Gogrof (1957) chromatographically studied the volatile oil of patchouli, and some sesquiterpenes were isolated.

The oils of *Thymus vulgaris, T. seropyllum*, and *Origanum creticum* were analyzed chromatographically by Gabel *et al.* (1962) for their thymol and carvacrol contents. Messerschmidt (1964) used TLC to study the effect of storage on the herbs *T. vulgaris* and *T. pulegioides*. It was noted that, although there were variations in the oil contents, the composition of the oils remained fairly constant. The volatile oils of *T. zygis* and *T. serpyllum* were further studied by Messerschmidt (1965), again using TLC and GLC to determine the differences in the two oils. From this study, the oil of thyme on the German market could be chemically classified as being obtained from a certain *Thymus* species. Using silica gel layers and a benzene–ethyl acetate (9:1) solvent system, Schratz and Qedan (1965) chromatographically fractionated into classes the components present in *T. serpyllum* oil. By further TLC using other mobile phases, each component class was separated into a number of constituents until they were able to isolate twenty-six components, of which eighteen were positively identified.

Ficicchia (1966) used silica gel layer chromatography to separate cineol, terpineol, and the essences of rosemary and thyme.

Winkler and Lunau (1959) used TLC to differentiate between *Curcuma xanthoriza* and *C. longa* (turmeric), with benzene–chloroform–menthanol (200:100:3) as a solvent system.

In 1965, Chopra chromatographically identified the acetates of terpineol, geraniol, linalool, and borneol as well as methylheptenone, linalool, nerol, borneol, and geraniol in cardamom oil, using benzene as the eluent.

In 1962, Sundt and Saccardi carried out an extensive study on the use of thin-layer and paper chromatography for the investigation of vanilla-like flavoring compounds. With the solvent systems used they were able to identify seven-

teen compounds at a sensitivity of 0.1 μg. Kahan and Fitelson (1964, 1965) used these results for the proposal of a technique to determine flavor additives in natural vanilla extracts. The method allows adulterants such as ethyl vanillin, methyl vanillin, piperonal, and propenyl guaethol to be determined. Synodinos *et al.* (1964) prepared their own layers from a mixture of silica gel, Celite, magnesium oxide, and starch and were able to identify a number of constituents in natural vanilla extracts using *n*-butanol saturated with 2% of ammonium hydroxide as an eluent.

9. Gas Chromatographic Techniques

The chemical analysis of food aromas has been an extremely difficult undertaking, principally for the following reasons: (1) the large number of components present; (2) the low concentrations of many of these compounds; (3) the complex relationship of chemical composition to olfaction; (4) the highly specific molecular structure–flavor relationship; and (5) the high degree of susceptibility of many of these compounds to chemical change.

The development of gas chromatography appears to provide a practical means of objective analysis of food aromas. James and Martin (1952) were the first authors to introduce GLPC as an analytical method for the determination of volatile esters of fatty acids ranging from formic acid to dodecanoic acid.

By virtue of the highly efficient fractionations possible with gas–liquid chromatographic columns, separation of complex mixtures becomes much less difficult. The development of hypersensitive detectors (ionization detectors) has provided a means of detecting concentrations at the parts-per-billion level. The very wide choice of column packings, lengths, and temperatures has made possible separation of compounds with small differences in molecular structure. Further, the gas chromatographic analysis may be carried out under conditions of low temperatures, inert atmospheres, and small sample size. Such an analysis is rapid, accurate, and reproducible. Consequently, attempts have been made to correlate gas chromatograms of foods including spices and spice essential oils with organoleptic evaluations. Some important aspects of GLPC of spices and spice essential oils are briefly reviewed here.

Teisseire (1962) presented reviews of the problems encountered in the application of gas chromatography to the raw materials of perfumery, including spice essential oils.

Nigam *et al.* (1963a) found the following trace constituents in ajowan oil by column and gas–liquid partition chromatography with SAIB and Reoplex 400 as stationary phases: camphene, β-pinene, 3-carene, and myrcene in the thymene fraction, and a hitherto unreported alcohol in PhOH eluate.

Kellogg *et al.* (1964) studied the gas chromatography of esters of plant acids and their identification in plant materials such as angelica.

Ishibashi *et al.* (1967) separated star anise oil from China into fourteen com-

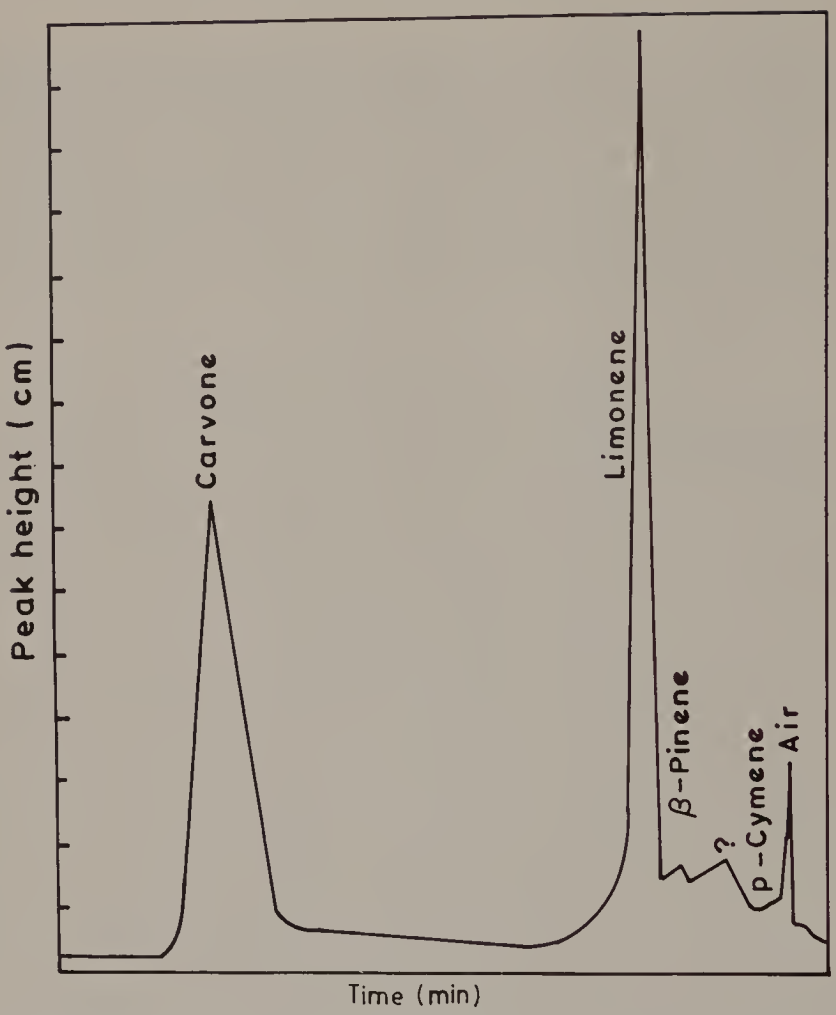


FIG. 4. Gas–liquid chromatogram of essential oil of caraway (*Carum carvi*). Column: polypropylene glycol(15%) on chromosorb-W; temperature 200°C; argon 60 ml/min; sample size 2. From Atal and Sood (1967).

ponents by precision distillation and gas chromatography, and identified each component by thin-layer chromatography and infrared spectral analysis. For comparison, the same analytical procedure was applied to a fennel oil from Europe, and three of the fourteen components were not detected.

The oil of the Japanese star anise (*Illicium verum*) was analyzed by gas–liquid chromatography by Cook and Haward (1966). The major constituents were found to be cineol (18.1%), linalool (10.1%), methyl eugenol (9.8%), a terpenyl acetate (6.8%), safrole (6.6%), and a sesquiterpene hydrocarbon of unknown constitution (7.23%). The composition of this oil differs widely from that of the commercially used star anise oil obtained from *I. verum*. The most striking difference between the two oils is found in the anethole content, which constitutes

88% of the commercial oil but only 1.2% of the oil investigated in the present study.

Wheeler *et al.* (1961) reported the analysis of some aldehydes and ketones in caraway and sage oils by gas chromatography at 150°C on a Craig polyester succinate column. They found carvone in caraway oil (58%), and in sage oil they reported thujone (54%) and camphor (2.6%).

Atal and Sood (1967) undertook a study of the quality of Indian *Carum carvi* to determine if it compares favorably with caraway of European origin. The oil of *C. carvi* of Lahaul origin appears to be of excellent quality. The presence of carvone and limonene was demonstrated by isolation of these constituents by fractionation and by preparation of suitable derivatives. Gas–liquid chromatography (Fig. 4) revealed that carvone and limonene together constitute the major bulk of the oil, the other constituents being present in very minor portions. The yield of oil from Lahaul caraway is 5 to 8%, and the carvone content is 63.3% (the pharmacopoeial standard is 53%).

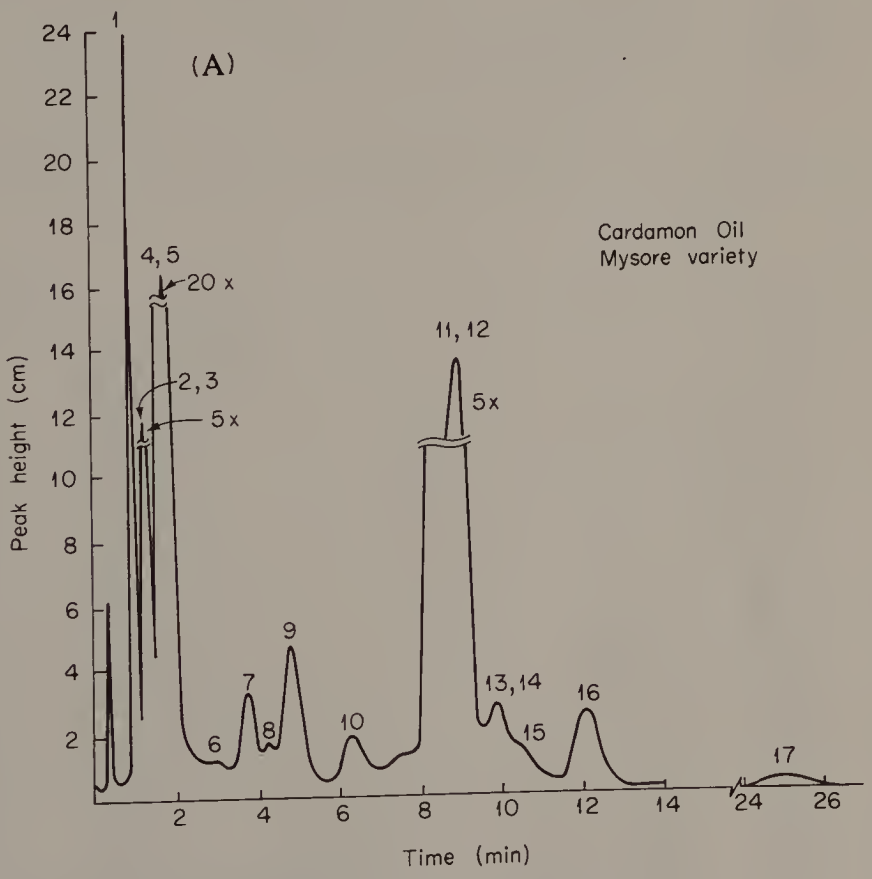


FIG. 5A. For caption, see p. 100.

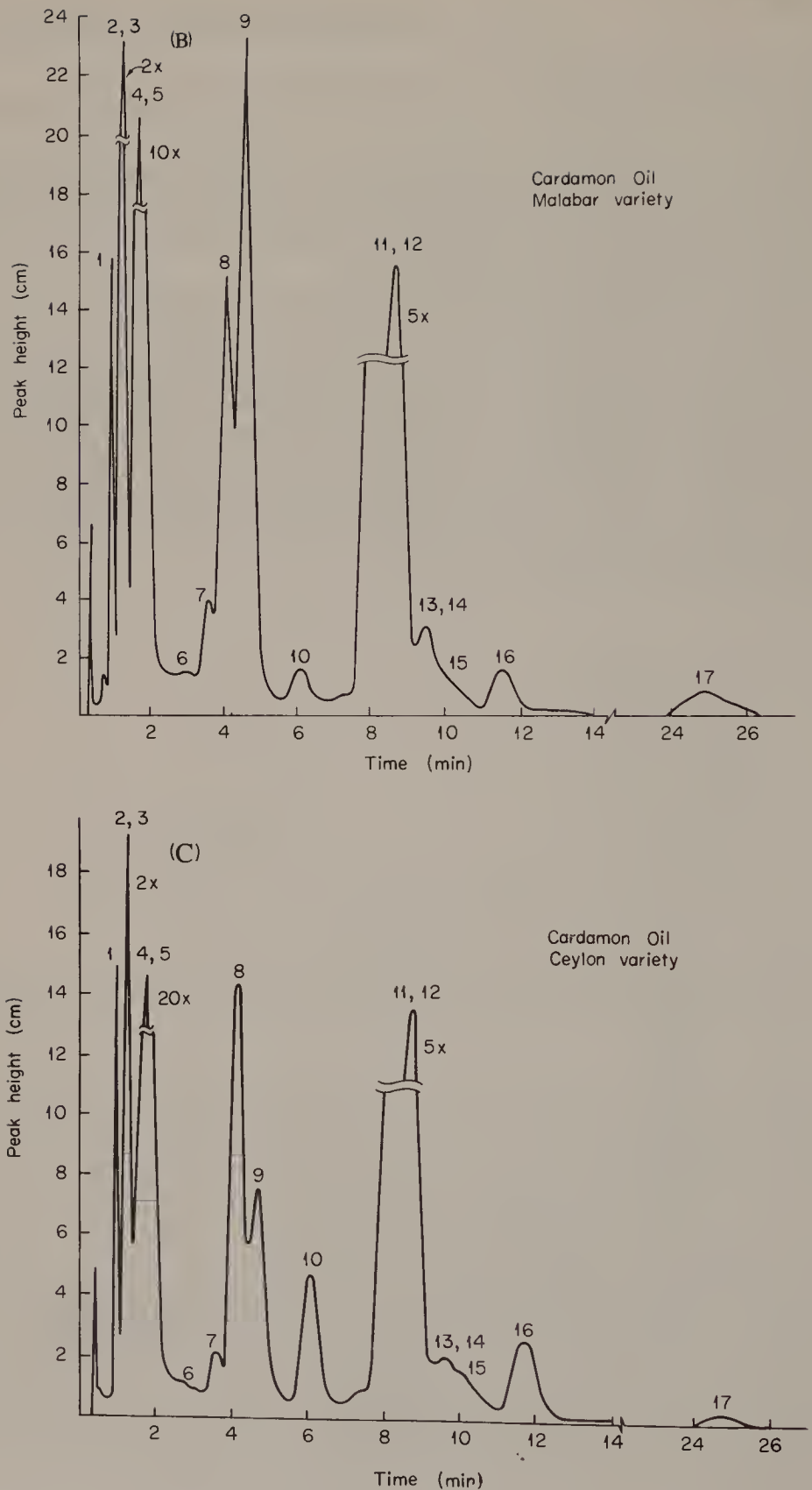


FIG. 5. Gas–liquid chromatograms of cardamom oils: (A) Mysore, (B) Malabar, and (C) Ceylon varieties. From Lewis *et al.* (1966).

Wellendorf (1966) studied the gas chromatography of freshly extracted essential oils from various types of commercial cardamom seeds obtained by Clevenger's method. The gas chromatograms were qualitatively similar. The major ingredients were identified as cineol and terpinyl acetate. No other peaks were identified, but some retention times corresponded to those of α-pinene, sabinene, and borneol. The α-terpineol frequently claimed as a constituent was absent in all the oils. The best-quality cardamom seeds have a high concentration of terpinyl acetate. Lewis *et al.* (1966) studied the differences in flavor of essential oils from three commercial varieties of cardamom—Mysore, Malabar (Kerala), and Ceylon or Sri Lanka (Fig. 5A–C)—by using thin-layer column and gas–liquid chromatography and found that the differences in the three oils were mainly quantitative (Table XXII).

Datta *et al.* (1962) studied the feasibility of using gas chromatography for the identification of the geographical origin of some important spices such as black pepper, ginger, cassia, and nutmeg oils (Figs. 6–8). Suitable conditions were developed for gas chromatographic separation of the components of oils (Table XXIII). Comparison of the peak heights of some of the major components of these spice oils showed marked and consistent differences among spices from

TABLE XXII COMPOSITION OF INDIAN VARIETIES OF CARDAMOM OILS[a]

Peak number	Constituent	Cardamom varieties		
		Mysore	Malabar	Ceylon
1	α-Pinene	1.4	0.7	0.7
2	Sabinene	3.2	4.6	3.8
3	Myrcene	0.2	0.2	0.1
4	Limonene	2.4	1.7	2.2
5	Cineol	41.0	26.5	36.0
6	*p*-Cymene	0.5	0.7	0.4
7	Methyl heptonone	1.2	1.5	0.8
8	Linalool	0.4	3.7	3.5
9	Linalyl acetate	1.6	7.7	2.6
10	β-Terpineol	0.8	0.7	2.1
11	α-Terpineol	0.8	1.0	1.0
12	α-Terpinyl acetate	30.0	34.5	30.0
13, 14	Neryl acetate (+ borneol?)	1.1	1.2	0.8
15	Geraniol	0.7	0.7	0.7
16	Nerol	1.4	0.6	1.6
17	Nerolidol	0.3	0.7	0.3
18	Solid (heptocosane?)	0.5	0.4	0.5
19	Unidentified compounds	12.5	12.9	12.9

[a] From Lewis *et al.* (1966).

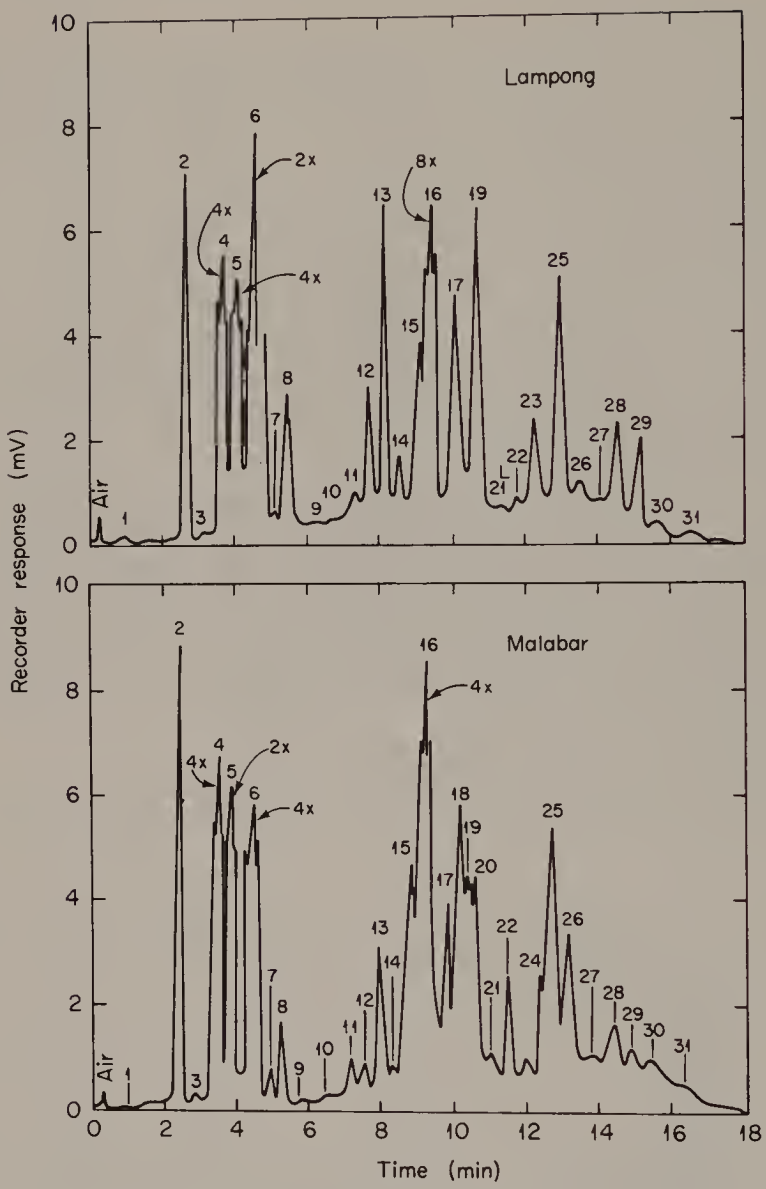


FIG. 6. Linear-temperature-programmed gas chromatograms of steam volatile oil from two black peppers. From Datta *et al.* (1962).

different geographical origins. The characteristic components were identified by micro-infrared spectroscopy (Table XXIV).

Wilson (1965) conducted studies on the separation of volatile flavors of celery essential oil. Gas–liquid chromatography showed fifty compounds. The flavor potency of the recovered essential oil was high.

Shellard (1967) examined fifteen samples of coriander fruits for volatile oil content according to Polish Pharmacopoeia III and British Pharmcopoeia 1963 and for linalool by gas–liquid chromatography. The Moroccan variety showed

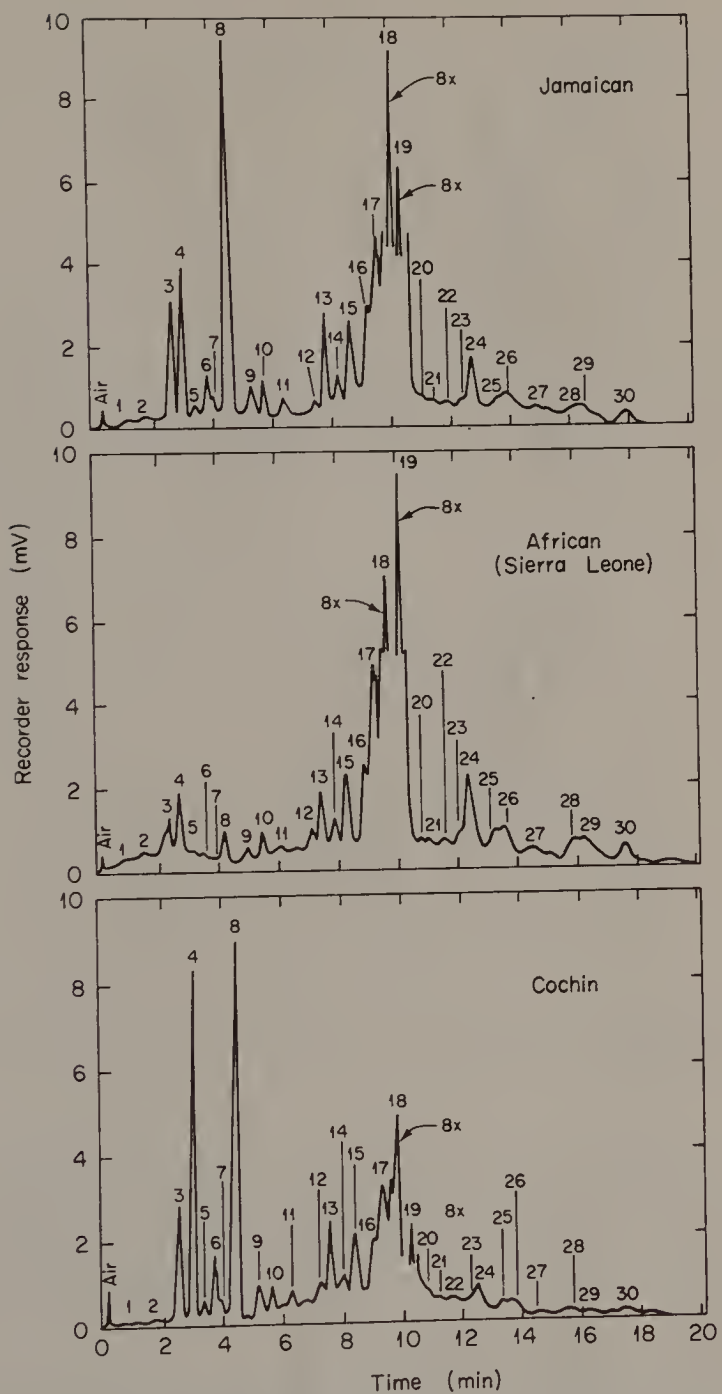


FIG. 7. Linear-temperature-programmed gas chromatograms of steam volatile oil from ginger. From Datta *et al.* (1962).

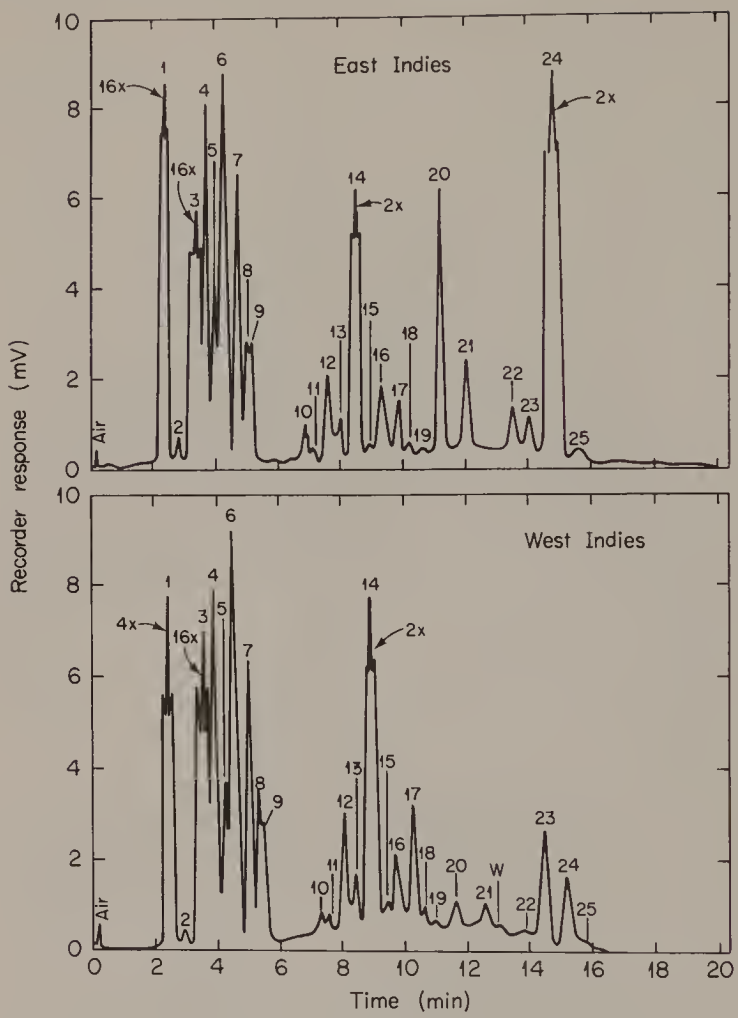


FIG. 8. Linear-temperature-programmed gas chromatograms of steam volatile oil from nutmeg. From Datta *et al.* (1962).

0.33%, the British variety 0.55%, and the Polish variety 1.15%. The British varieties grown in Poland showed as much as 0.85% oil. Several physiochemical constants were reported for the oils examined as well as their infrared, spectral, and composition data obtained from thin-layer chromatography. The oil prepared from the Polish variety (*Coriandrum sativum* var. *midrocaroum*) met the requirements of the British Pharmacopoeia.

Jain *et al.* (1962) studied gas–liquid chromatography of monoterpenes and its application to essential oils. The monoterpenic fractions of nine essential oils (including dill seed oil) collected by fractional distillation with an ice-cold condenser to prevent the loss of any low-boiling material and the relative corrected retention volume of the monoterpene with *d*-limonene as the reference compound were tabluated. The data presented were obtained on silicon and Car-

TABLE XXIII GAS–LIQUID CHROMATOGRAPHY OPERATING CONDITIONS[a,b]

Spice sample	Temperature (°C)			Length and type of column	Liquid phase	Helium gas flow rate (ml/min)	Sample size (μl)	Chart speed (inches/min)
	Column	Detector	Injector					
Cassia								
Isothermal	185	250	250	6-ft hairpin	Craig	40	2	0.5
Linear program	89–220	300	300	5-ft coil	succinate (15%)	50	2	0.2
Black pepper								
Isothermal	195	250	250	6-ft hairpin	Carbowax	40	5	0.5
Linear program	89–240	300	300	5-ft coil	20M (15%)	50	2	0.2
Nutmeg								
Isothermal	205	250	250	6-ft hairpin	Carbowax	40	5	0.5
Linear program	89–240	300	300	5-ft coil	20M (15%)	50	2	0.2
Ginger								
Isothermal	210	250	250	6-ft hairpin	Carbowax	40	5	0.5
Linear program	89–240	300	300	5-ft coil	20M (15%)	50	2	0.2

[a] The inert support in all columns was 60–80 mesh Chromosorb-W.

[b] From Datta *et al.* (1962).

TABLE XXIV IDENTIFICATION OF IMPORTANT COMPONENTS OF SPICE VOLATILE OILS OF DIFFERENT ORIGINS[a]

Spice	Origin of spice	GLC peak number	Infrared identification
Black pepper	Malabar, Lompong	5	β-Pinene
		6	D-Limonene
		16	β-Caryophyllene
Nutmeg	East Indies, West Indies	1	α-Pinene
		3	Camphene
		14	*d*-Terpineol
		24	Myristicin[b]
Ginger	Jamaican, Cochin, and African (Sierra Leene)	4	Camphene
		8	β-Phellandrene
		18	Zingiberene
		19	Zingiberel
Cassia	Saigon, Korintji, and Batavia	1	Benzaldehyde
		Last	Cinnamic aldehyde

[a] From Datta *et al.* (1962).

[b] This material is an unsaturated ether without OH or CO groups. It may be myristicin, but neither reference material nor infrared spectra were available for positive identification.

bowax columns and can be used to identify these products in essential oils of dill, ginger, and spearmint.

Schultz and Mohrmann (1965b) determined the conditions effective for successful gas chromatography of the essential oil of garlic. Constituents found included $(CH_2{:}CHCH_2)_2S_3$, Me_2S, Me_2S_2, $(CH_2{:}CHCH_2)_2S_3$.

Unchanged mercaptans were not present to any extent when the distillate oil was triturated with $HgCl_2$, extracted with CCl_4, taken up in $CHCl_3$, and analyzed by gas chromatography. This was explained as possibly being due to their reaction with $HgCl_2$ to form mercaptides or to thermal decomposition of the oil constituents perhaps catalyzed by the metallic fractometer material. Comparative studies with oils of *Allium cepa* (onion) and *A. ursinum* (ramsoms) showed strong qualitative similarities but very distinctive quantitative differences.

From hydrolytes of horseradish *(Cochlearia armaracia)*, black mustard *(Sinapsis nigra)*, and wasabi *(Wasabia japonica)*, which are used for preparing commercial mixed horseradish (wasabi) powder, the acrid components were collected after steam distillation and solvent extraction by Kojima *et al.* (1966). The acrid components were analyzed by GLC, and twenty components were detected. The main component in horseradish was β-phenylethyl isothiocyanate, but only small amounts of it were found in black mustard and wasabi. The main acrid components in commercial wasabi powder were identified as allyl and β-phenylethyl isothiocyanate; their relative contents could be distinguished in

raw materials used for preparing commercial wasabi powder. This method was applied to several kinds of commercial powders, and it was shown that they were prepared mainly from horseradish. The results suggested that the method described is useful for quality control.

Horose *et al.* (1960) investigated the essential oils of *Juniperus rigida* from Japan, *J. communis* from Italy, and commercial juniper berry oil by fractionation *in vacuo*, gas chromatography, liquid chromatography, and infrared spectroscopy. *Juniperus rigida* oil was composed of *l*-pinene 36.3%, myrcene 12.9%, 3-carene 0.8%, limonene 1.6%, *p*-cymene 0.9%, β-elemene 2.0%, caryophyllene 0.4%, humulene 0.4%, γ-cadinene 0.2%, terpeneniol 0.2%, a mixture of borneol and citronellol 0.2%, a mixture of bornyl acetate, citronellyl acetate, and terpinyl acetate 0.13%, anethole 0.05%, and unknown hydrocarbon 1.1%. *Juniperus communis* and juniper berry oils contained sabinene, camphene, *l*-terpinene, γ-terpinene, terpinolene, *l*-terpineol, ethyl caprylate, and an unknown unsaturated hydrocarbon, b.p. 223° to 250°C, in addition to the ingredients of *J. rigida* oil. Bruno (1961) studied the principal components of *J. sabina* by gas chromatography and successively identified them by suitable sensitive reactions. In *J. sabina*, *d*-sabinene, *l*-pinene, sabinyl acetate, and limonene were identified.

Nigam *et al.* (1963b) combined GLPC and TLC into a single microphysicochemical technique for the simultaneous determination of piperitone and piperitone oxide in *Mentha arvensis* (Japanese mint) and *M. piperita* (Mitcham peppermint). The authors have also indicated future applications of this technique.

Wrolstad and Jennings (1964) investigated, among other essential oils, the oils of sweet marjoram, thuja, and bay as sources of authentic samples of monoterpene hydrocarbons.

Jaureguiberry and Wolff (1962) subjected oil of nutmeg to gas chromatography. Fraction A was pure α-pinene, whereas fraction B was largely β-pinene but apparently contained something else. Fractional distillation did not effect separation, but gas chromatography effected a clean separation of fraction B into β-pinene and (+)-sabinene. Identification of (+)-sabinene was accomplished by ultimate analysis and ultraviolet, infrared, and nuclear magnetic resonance spectra. Curves for the latter two are reproduced and discussed in detail. Lee *et al.* (1962) related organoleptic grades with the chemical composition of eleven commercial samples of nutmeg oil by gas chromatography. At least thirty-three components were found, eight of which were positively identified by infrared spectra and seven others tentatively identified by their relative retention times. On the Barber–Colman DEGS run, in addition to twenty-six components, safrole in 1 to 2% concentration was eluted after 6.5 hours. A high-boiling major component in 5 to 10% concentrations, positively myristicin, was seen in the Apiezon L run. The unacceptable oils had significantly less limonene and linalool than the acceptable oils. Other compounds identified were α-pinene and

α-limonene, camphene, β-pinene, cumene menthone, β-terpineol, terpineol, eugenol A, isoeugenol, cyclamen aldehyde, caryophyllene, and menthyl isovalerate.

Mackay *et al.* (1961) used gas–liquid chromatography with ionization-type detectors to demonstrate differences in the composition of the unconcentrated vapor over good and deteriorated peppermint oil, real and imitation banana flavors, coffee, various grades of brandy, whisky, crushed onions before and after treatment with chlorophyllin, and cigar and cigaret smoke. Some of the compounds in peppermint oil, brandy, and whisky were identified. Lacroix *et al.* (1967) studied the application of electron-capture gas chromatography in the determination of flavor components of onions. Chromatographs of the more volatile fractions and hexane-soluble components of six varieties of onions show qualitative and quantitative differences among them. Onions of one variety obtained from seeds and from sets gave essentially the same chromatograms, indicating that the method of propagation has little effect on the pungent components.

Calzolari *et al.* (1968) conducted a gas chromatographic study of the essential oil of origanum with both capillary and packed columns. In addition, some new components of the essential oil of origanum were found by coupling preparative gas chromatographic analysis and infrared spectroscopy. Stahl *et al.* (1969b) devised both a GLC and a TLC procedure to differentiate the origin of oreganos. The gas chromatographic procedure utilizes the observation that the ratio of the concentration of carvacrol to thymol in Greek oregano is approximately 7 : 1, whereas in Mexican oregano it is 1 : 1. This observation can be reinforced by a fluorescene TLC technique, using a hexane–ethyl acetate–tetrahydrofuran mixture as the developing solvent. A two-dimensional TLC using acetone–ether–methylene chloride for the second development affords a clean-cut separation of all spots and is particularly useful in the diagnosis of mixtures.

Bernhard (1957) reported the separation and identification of some terpenes in pepper oil by GLPC. Hasselstorm *et al.* (1957), after reviewing the available literature on the composition of volatile black pepper oil, reported the detailed composition of Indian (Malabar variety) black pepper (Table XXV). Lewis *et al.* (1969b,c) reported and discussed the differences in yield and composition of pepper oils of seventeen Indian varieties of black pepper (Table XXVI). The available literature is summarized in Table XXVII. Jennings and Wrolstad (1961) reported a comprehensive study of components of commercial black pepper oil by gas chromatography and infrared spectroscopy. Wrolstad and Jennings (1962) studied the changes in the oil by gas chromatography. Two components were labile at relatively low temperatures. Two other components that normally occur in minor amounts increased in quantity on heating and were identified by ultraviolet and infrared spectroscopy as alloocimene isomers. The relative amount of alloocimene in black pepper oil could be a useful quality control criterion, if the method of preparation, etc., is also considered. Light

TABLE XXV COMPONENTS ISOLATED FROM BLACK PEPPER OIL (*PIPER NIGRUM*)[a]

Compound	Formula	B.p. or m.p. (°C)	Estimated oil (%)	Characterization	M.p. (°C)	Analysis					
						Calculated			Found		
						C	H	N	C	H	N
α-Pinene	$C_{10}H_{16}$	157–161	14	Nitrolpiperidide	117	72.00	10.40	11.20	72.31	10.43	11.43
				Myrtenal semicarbazone	206	63.77	8.21	20.29	63.93	8.02	20.30
β-Pinene	$C_{10}H_{16}$	164.5–168	23	Nopinic acid	126–127	65.18	8.75	—	65.41	8.41	—
l-α-Phellandrene	$C_{10}H_{16}$	175–178.5	7	Maleic anhydride adduct	125	71.80	7.69	—	71.72	7.67	—
dl-Limonene	$C_{10}H_{16}$	178.5–181	25	Tetrabromide	124–125	26.34	3.53	—	26.37	3.42	—
β-Caryophyllene	$C_{15}H_{24}$	126.5/20 mm	19	Phenylurethane of caryophyllene alcohol	135–136	77.38	9.15	—	77.64	9.13	—
Epoxydihydrocaryophyllene	$C_{15}H_{24}O$	M.p. 61–62	0.1	Mixed melt with authentic specimen	—	81.80	10.93	—	81.88	10.67	—
Phenylacetic acid	$C_8H_8O_2$	M.p. 76.5–77	0.2	Mixed melt with authentic specimen	—	70.59	5.92	—	70.81	6.04	—
Dihydrocarveol	$C_{10}H_{18}O$	108–124/20 mm	2	3,5-Dinitrobenzoate	120.5–121.5	58.61	5.80	8.05	58.72	5.75	8.34
Piperonal	$C_8H_6O_3$	108–124/20 mm	0.5	Semicarbazone	218–219	52.17	4.38	20.28	52.53	4.16	19.96
Cryptone	$C_9H_{14}O$	108–124/20 mm	0.1	Semicarbazone	185	61.54	8.71	—	61.39	8.79	—
Piperidine	$C_5H_{11}N$	106	0.1	*N*-Phenyl-*N'*,*N* pentanethylene thiourea	99	65.41	7.31	12.72	65.54	6.99	12.92
(Alcohol)	$C_{10}H_{18}O$	103–105/13 mm	0.1	3,5-Dinitrobenzoate	117–117.5	58.62	5.78	8.04	58.54	5.74	8.28
(Alcohol)	$C_{15}H_{26}O$	100/1 mm[b]	0.1	—	—	81.08	11.71	—	81.19	11.47	—
(Alcohol)	$C_{15}H_{24}O$	110/1 mm[b]	0.1	—	—	81.81	10.90	—	81.83	11.08	—
Citronellol (?)	—	—	0.1	Semicarbazone of pyruvic acid ester	—	—	—	—	—	—	—
	$C_{15}H_{32}O_4$	M.p. 97	—	—	—	64.81	11.67	—	64.45	11.91	—
	$C_9H_{16}O_2$	M.p. 161	0.1	—	—	69.23	10.26	—	69.20	10.32	—
	$C_{10}H_{10}O_3$	M.p. 234–235	0.1	—	—	74.07	6.17	—	73.64	5.87	—
Higher boiling components	—	—	4.0	—	—	—	—	—	—	—	—
Residue	—	—	2.3	—	—	—	—	—	—	—	—

[a] From Hasselstrom *et al.* (1957).

[b] Bath temperatures.

TABLE XXVI VARIETAL DIFFERENCES IN YIELD AND PHYSICOCHEMICAL COMPOSITION OF PEPPER OIL[a]

Pepper variety	Oil yield (% v/w)	Ref. index, (25°C)	Specific gravity, 30°C	Optical rotation, 25°C	Constituents (%)									
					α-Pinene	Camphene	β-Pinene	Sabinene + myrcene	Limonene	p-Cymene	Bergamotene	Caryophyllene	Humulene	Selinene
Panniyur-1	2.48	1.4790	0.9082	−13.0	5.9	—	10.6	42.4	22.0	2.2	—	16.8	—	—
Karimunda	2.56	1.4791	0.9246	−15.0	5.3	Trace	15.8	25.5	25.7	1.8	—	21.0	2.7	—
Karivally	2.28	1.4777	0.9226	−12.6	10.6	Trace	15.5	30.8	26.4	1.7	—	15.0	Trace	Trace
Kottanadan	2.48	1.4789	0.9175	−14.0	14.9	Trace	21.5	14.8	26.1	Trace	Trace	17.6	Trace	—
Chumala	2.40	1.4800	0.9021	−24.0	8.8	—	31.9	Trace	25.8	—	Trace	33.3	—	—
Vally	2.00	1.4810	0.9098	−8.4	7.1	—	9.5	28.9	24.5	4.3	Trace	25.5	Trace	—
Kumbhakodi	2.40	1.4771	0.9004	−27.6	11.1	—	35.2	9.5	24.1	1.8	Trace	18.0	Trace	—
Kalluvally	3.08	1.4770	0.8995	−17.8	8.5	—	24.8	27.3	18.6	—	—	20.7	—	1.7
Karinkotta	3.00	1.4781	0.8895	−15.2	6.3	Trace	16.7	24.9	24.9	3.8	—	20.7	—	1.9
Balankotta	2.60	1.4765	0.8806	−3.0	12.8	—	11.6	24.3	24.5	2.3	—	22.4	—	—
Perumkodi	3.80	1.4771	0.8837	−23.4	9.3	—	26.9	17.1	19.1	Trace	0.6	27.0	—	—
Kuthiruvally	3.20	1.4750	0.9213	−26.0	10.7	—	35.5	7.1	31.1	Trace	1.0	10.9	2.1	—
Mumdi	3.10	1.4771	0.8672	−25.6	10.1	—	32.9	Trace	26.4	—	—	30.5	—	—
Narayakodi	3.20	1.4782	0.9066	−22.0	6.8	—	29.0	14.7	29.0	—	5.3	10.3	4.9	—
Arikottanadan	3.52	1.4750	0.9187	−13.6	19.4	—	33.6	Trace	27.3	—	1.0	17.5	3.7	—
Uthirankotta	2.70	1.4795	0.9153	−18.0	9.0	—	25.2	Trace	39.8	4.0	2.6	17.0	2.4	—
Karimunda-Thodupuzha	3.00	1.4773	0.8783	−15.0	4.5	—	22.5	27.5	23.4	1.2	—	19.0	—	1.9

[a] From Lewis *et al.* (1969b,c).

TABLE XXVII COMPOSITION OF PEPPER OIL AS REPORTED IN THE LITERATURE

Constituent	Percentage reported by					
	Hasselstorm *et al.* (1957)	Jennings and Wrolstad (1961)	Ikeda *et al.* (1962)	Nigam and Handa (1964)	Wrolstad and Jennings (1965)	Lewis *et al.* (1969b)[a]
α-Pinene	14	11.6	5.3	7.4	22.1	4.5–19.4
β-Pinene	23	13.7	7.8	4.8	11.1	9.5–35.5
Camphene	—	—	0.5	0.6	—	Trace
Sabinene	—	—	11.6	15.7	21.3	Trace–42.4
Myrcene	—	—	3.7	10.3	2.6	
Limonene	25	26.3	12.3	22.2	11.1	18.6–39.8
α-Terpinene	—	—	0.17	—	—	—
γ-Terpinene	—	—	0.05	0.8	0.85	—
p-Cymene	—	—	0.74	1.5	0.85	Trace–4.0
α-Thujene	—	—	1.7	—	2.6	—
α-Carene	—	—	8.6	—	10.2	—
Ocimene	—	—	0.05	—	—	—
α-Phellandrene	—	—	2.1	5.3	1.7	—
β-Phellandrene	7	—	2.4	—	1.7	—
α-Bergamotene	—	—	—	3.7	—	Trace–5.3
β-Caryophyllene	19	13.3	—	18.1	16.6	10.5–33.5
α-Humulene	—	—	—	6.0	—	2.1–4.9
α-Selinene	—	—	—	—	—	Trace–1.9
Epoxydihydro-caryophyllene	0.1	—	—	—	—	—
Phenylacetic acid	0.2	—	—	—	—	—
Dihydrocarveol	2	—	—	—	—	—
Piperonal	0.5	—	—	—	—	—
Cryptone	0.1	—	—	—	—	—
Piperidine	0.1	0.4	—	—	—	—
Citronellol	0.1	—	—	—	—	—
Alcohol ($C_{14}H_{12}O$)	0.1	—	—	—	—	—

[a] Based on variations in seventeen varieties of Indian black pepper.

causes certain photochemical changes, such as a decrease in at least one fraction and an increase in another. Datta and Susi (1962) reported the gas chromatographic separation of oxygen-containing terpene compounds on low-temperature columns. Wrolstad and Jennings (1965) separated monoterpene hydrocarbons of black pepper both by vacuum distillation and by TLC and GLC followed by infrared and ultraviolet spectroscopy of the products, which revealed the following: α-pinene 26%, α-thujene 3%, β-pinene 13%, sabinene 3%, carene 12%, myrcene 3%, α-phellandrene 2%, limonene 13%, β-phellandrene 2%, γ-terpinene 1%, *p*-cymene 1%, and terpinolene 1%. Wrolstad (1965) also re-

ported the terpene hydrocarbons of *Piper nigrum*. Muller and Jennings (1967) isolated individual sesquiterpene hydrocarbons from black pepper oil by column chromatography followed by preparative and capillary gas chromatography. On the basis of matching retentions and of infrared and, where applicable, ultraviolet spectra of the isolated components, the following were identified as major compounds of the sesquiterpene hydrocarbon fraction: δ-elemene, α-copaene, β-elemene, α-*cis*-bergamotene, α-santalene, α-*trans*-bergamotene, β-caryophyllene, α-humulene, β-selinene, α-selinene, β-bisabolene, δ-cadinene, and calamenene. Muller *et al.* (1968) further studied the volatile compounds of black pepper by techniques involving infrared spectroscopy of components isolated and purified by repetitive gas chromatographic separations. α-Cubene, isocaryophyllene, and γ-muurolens were identified among the minor constituents, and an additional major constituent was isolated and identified as β-farnesene. Mechanisms that explain the observed co-occurrence of sesquiterpenes in natural products are discussed. Pruthi (1968a), while reviewing the chemistry and quality evaluation of black pepper, briefly covered its GLPC aspects also. Buttery *et al.* (1969) studied the aroma component of bell pepper.

Porcaro and Johnston (1961) reported a rapid method for the determination of menthol isomers on two columns with two substrate types. The packings used were Carbowax 20M and silicon oil DC-710 on Chromosorb-W. Examples are given of the chromatograms obtained, and an application to the analysis of peppermint oil is also given. Smith and Levi (1961) analyzed *Mentha piperita* and *M. arvensis* by GLPC (Figs. 9A and 9B). The composition and criteria thus established were utilized for (1) determination of geographical origins, (2) recognition of biochemical relationships governing formation of oil in the plant, (3) evaluation of the manufacturing process, and (4) detection of subtle adulterations. Data and their treatment should prove of value not only to processors of essential oils, but also in characterization, analysis, and quality control of complex natural and synthetic compositions produced by the food, drug, and cosmetic industries. Hefendehl (1962) discussed the theoretical and practical aspects of gas chromatography of essential oils and of peppermint oil in particular. A description is given of possible methods for identifying individual peaks of a diagram. The bases of preparatory gas chromatography applied to essential oils are explained. Also included are a definitive treatment of the possible decomposition of labile oil components due to the effect of temperature, carrying materials, and carrying gas (H), a quantitative evaluation of the oil diagram, and an evaluation of gas chromatography with relation to an understanding of the biogenesis of the components of the essential oil.

Jansen and Vander Hold (1963) studied the GLC of rue oil and peppermint oil and identified the characteristic peaks for rue oil. Peppermint oil was more difficult to identify, but gave two distinctive peaks. Qualitative data were obtained for rue oil, but not for peppermint oil. Von Rudloff and Hefendehl (1966)

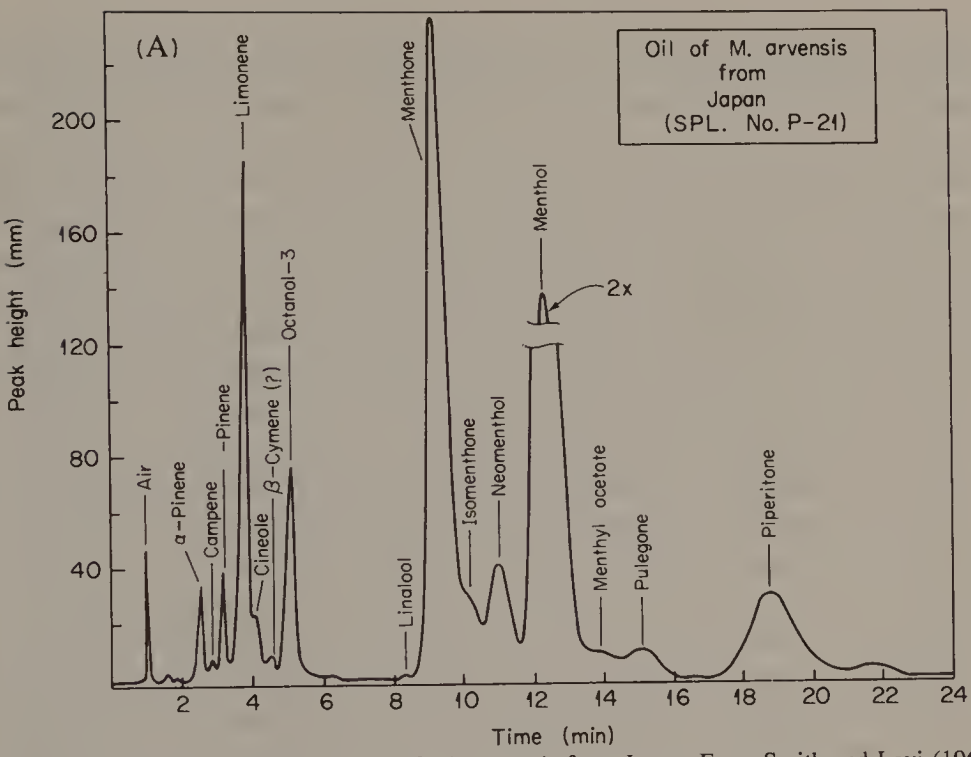


FIG. 9A. Gas chromatogram of oil of *M. arvensis* from Japan. From Smith and Levi (1961).

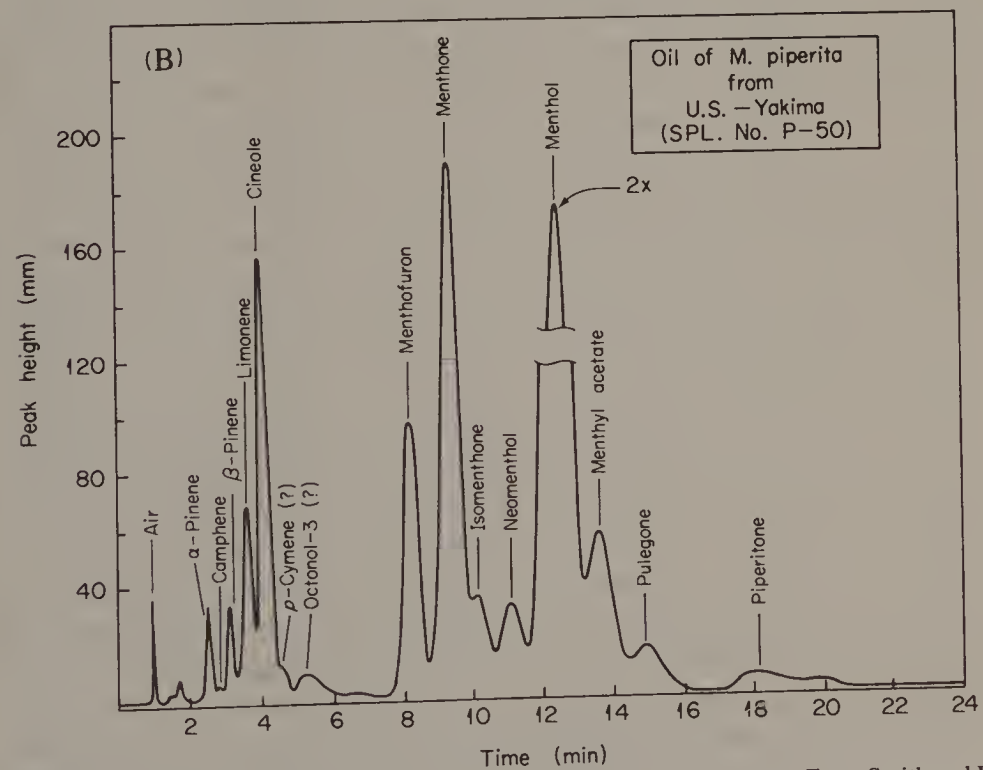


FIG. 9B. Gas chromatograms of oil of *M. piperita* from the United States. From Smith and Levi (1961).

found the volatile oil of the North American wild mint to consist mainly of *d*-pulegone (80 to 90%) and smaller amounts of α-pinene, β-pinene, sabinene, limonene, 1,8-cineol, 1-octen-3-ol, menthone, isomenthone, piperitone, *cis*- and *trans*-pulegone oxide, and piperitenone. Trace amounts of γ-terpinene, menthofuran, β-carophyllene, and α- and δ-cadinene were also isolated, and camphene, *p*-cymene, terpinolene, sabinene hydrate, isopulegone or its stereoisomer, β-elemene, and α-terpineol were tentatively identified. A labile autoxidation product of pulegone was also detected. Significant seasonal changes in the quantitative oil composition occurred only in very young plants. Maximum oil yield was obtained at the start of flowering. Practically no variation in the oil composition was recorded for plants from different localities. Goryaev *et al.* (1967) advocated that the most reliable method for the evaluation of mint essential oil is GLC. All components of the oil can be determined quantitatively by this method. Polyethylene glycol of molecular weight 2000 is the best liquid stationary phase to affect the optimal resolution. A 255 × 0.8-cm steel column is used for the analysis, which is performed at 158° to 159°C with nitrogen as carrier gas and with a flame ionization detector. The major constituents are menthol and menthyl acetate. The content of menthol as determined by a chemical method is 43.12%, and by GLC 44.50%. The content of methyl acetate determined chemically is 25.80%, and with gas chromatography it is 22.40%. Other components are menthone, isomenthone, neomethone, pulegone, and piperitone. Calibration graphs and fenchone as an internal standard are used for calculation of peak areas. Hefendehl *et al.* (1967) incorporated radioactive acetate and mevalonate into the volatile oil of *Mentha piperita* and *M. arvensis*. All terpenes studied, including menthol and methofuran, showed radioactivity within 5 minutes of feeding $^{14}CO_2$. Isolation of individual monoterpenes by GLC and determination of their specific activities upon supplying $^{14}CO_2$ for various periods of metabolism confirmed the Reitsema biosynthetic sequence (1954) in its major aspects, but it could not be confirmed that piperitenone is the first isolable precursor of the two sequences.

Nigam and Levi (1963) reported the occurrence of a peak emerging between 3-octanol and menthone in spearmint oil gas chromatogram, which permitted a reliable distinction to be made between *Mentha spicata* and *M. cardiaca* largely by two components—*trans*-sabinene hydrate and *trans*-sabinene acetate. The tertiary alcohol is known to occur in American peppermint oil (0.8%). The presence of the ester in *M. piperita* was also established for the first time. Smith *et al.* (1963) analyzed spearmint oils by GLPC. The compositional criteria established were used for the determination of geographical origin and the recognition of genetic variations among plants. Over 1% of menthone was found in American oils derived from *M. cardiaca*, but less than 0.5% of the 3-oxygenated terpene was detected in oils distilled from *M. spicata*. Likewise, an unidentified component occurring in *M. spicata* to the extent of about 1%, but present only in

traces in *M. cardiaca* oils, permitted reliable distinctions. Experimental results for canone contents, illustrative of the selectivity of the gas chromatographic technique, are compared and correlated with those obtained by conventional ultraviolet analysis.

Niegisch and Stahl (1956) studied the gaseous emanation products of onion. Carson and Wong (1961a) isolated a number of the more volatile flavor components of onions by GLPC and identified them by infrared and chemical derivatization. In particular, methyl disulfide, methyl trisulfide, methyl-*n*-propyl disulfide, methyl-*n*-propyl trisulfide, *n*-propyl disulfide, and *n*-propyl trisulfide were identified (Fig. 10). Neither monosulfide nor allylic disulfides could be detected.

Coffman and Schwecke (1962) conducted quantitative and qualitative analyses for piperonal, vanillin, and ethyl vanillin in food-flavoring powders by gas chromatography. Jackson (1966) concentrated the trace components in vanilla extracts and analyzed them by gas chromatography. This technique permits the detection of more of the volatile components than was previously possible. Levins and Ottenstein (1967) studied the effect of copper, aluminum, stainless steel, and glass tubing on absorption of polyol, and vanillin was tested with columns packed with 5% Carbowax 20M or 5% FFAP on Chromosorb GAW-DMCS at a column temperature programmed at 4.0°/min from 130° to 230°C for Carbowax and 4.0°/min from 130° to 260°C for Chromosorb. Stainless steel and aluminum tubing adsorb the samples strongly, whereas copper and glass do not. More than one type of adsorption site appears to be involved, but coating the internal wall of the metal tubing with a thin film of a polar stationary phase essentially eliminates adsorption losses by blocking active sites on the metal surface. Mendez (1968), employing gas–liquid chromatography, separated five phenols—catechal, pyrogallol, phloroglucinol, vanillin, and syringaldehyde—with average retention times of 5.0, 7.0, 7.8, 3.6, and 8.2 minutes, respectively. Helium carrier gas and helium flame ionization detection were used. Well-defined peaks were obtained for the five naturally occurring phenols.

Guenther *et al.* (1959, 1961, 1963, 1965, 1966, 1967, 1969, 1971, 1973, 1975, 1977) reviewed the GLC of vanilla extract and related analytical techniques.

Lawrence *et al.* (1969a) investigated the oil of galangal *(Alpinia officinarum)* by GLPC of fractions obtained by vacuum distillation followed by alumina and $AgNO_3$ alumina column chromatography. The presence of α-pinene and cineol reported by previous workers was confirmed. In addition, the oil was found to contain forty-six previously unreported constituents.

By employing column and gas chromatography, Nigam and Levi (1963) identified some hitherto unreported constituents in wild ginger oil *(Zingiber zerumbet)*.

Brodnitz *et al.* (1971) found diallyl thiosulfinate to be a major constituent of solvent-extracted garlic. Upon gas chromatography, diallyl thiosulfinate undergoes dehydration, leading to the formation of two isomeric disulfides. At room

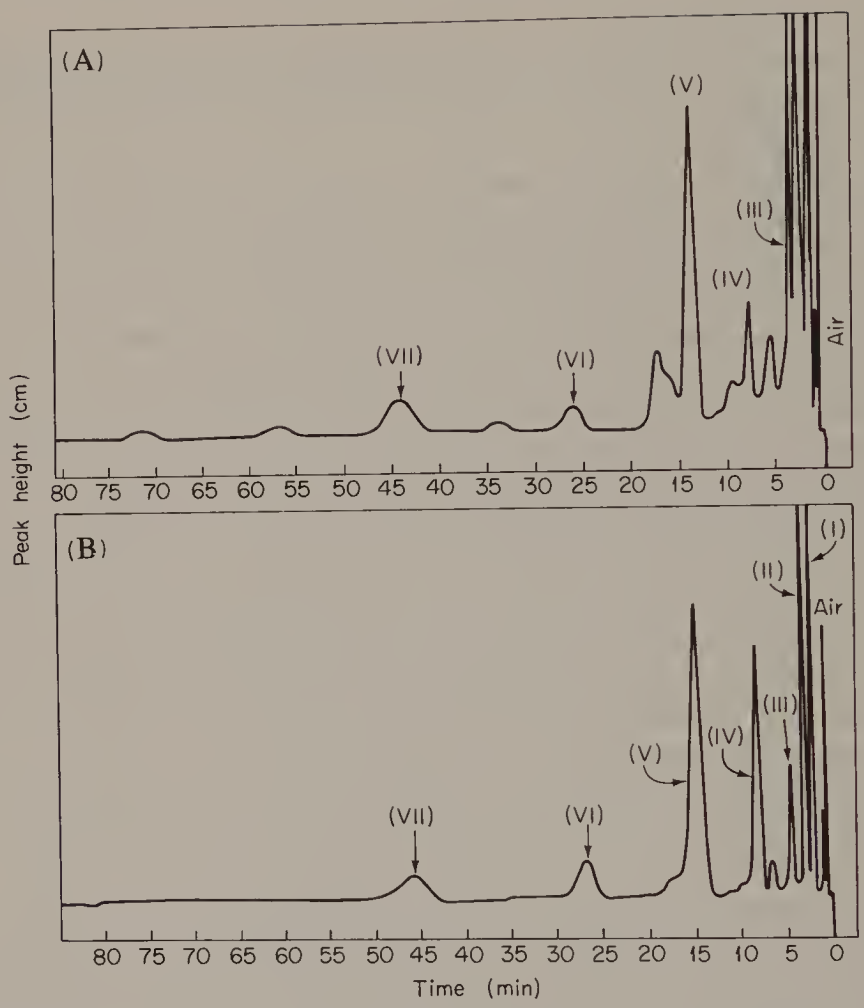


FIG. 10. Gas–liquid partition chromatograms of onion volatiles. (A) Isopentane extraction, Carbowax-1540 column. (B) Carbon adsorption, Reoplex 400 column, 1/4 inch × 6 feet; temperature 150°C, flow rate 45 ml of helium per minute; filament current 225 mA, 4-mV full-scale deflection. Sample volume 5 μl. Peak identification: (I) Ethyl and isopropyl alcohols. (II) *N*-Propyl alcohol. (III) Methyl disulfide. (IV) Methyl-*N*-propyl disulfide. (V) *n-Propyl disulfide and methyl trisulfide. (VI) Methyl-n*-propyl trisulfide. (VII) *n*-Propyl trisulfide. From Carson and Wong (1961a).

temperature, it undergoes a rearrangement. After 24 hours, sulfur dioxide and diallyl mono-, di-, and trisulfides are the major products of this reaction. The role of thiosulfinates in the formation of flavor components of garlic and other alliums is discussed.

By using infrared, ultraviolet, NMR, mass spectra, etc., Varo and Heinz (1970b) isolated, identified, and characterized 1,4-*p*-menthadien-7-al from cumin seeds.

Several other workers have reported the volatile flavor components of spices. Thus, Bandyopadhyay *et al.* (1973) reported comparative studies on the chemical

aspects of white and Red Globe onions by using GLC, TLC, ultraviolet, and infrared spectroscopy. Galetto and Bednarzyk (1975) tried to correlate more accurately onion oil flavor with chemical composition using multiple regression analysis techniques (quantitative GLC data) with organoleptic evaluation. The major onion compounds of overall onion flavor significance are represented by peaks 1, 5, and 7 (methyl propyl disulfide, methyl propyl trisulfide, and dipropyl trisulfide, respectively).

Freeman (1975) studied the distribution of flavor components in garlic, leek, and onion. Freeman and Whenham (1975b) made a survey of volatile components of *Allium* species and cultivars, mostly edible, in terms of *S*-alkyl-1-cystein sulfonides present as flavor precursors. Freeman and Whenham (1975a) studied the losses of onion flavor components during processing, cooking, dehydration, and freezing. The mechanism by which these losses occur has been investigated on the basis of the allinase-precursor system and has been shown to include (1) complete or partial enzyme destruction; (2) partial nonenzymatic destruction of precursors; and (3) partial enzymatic hydrolysis of precursors with loss of volatile reaction products.

The occurrence of *N*-isobutyldeca-*trans*-4-dienamide (Dhar and Atal, 1967) and sesamin in *Piper longum* has been reported (Atal *et al.*, 1966). Gupta *et al.* (1972) have reported the occurrence of methyl piperate in *P. officinarum*. Russell and Else (1975) have reported the volatile compositional differences between cultivars of black pepper, and Debrawere and Verzele (1975) have reported new constituents of the oxygenated fraction of pepper essential oil.

Gough and Goodhead (1975), using GLC and mass spectrometry (MS), have reported the occurrence of volatile nitrosamines in spice curing salt premixes for meat products.

Hunter *et al.* (1975) reported the combined GC–MS analysis of tamarind volatiles, which has resulted in the identification and confirmation of sixty-one major components including among others, pyrazines and thiazoles, which are formed normally during roasting of several foods. The aroma profile of tamarind appears to be similar to that of citrus notes, warm spice-like flavors, and flavors of some roasted foods.

Earlier, Misra and Chandra (1973) discussed the chemistry and manufacture of vanillin. The Analytical Methods Committee (1973) discussed GLC of essential oils.

10. Proton Magnetic Resonance

Wallace *et al.* (1963) conducted proton magnetic resonance studies on lignans from nutmeg hydroxyotobain and isootobain and described their structures. They also obtained the conformations of the molecules and the configurations of their asymmetric centers.

11. X-Ray Diffraction

Meranger *et al.* (1967) reproduced x-ray diffraction patterns for semicarbazone of β-ionone, *p*HCHO, tridecanone, citronellal, acetophenone, carvone, methylheptenone, vanillin, and anisaldehyde, and tabulated the *d*-spacings. In most cases, positive identification of the derivatives can be made by the position of the strongest line. With camphor, vanillin, and heliotropin, however, more than one line should be used for identification.

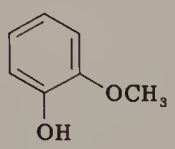
12. Raman Spectra

Mohan (1962) vacuum-distilled the essential oils of cassia, cinnamon, caraway, and spearmint twice, and immediately used the colorless distillates for the determination of their Raman spectra with an E 6122 prism glass spectrograph with a Hilger source unit and Agfa Isochrome plates. The frequencies of the oils and their known major constituents are given in five tables. The Raman effect enables one to identify the principal constituents of the oils but not the minor ones.

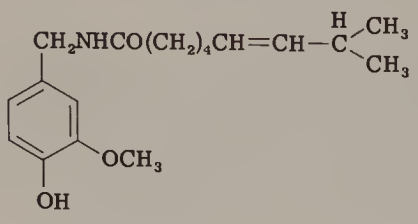
B. Determination of Some Major Principles

1. Pungent Principles

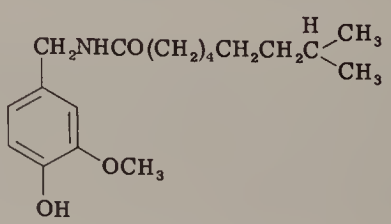
Several pungent compounds found in nature are derivatives of *o*-methoxyphenol:



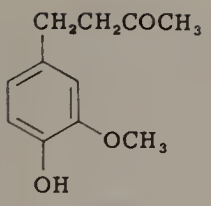
Examples of this group are (1) capsaicin and dihydrocapsaicin, which are constituents of the red pepper spice; (2) zingerone, shogaol, and gingerol, which are constituents of the ginger root; (3) eugenol, found in cloves; and (4) isoeugenol, found in nutmeg and various essential oils (Kulka, 1967).



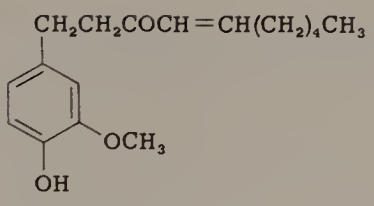
(Capsaicin)



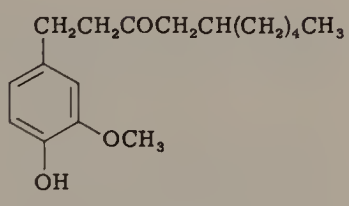
(Dihydrocapsaicin)



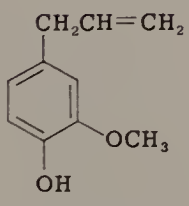
(Zingerone)



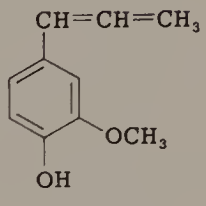
(Shogael)



(Gingerol)



(Eugenol)



(Isoeugenol)

The degree of pungency and the character of taste sensation vary markedly among these chemicals. In certain cases, chemicals having a taste sensation are also odor contributors. Eugenol, in addition to its familiar burning sensation, also has odor, which stimulates the odor preceptors in a positive manner. In contrast, piperine and capsaicinoid are *odorless*. Piperine is responsible for the characteristic "bite" sensation. Since black pepper is the most popular and the most widely used spice in the world, piperine has been investigated in great deatil, particularly when black pepper was in short supply, to find a commercially feasible synthesis or a replacement. The chemistry of piperine and other pungent principles such as chavicine and piperettine and analytical techniques for the determination of pungent principles (piperine, etc.) have been reviewed (Rogers, 1966; Pruthi, 1969b,c,d, 1970a,b).

The analytical techniques for some of the major pungent principles in some spices are briefly reviewed and discussed below.

a. Pungent Principles in Black and White Pepper. Labruyere (1966) and subsequently, Pruthi (1969b,c,d, 1970a,b) have presented critical exhaustive reviews on the working principle, specificity, sensitivity, and comparative merits and demerits of thirteen analytical techniques for the determination of pungent principles (piperine, etc.) in black and white pepper (Table XXVIII).

The advantages and disadvantages of different techniques are briefly discussed below.

The nitrogen method is not specific, as is indicated frequently in the literature (Fagen *et al.*, 1955; Tausig *et al.*, 1956; Genest *et al.*, 1963), because the related nonvolatile, ether-soluble, nitrogen-containing compounds interfere in the estimation. The incomplete decomposition of piperine may also affect the results.

TABLE XXVIII ANALYTICAL TECHNIQUES FOR THE DETERMINATION OF PIPERINE (PUNGENCY) IN PEPPER[a]

Methods	References
Chemical methods	
1. Kjeldahl's nitrogen estimation	Winton and Winton (1945)
2. Iodometric method	Duro (1961)
3. Alkali hydrolysis method	Labruyere (1966)
Physicochemical methods—instrumental methods	
Colorimetric methods	
1. Chromotropic acid method (American Spice Trade Association, 1968)	Bricker and Johnson (1945), Bricker and Veil (1950), Beroza (1954), and Lee (1956)
2. Reaction with ammonium reineckate	Kum-Tat (1961)
3. Nitric acid method	Graham (1965a)
4. Komaroswsky reaction using thiourea	Graham (1965b)
5. Reaction with Labat reagent	Graham (1965c)
6. Phosphoric acid method	Graham (1965d)
7. Reaction of piperidine with CS_2	Shankaranaryana *et al.* (1970)
Ultraviolet spectrophotometric methods	Fagen *et al.* (1955), Tausig *et al.* (1956), Ramachandra Rao *et al.* (1960), Genest *et al.* (1963)
Other instrumental methods	
1. Polarographic method	Hanc and Santavy (1946)
2. Infrared analysis method	Pleat *et al.* (1952)
3. Gas chromatographic method	Parker *et al.* (1963)

[a] From Pruthi (1969a-d, 1970a).

Unsaturated fats and some components of essential oils may influence the results of polarographic (Hanc and Santavy, 1946, 1947; Fagen *et al.*, 1955) and iodometric methods (Duro, 1961) unfavorably (Labruyere, 1966).

The ultraviolet spectrophotometric method (Fagen *et al.*, 1955; Tausig *et al.*, 1956; Ramachandra Rao *et al.*, 1960; Genest *et al.*, 1963) has a high specificity for piperine, owing to the typical system of conjugated double bonds, and is the best existing method for the determination of piperine. Besides, piperettine can also be determined by this method. In the author's view it could serve as a reference method for the determination of piperine.

The chromotropic method (American Spice Trade Association, 1968) uses a reaction given by all molecules that release formaldehyde under the conditions of the analysis, not merely compounds with a methylenedioxy grouping such as piperic acid and piperonal, but also products such as dextrose (Sawicki *et al.*, 1962). Still other carbohydrates may also interfere.

All compounds with a benzene nucleus that can be nitrated may interfere with the colorimetric method (Graham, 1965a) using nitric acid.

The colorimetric method (Graham, 1965b) using thiourea in H_2SO_4 and an aromatic aldehyde seems inferior to the polarographic and iodometric methods, as unsaturated compounds formed by concentrated sulfuric acid may interfere. Thus, the specificity of the method does not appear to be satisfactory.

The colorimetric method (Graham, 1965c) using phosphoric acid, however, is very promising. It is rather specific for piperine, is fast, allows the direct determination of piperine in pepper, and needs no special apparatus other than a visible-range spectrophotometer or an ordinary photoelectric colorimeter.

If an ultraviolet spectrophotometer is not available in any laboratory, only the colorimetric methods (Kum-Tat, 1961; Graham, 1965a,c) may be acceptable as alternative methods for the determination of piperine. The chromotropic acid method usually gives reproducible results. Labruyere (1966) also confirms this viewpoint. According to Genest *et al.* (1963), results obtained are higher in comparison with spectrophotometric values. In applying the first four colorimetric methods to three samples of pepper, Graham (1965d) found that all four colorimetric methods gave the same high results in comparison with the ultraviolet method. Considering the high specificity of the phosphoric method and the low specificity of the method involving the Komarowsky reaction (Graham, 1965b), this result is surprising. Graham (1965b) seems to hold the opinion that discrepancy between the ultraviolet method and the colorimetric methods is largely due to the presence of piperettine. The work of Genest and co-workers (1963), however, shows that the "apparent percentage piperine," calculated from the absorbance at 343 μm and without considering the absorbance at 364 μm, includes the greater part of the piperettine. Therefore, the high results of the colorimetric methods are only to a very small extent due to the presence of piperettine. The real cause must yet be determined. This aspect needs further careful study.

It may also be stated that a method that gives higher results is not necessarily to be rejected if one aims to assess the intrinsic value of the product as a spice in terms of its total pungency. In pepper and in its derived products, compounds other than piperine may also contribute to pungency. Besides, the vinyl homolog piperettine and the stereoisomer chavicine have been detected in pepper. Part of the results of Genest and co-workers (1963) point to the presence of methylene caffeic acid and piperidide, although other results are also conflicting.

The literature reveals many cases of dimerization of ethylenic compounds to cyclobutane derivatives and reversed depolymerization of cyclobutane compounds to ethylenic derivatives under the influence of light and heat (Bernstein and Quimby, 1943; Ruber, 1913; Roberts and Sharts, 1962). The investigations of Staudinger and Muller (1923) justify the supposition that these compounds, if present, contribute to the pungency of pepper. Possibly, some of these substances are determined by the methods of Kum-Tat (1961) and Graham (1965a,b), but they are not, or not completely, determined by the methods of the American Spice

Trade Association (1968) and Graham (1965c), owing to the higher specificity of the latter methods for piperine.

The "hydrolysis method" for the determination of piperine and other bite principles of pepper and pepper oleoresins attacks the amidic bond of the piperidine ring, which is essential for the pungency, in contrast with the methylenedioxy grouping and the aliphatic double bonds (Hasselstorm *et al.*, 1957; Staudinger and Muller, 1923). The point of attack differs from that of all other methods. Possible interferences need not overlap those of existing procedures. For instance, piperonal, which may cause high results with both the chromotropic acid and nitric acid methods, does not interfere. Piperic acid, which interferes with all colorimetric methods, does not influence the hydrolysis method. Thus, the ultraviolet spectrophotometric method, the chromotropic acid method, the nitric acid method, the phosphoric acid method, and the hydrolysis method can furnish complementary data.

Pruthi (1970b) reported the results of an international collaborative study conducted by workers in India, France, The Netherlands, and the United Kingdom on the performance of two ultraviolet spectrophotometric methods, the chromotropic acid method (American Spice Trade Association, 1968), and the alkali hydrolysis method (Labruyere, 1966), and made several useful suggestions for further research on the development of a simple, rapid, reliable, and inexpensive method.

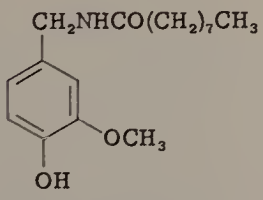
Ananthakrishnan and Govindarajan (1975) have adopted Labruyere's alkaline hydrolysis method for estimating total pungent principles for use directly on pepper powder. This simple method is more rapid, as it obviates the time-consuming step of oleoresin preparation required by all other methods. Besides, this improved method does not require any special equipment.

b. Capsaicin in Capsicums. It was in 1876 that Thresh (1876, 1878) isolated for the first time the pungent principle from capsicums and called it capsaicin. Starting their work on the structure of capsaicin, Nelson (1919) and Nelson and Dawson (1923) declared it to be the amide of vanillylamine and isodecanoic acid. Further work on the chemistry of capsaicin has been reviewed by Newman (1953), Rogers (1966), and Pruthi (1970c).

Kosuge *et al.* (1965) established that the pungent principle of red pepper consists not of one chemical but actually of the unsaturated and saturated amides—that is, capsaicin and dihydrocapsaicin. The mixture of these two amides was named *capsaicinoid,* which is odorless.

With regard to the pungency of this group of compounds, it can be stated that an aromatic ring having a phenolic hydroxyl group and an ether group such as methoxy in ortho position to each other is a basic prerequisite. A side chain is also necessary. The length and composition of this side chain are important. The pungency is greatly enhanced by an acid amide group, in this instance vanillylamide, as found in the capsaicinoid molecule (Kulka, 1967).

On the basis of these observations, the synthetic compound nonoylvanillylamide was prepared. This has considerable pungency and heat (Kulka, 1967).



Nonoylvanillylamide

Friedrich and Rangoonwala (1965) studied the separation of capsaicin and nonanoic acid vanillylamide. Kosuge and Furuta (1970) further studied the pungent principles, which consist of capsaicin, dihydrocapsaicin, nordihydrocapsaicin, homodihydrocapsaicin, and two or more analogs of these materials. Thin-layer chromatography and open tubular gas chromatography showed that the natural pungent mixture contains no cis isomer of capsaicin. The chemical structure of nordihydrocapsaicin was determined as *N*-(4-hydroxy-3-methoxybenzyl)-7-methyloctanamide by gas chromatography, infrared spectrometry, mass spectrometry, and NMR spectroscopy. Homodihydrocapsaicin was identified as *N*-(4-hydroxy-3-methoxybenzyl)-9-methyldecamide.

A number of analytical techniques have been reported from time to time since 1912 for assaying the pungency or capsaicin (Joint Committee of the Pharmaceutical Society, and the Society for Analytical Chemistry, 1959, 1964). The chronological developments in analytical techniques are summarized in Table XXIX, along with their working principle and remarks. The thirty-five methods suggested have been categorized as organoleptic, photometric, spectrophotometric, polarographic titration, paper chromatographic, thin-layer chromatographic, and GLC techniques (Table XXX). See also Barnyai and Szabolis (1976).

The pungency or capsaicin content of capsicums varies widely among varieties, seasons, places of origin, etc. The available data on the capsaicin content of paprikas, peppers, and chilies grown in different parts of the world (Africa, Ethiopia, Hungary, India, Japan, Mexico, Turkey, Uganda, the United States, and other countries) have been summarized in Table XXXI. Thus, the capsaicin content varies from 0.0 in sweet peppers to as high as 1.86% in Indian chilies, which is the highest on record. See also Balbag *et al.* (1968).

André and Mile (1975) have developed a new, simple, and sensitive TLC method for the determination of capsaicin, even at levels as low as 0.1 μg of capsaicin per spot. This method can readily be used to distinguish the paprika varieties according to their degree of pungency.

c. Pungency in Celery. The factor that produces the burning and numbing taste fraction in celery was isolated by Pan (1960) as a slightly oily liquid. Chem-

TABLE XXIX ANALYTICAL TECHNIQUES FOR THE DETERMINATION OF CAPSAICIN

Author	Method	Remarks
Scoville (1912)	Scoville organoleptic test—based on threshold sensory evaluation	Unsatisfactory. Results not reproducible.
Gibbs (1927)	Photometric method	Needs improvement.
von Fodor (1930, 1931)	$VOCl_3$ method (vanadium oxytrichloride)	Color must be determined within 20–30 seconds; otherwise interference by other color reactions takes place.
Tice (1932)	$VOCl_3$ method	
Berry and Samways (1937)	Threshold sensory evaluation	Low sensitivity. Lack of reproducibility.
Hayden and Jordan (1941)	Threshold sensory evaluation	Results not reproducible.
Nogrady (1943)	Titration with picric acid and fluorescence desorption	
Buchi and Hippenmeier (1948)	Phosphomolybdic acid method	Colorimetric method. Time-consuming.
North (1949)	Phosphostungstic–phosphomolybdic acid–vanillin method (Folin–Denis reagent)	Colorimetric method.
Newman (1953)	On Scoville test	Defects of Scoville test pointed out.
Fujita *et al.* (1954)	Paper partition chromatography and colorimetric method	Simple technique.
Schenk (1954)	Ammonium vanadate and HCl method	Photometric method.
Schulte and Krueger (1955)	Diazobenzene sulfonic acid method	Colorimetric method. A satisfactory method.
Spanyar *et al.* (1956a)	Sulfanilic acid method	Authors surveyed the difficulties in earlier method. This method gives considerably accurate and reproducible results.
Spanyar *et al.* (1956b)	Polarimetric titration with *p*-diazobenzene sulfonic acid	Applicable to samples containing capsaicin more than 30 mg/100 g and moisture not above 20%. Mean error less than ±10%.
Schenk (1957)	$VOCl_3$ method	Optical inspection and color comparison. Suitable for rough assessments in pharmacy. A good review of earlier methods presented.
Spanyar *et al.* (1957)	Diazosulfanilic acid method	Applicable to samples containing capsaicin above 5 mg/100 g and moisture not above 20%. Accuracy ± 10%.
Schulte and Krueger (1957)	Photometric method	
Suzuki *et al.* (1957)	Ultraviolet spectrophotometric method	

Waldi (1958)	Paper chromatographic method	The fat extracted from paprika causes elongation of spots and tailing.
Spanyar *et al.* (1958)	Sulfanilic acid method, modified	A quick method; rough results obtainable within 4 minutes.
Zitko (1957)	Same as above	
Benedek (1959a)	Ammonium vanadate photometric method	Sensitivity: 0.01 mg of capsaicin.
Spanyar *et al.* (1958)	Photometric method	Satisfactory.
Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry (1959)	Column chromatographic purification and spectrometry	A review and improvement of available methods discussed.
Csedo *et al.* (1960)	Review of analytical methods	Authors recommend Spanyar method.
Tiechert *et al.* (1961)	Thin-layer chromatography	Satisfactory.
Brauer and Schoon (1962)	Ultraviolet spectrophotometric method	Satisfactory but time-consuming, involving costly equipment.
Kosuge and Inagaki (1962)	Phosphomolybdate method and colorimetric method	
Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry (1964)	Spectrophotometric and modified colorimetric methods	Spectrophotometric and colorimetric methods, recommended.
Heusser (1964)	Thin-layer chromatographic method	Applicable to samples containing at least 0.1 mg of capsaicin. Not sensitive for lower concentrations. Time-consuming.
Csedo and Kopp (1964)	Ammonium vanadate	
Friedrich and Rangoonwala (1965)	Thin-layer chromatography	
Karawya and Balbao (1967)	Micro method, diazobenzene sulfonic acid method	
American Spice Trade Association (1968)	Scoville test	Subjective test recommended.
Spanyar and Blazovich (1969)	Thin-layer chromatographic method using ferric chloride and potassium ferricyanide reagent	Reaction not very specific. Coloring pigments interfere. Evaluation to be completed within 1–2 minutes after spraying to avoid interference by other pigment compounds.
Blazovich and Spanyar (1969)	Thin-layer chromatographic method	Applicable to oleoresin and other preparations containing large amounts of capsaicin.
Hollo *et al.* (1969)	GLPC method	
Hartman (1970)	GLPC method	
Mathew *et al.* (1971)	TLC method	Satisfactory.
Govindarajan and Ananthakrishnan (1974)	Paper partition chromatography	Simple, inexpensive, adoptable.
Indian Standards Institution (1976)	Scoville test modified	Adoptable method.
Govindarajan *et al.* (1977)	Scoville heat units (organoleptic evaluation)	Modified Scoville test, useful method.

TABLE XXX CLASSIFICATION OF ANALYTICAL TECHNIQUES FOR THE EVALUATION OF PUNGENCY (CAPSAICIN) IN CAPSICUMS[a]

Technique/basis	Authors
I. Threshold sensory (organoleptic) evaluation	Scoville (1912); Berry and Samways (1937); Hayden and Jordan (1941); Indian Standards Institution (1976); Govindarajan *et al.* (1977)
II. (1) Photometric methods	Gibbs (1927); Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry (1959)
(2) $VOCl_3$ method (vanadium oxytrichloride)	von Fodor (1930, 1931); Tice (1932); Schenk (1954, 1957); Benedek (1959a); Csedo and Kopp (1964)
(3) Phosphomolybdic acid method	Buchi and Hippenmeier (1948); Kosuge and Inagaki (1962)
(4) Follin–Denis reagent: phosphotungstic phosphomolybdic–vanillin method	North (1949)
(5) Diazobenzenesulfonic acid method	Schulte and Krueger (1955); Spanyar *et al.* (1957); Karawya and Balbao (1967)
(6) Sulfanilic acid method	Spanyar *et al.* (1956 , 1958); Zitko (1957)
III. Ultraviolet spectrophotometric methods	Suzuki *et al.* (1957); Brauer and Schoon (1962); Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry (1964)
IV. Polarographic titration with *p*-diazobenzene sulfonic acid	Spanyar *et al.* (1956)
V. Paper chromatography and colorimetry	Fujita *et al.* (1954); Waldi (1958); Govindarajan and Ananthakrishnan (1974)
VI. Thin-layer chromatography	Tiechert *et al.* (1961); Heusser (1964); Friedrich and Rangoonwala (1965); Blazovich and Spanyar (1969); Spanyar and Blazovich (1969); Mathew *et al.* (1971)
VII. Titration with picric acid and fluorescence desorption	Nogrady (1943)
VIII. Gas–liquid chromatographic methods	Hollo *et al.* (1969); Hartman (1970)

[a] From Pruthi (1970c), further updated.

ical tests indicated the presence of carbonyl and aromatic acid groups. Further work is necessary in this area.

d. Pungency in Garlic. Pungency in garlic is due mostly to allicin, a sulfoxide, and other sulfur compounds (Cavallito and Bailey, 1944; Stoll and Seebeck, 1947, 1948, 1949a,b,c; Zwergal, 1952). Previous studies on the quantitation of pungency in garlic and onion have depended mainly on the determination of volatile sulfur (Platenius, 1935; Currier, 1945; Kohman, 1952; Jager, 1955). Farber (1957) studied the chemical evaluation of the pungency of garlic and

TABLE XXXI VARIATIONS IN CAPSAICIN CONTENT IN IMPORTANT VARIETIES OF RED PEPPER GROWN IN DIFFERENT PARTS OF THE WORLD

Variety or type	Source	Capsaicin (% w/w)	References
1. Paprikas	Different sources	0.00–0.10	Kisgyorgy *et al.* (1962), Natarajan *et al.* (1968b), CAL (1969)
2. Hungarian paprika (ground)	Hungary	0.022–0.03	Pruthi (1969a)
3. Mambasas var.	Africa	0.60–1.10	Louis Sair (personal communication, 1969)
4. Red pepper	United States	0.25–0.45	Louis Sair (personal communication, 1969)
5. Hontakas	Japan	0.40–0.50	Louis Sair (personal communication, 1969)
6. Santaka	Japan	0.55–0.65	Louis Sair (personal communication, 1969)
7. Ethiopian chilies	Ethiopia	0.35	Louis Sair (personal communication, 1969)
8. Uganda var.	Uganda	0.85	Suzuki *et al.* (1957)
9. Turkish	Turkey	0.083	Suzuki *et al.* (1957)
10. Mexican pequinos	Mexico	0.26	Suzuki *et al.* (1957)
11. Santana	Japan	0.058	Suzuki *et al.* (1957)
12. Abyssinian chilies		0.075	Suzuki *et al.* (1957)
13. Indian chilies	India	1.05–1.59	Deb *et al.* (1963),
		0.60–1.86	Ramanujam and Tewari (1968)

onion by the measurement of volatile reducing substances. Pruthi *et al.* (1959b) standardized the technique for the estimation of pungency as volatile reducing substances in fresh and dehydrated garlic, garlic powder, and garlic salt.

e. Pungency in Ginger. Pravatoroff (1967) has reviewed the chemistry of ginger, with special reference to its pungency and the various factors affecting it. Connel (1969) studied the pungent principles present in fresh and stored ginger and their importance in ginger products. The major pungent principle was isolated from a commercially prepared sample of ginger oleoresin and, by usual spectroscopic and chemical techniques, was identified as shogaol. Processing and storage of oleoresin derived from ginger can result in chemical conversion of 50% of the product to other substances. This is the result of conversion of gingerol to shogaol and zingerone. The author further suggests that shogaol and zingerone do not occur naturally in fresh ginger rhizomes. Chemical changes of oleoresin at acid pH can occur at five times the rate observed at neutral pH and can be accelerated by elevated temperatures. Conversion of gingerol to the less pungent shogaol and thence to nonpungent residues is undesirable because of loss of pungency and an accumulation of nonpungent residues. Decomposition of gingerol to zingerone and aliphatic aldehydes must therefore be avoided (Connel, 1969). Eschemoser and Schinz (1950) reported on the constitution of zingiberene.

Connel and Sutherland (1969) also conducted the reexamination of gingerol, shogaol, and zingerone in dried rhizomes of ginger and reported interesting and new findings. Connel (1970a) also later isolated a new series of pungent compounds, the 1-(4-hydroxy-3-methoxyphenyl) alkan-3-ones, in ginger and in grains of paradise. Connel (1970b) systematically discussed the chemistry of essential oil and oleoresin of ginger, including the pungent principles and their chemical modification during preparation and storage of ginger oleoresin.

f. Pungency in Mustard. The International Organization for Standardization (1971) has described the argentimetric procedure for the estimation of allyl isothiocyanate in black mustard *(Brassica nigra)* and Indian mustard *(B. juncea)* and photometric and argentimetric methods for the estimation of *p*-hydroxybenzyl isothiocyanate in white mustard (*Sinapis alba*), in addition to other physicochemical specifications for mustard seeds.

Twelve collaborating laboratories have compared the AOAC method and the newly proposed GLC method for the determination of allyl isothiocyanate in whole mustard seeds. The GLC method has been recommended for adoption as the official first action method (Anderson, 1970).

Earlier, Kjar and Jart (1957) reported the separation of volatile isothiocyanates by gas chromatography. Pandey and Bholenath (1960) described a new method for augmenting the allyl isothiocyanate content in the oil milling of rape and mustard seeds. Mustakas *et al.* (1965) developed an improved method for isolating the pungent factor and for controlling protein quality.

g. Pungency in Onion. A survey of the literature (Schwimmer and Weston,

1961) on the chemical and enzymological properties of the onion strongly suggests that its pungency arises as a result of the interaction of *S*-substituted-L-cysteine sulfoxide derivatives and enzymes of the allinase type when the integrity of the onion tissue is destroyed by comminution or other means:

$$RSCH_2CHNH_2COOH + H_2O \rightarrow (RSOH) + NH_3 + CH_3COCOOH \text{ (pyruvic acid)}$$

The presumed unstable initial products, RSOH (sulfenic acid), can then react in several ways to form sulfur-containing odoriferous substances, which impart the characteristic pungency to homogenates of onion (Stoll and Seebeck, 1949a,b,c).

Schwimmer *et al.* (1964) reviewed the major advances in the past several years in our knowledge of onion flavor precursors (Carson and Wong, 1961b; Virtanen and Spare, 1962), enzymatic action (Kupiecki and Virtanen, 1960; Schwimmer *et al.*, 1960; Schwimmer and Weston, 1961; Schwimmer and Mazelis, 1963), and derived volatile end products (Carson and Wong, 1961a; Virtanen and Spare, 1962; Spare and Virtanen, 1963), which have established the major pathways leading to the characteristic odor developed by comminuted onion tissue. Galetto and Hoffman (1976a,b) reported on the flavor of alluims.

On the basis of the demonstrated stoichiometry of the above reaction, it was suggested (Schwimmer and Weston, 1961) that a correlation may exist between overall aroma, as measured by the olfactory threshold value, and the amount of enzymatically produced pyruvic acid (pyruvate) in the juice of freshly comminuted onion. Demonstration of a highly significant correlation (Schwimmer and Guadagni, 1962) showed that the determination of pyruvic acid in juice constitutes a reliable, simple, and convenient method for measuring the overall odor intensity of fresh onion. Extension of the pyruvate method to commercially dehydrated onion products would be of considerable practical importance as a means of standardizing the quality of these products, and as a control tool for following the fate of aroma during processing. Accordingly, Schwimmer *et al.* (1964) tested eighteen samples of onion powder from three onion dehydration firms for pyruvic acid content and for odor intensity after reconstitution with either water or acid. The water-reconstituted samples always contained more pyruvic acid and had a lower olfactory threshold than the acid-treated samples, thus showing that both enzyme and precursor substrate survived processing. Both pyruvic acid and odor intensity averaged about 1.8 times as high in the water-reconstituted samples as in the HCl-reconstituted samples. Comparison with previous results indicates that not all the potential odor intensity and pyruvic acid survived processing. Measurement of total pyruvic acid may be of value in assessing at least certain aspects of the flavor quality of dehydrated onions.

Previous studies on the quantitation of the pungency of onion and garlic have depended mainly on the determination of the volatile sulfur content (Currier,

1945; Kohman, 1952; Platenius, 1935; Schuphan, 1937). These studies have been based on the concept that the constituents responsible for onion and garlic odor and flavor were sulfur compounds, which could be separated by distillation (Semmler, 1892; Stoll and Seebeck, 1951). A study of the gaseous constituents of onion has been reported in which vapor chromatography, mass spectrometry, and infrared spectrometry were used to identify a number of the volatile sulfur and other compounds (Niegisch and Stahl, 1956).

The methods hitherto proposed for the quantitative estimation of the pungency of onion and garlic have involved many preparative steps and hence are time consuming. Their adaptation for plant or laboratory testing of many samples is therefore not too practical.

A procedure for the quantitative determination of constituents that are volatile at room or ambient temperatures has been described (Lang *et al.*, 1954). In a survey of its more general applicability (Farber, 1949), it was shown (1) that onions and garlic contain substances volatile at ambient temperatures that are able to reduce alkaline potassium permanganate solutions, and (2) that the content of these volatile reducing substances (VRS) diminished at the same time as the organoleptically judged pungency. Later, the same author (Farber, 1957) reported a somewhat more detailed study of the applicability of the VRS method to evaluate the pungency of onion and garlic. Its chief purpose is to attract the attention of those food technologists directly concerned with the problem to the usefulness of the VRS procedure as a fast, easily carried out, quantitative technique to complement and supplement the organoleptic judgment of the pungency of onion and garlic products. No attempt has been made to standardize the procedure, to set up practical ranges of values, or otherwise to adapt the method for practical, everyday application. Pruthi *et al.* (1959d) standardized the method for the estimation of VRS in fresh and dehydrated garlic.

The presence of relatively large amounts of pyruvic acid in onion was first detected qualitatively by Bennet (1945). Morgan (1946) proved its presence by isolation of its 2,4-dinitrophenylhydrazone (DNPH) from an unheated macerate and demonstrated that it arises enzymatically from precursors. Vilkki (1954) showed that these precursors are converted to both pyruvate and ammonia by onion juice. Schwimmer *et al.* (1960) demonstrated that allinase of onion produces pyruvate and ammonia in amounts equivalent to that of the disappearance of the substrate, as well as less than the stoichoimetric amounts of thiosulfinates.

The above summary of recent investigations on the mechanism of production of volatile flavor components suggests several approaches toward the objective evaluation of pungency on onions. The most direct overall approach would be the determination of volatile sulfur. This was used in the pioneering work of Platenius (1935). He determined sulfur (as barium sulfate after oxidation with bromine) in a distillate after acid hydrolysis of the onion at high temperatures for prolonged periods. Currier (1945) improved this method by using lower tempera-

tures and by converting sulfur eventually to methylene blue, thus affording in effect a colorimetric determination of sulfur. Kohman (1947), aware of the enzymatic nature of the production of the sulfur volatiles, employed steam distillation of onion macerates in a special apparatus, followed by conversion of the volatile sulfur components to barium sulfate. There appears to be a highly significant correlation between dry weight and pungency (Platenius and Knott, 1941).

Other methods that might be used on the basis of present knowledge of the properties of the sulfur volatiles are antibiotic activity (Virtanen and Matikkala, 1959); reaction with *N*-ethyl maleimide (Carson and Wong, 1961a,b; Schwimmer *et al.*, 1960); reaction with thiamine to form analogs of allithiamine (Morgan, 1946), which possess characteristic ultraviolet absorption spectra (Gmelin, 1959); and gas chromatography (Schwimmer *et al.*, 1960). The concentration of onion components related to pungency as reported by different workers is summarized in Table XXXII.

As an alternative to estimation of sulfur volatiles, the determination of increases in ammonia or pyruvic acid in macerates is suggested. The large amounts of endogenous ammonia render the determination of ammonia unattractive. On the other hand, the determination of pyruvic acid as a measure of flavor has already been applied to garlic by Jager (1955) and to Mexican varieties of garlic (Alfonso and Lopez, 1959).

Schwimmer and Weston (1961) have demonstrated that onion homogenates from different varieties and lots of onion produce pyruvic acid in varying amounts, which appear to correlate well with the generally accepted pungency of these varieties. Schwimmer and Guadagni (1967) also studied the kinetics of the

TABLE XXXII CONCENTRATION OF ONION COMPONENTS RELATED TO PUNGENCY[a]

Determination of components	Concentration (μmoles/g onion)	References
Pyruvic acid	2.0–19.0	Schwimmer and Weston (1961)
Total volatile sulfur	2.0–5.0	Currier (1945), Niegisch and Stahl (1956), Renis and Henze (1958)
Steam-distilled sulfur	2.0–7.0	Kohman (1947)
MCSO[b] + PCSO[c]	1.6–1.9	Stoll and Seebeck (1949a,b)
MCSO + PCSO + cycloalliin	5.0–11.0	Stoll and Seebeck (1949a,b)
Total sulfur in methanol extract[d]	18.0	Carson and Wong (1961a)
Pyruvic acid–garlic	47.0–63.0	Alfonso and Lopez (1959), Gmelin (1959)

[a] From Schwimmer and Weston (1961).
[b] MCSO: *S*-methyl-L-cysteine sulfoxide.
[c] PCSO: *S*-propyl-L-cysteine sulfoxide.
[d] Present in Southport white globe onion.

enzymatic development of pyruvic acid and odor in frozen onions treated with cysteine C—S lyase. Freeman and Whenham (1976) reported on the nature and origin of flavor components of onions and related species.

2. Cinnamaldehyde in Cinnamon

Rivas (1932) described a semimicro method for the determination of cinnamaldehyde in cinnamon bark. Mouton (1939) reported the determination of benzaldehyde and cinnamaldehyde as 2,4-dinitrophenylhydrazones and its possible application to an assay of galenic preparations of cherry laurel and cinnamon. Wachsmuth and Lenaers (1946) developed a colorimeter method for the determination of cinnamaldehyde with *p*-phenylenediamine. The American Spice Trade Association (1968) described a chemical method for the determination of cinnamaldehyde in steam-volatile oil from cinnamon or cassia by indirect titration.

3. Coloring Pigments

The color of spices is important from the point of view of quality as well as economic worth. The coloring pigments and the coloring power of only three spices have received some attention—red pepper or paprika, saffron, and turmeric.

a. Coloring Pigments in Red Pepper. A number of carotenoids have been isolated and characterized from red peppers *(Capsicum annuum)* or the dehydrated product, paprika (Curl, 1962; Cooper *et al.*, 1962; Chonoky *et al.*, 1963; De La Mar and Francis, 1969). The major red pigment is capsanthin, which was isolated in crystalline form as early as 1927 (Karrer and Jucker, 1950), but the current structure was assigned relatively recently (Barber *et al.*, 1960, 1961). Faigle and Karrer (1961) determined the asymmetry of the carbon-5 position in the five-membered ring of capsanthin, and Cooper *et al.* (1962) established that the hydroxyl in the cyclopentane ring is trans to the polyene chain. Capsanthin occurs as the dilaurate in paprika (Phillip *et al.*, 1971). The oxidation degradation sequence for capsanthin in oxygen was worked out by Phillip and Francis (1971a).

Sharma and Seshadri (1955) reported the unusual presence of anthocyanin pigments in the unripe fruits of four varieties of Indian chilies, all of which contained petunidin diglycoside.

Column chromatography has been the traditional method for separation of carotenoids (Davies, 1965). For example, with paprika carotenoids, Sea Sorb 43 is effective, but it does cause some isomerization and oxidation of pigments; hence, a more rapid method is desirable. Paper chromatography based on impregnated papers (Booth, 1962) and papers with suitable fillers (Jensen and Jensen, 1959; Jensen, 1960) has been suggested. Separations are described based on thin-layer systems on alumina (benzene), silica gel G (methylene chloride–ether, petroleum ether–benzene, undecane–methylene chloride, and methylene

chloride–ethyl acetate), calcium hydroxide (hydrocarbon mixture–methylene chloride), secondary magnesium phosphate (carbon tetrachloride, benzene, and petroleum ether–ether), silica gel G mixed with rice starch (*m*-hexane–ether), calcium hydroxide (benzene, benzene–methanol, and petroleum ether–benzene) (Stahl *et al.*, 1963; Demole, 1958, 1959; Bunt, 1964; Bollinger, 1965). Stahl and co-workers (1963) reported that not all carotenoid mixtures could be separated with a single solvent and a single absorbent in a thin-layer system. Phillip and Francis (1971b) reported the development of a solvent system for the thin-layer separation of paprika carotenoids as well as the physicochemical properties of capsanthin and derivatives. Rosebrook *et al.* (1968a) and Rosebrook (1971) reported the results of systematic collaborative studies on a method for the detection of the extractable color in paprika and paprika oleoresin. The color in paprika is extracted with acetone, and absorbance is measured at 460 mμ. The method incorporates a standard color solution as a spectrophotometric check. The method has been recommended for adoption. In a series of papers, Moster and Prater (1952, 1957a,b,c) described the extractable color, the color scale, the color of paprika oleoresin, and the structure of the coloring matter of paprika. Cholnoky *et al.* (1958) reported on the carotenoid pigments in yellow paprika, and Curl (1964) reported on carotenoids of green bell pepper. Thus, the subject of the assessment of color of capsicums and paprika has been investigated by a number of workers in different countries. Their findings have been condensed in Table XXXIII.

Shuster and Lockhart (1954) have reported on the comparative composite color grading of paprika by visual score, Lovibond units, C-units, and absorbancy at 462 mμ (Table XXXIV), and Lease and Lease (1956a) have reported on the different fractions of pepper pigments of different varieties of cayenne pepper and paprika as percentage of total color of the hexane extract (Table XXXV). Pruthi (1969a) reported on the loss in color (capsanthin and capsorubin) in Hungarian paprika during storage. Kanner *et al.* (1976, 1977, 1978) studied the carotene-oxidizing factors in red pepper.

b. Pigments in Onion Purée. Shannon *et al.* (1967a,b) investigated the reactions involved in and the conditions affecting the formation of pink pigments from precursors isolated from purées of white onion. According to Michaluk and Oswiecimska (1961) and Schwimmer (1967), the scales of yellow onion contain glycosides of quercetin. He has also reported the possible development of a bitter substance from flavonoid.

c. Coloring Pigments in Saffron. Before the advent of coal-tar dyes, saffron constituted an important source of dyestuffs. It is now used chiefly as a coloring and flavoring agent for foods. Sastry *et al.* (1955a) and Madan *et al.* (1966) have reviewed, among other aspects, the coloring pigments in saffron including crocin, crocetin, carotene, lycopene, zeaxanthin, and picrocrocin. Crocin constitutes the major pigment. One part of saffron in 10,000 parts of water gives a color equiva-

TABLE XXXIII ANALYTICAL TECHNIQUES FOR COLOR EVALUATION OF CAPSICUMS, AND FACTORS AFFECTING COLOR RETENTION THEREIN

Author	Title/principle of method	Remarks
Moster and Prater (1952)	Measurement of extractable color of capsicum	For routine analysis.
Shuster and Lokhart (1954)	Objective method of color evaluation, color grading, absorption spectra	
Moster and Prater (1957a)	Extraction of color	
Moster and Prater (1957b)	A linear color scale	
Moster and Prater (1957c)	Measurement of the color of oleoresins of paprika	
Garcia (1959)	Determination of capsanthin and capsorubin content	
Benedek (1959b)	Determination of color	
Pohle and Gregory (1960)	Spectrophotometric method, calibration with β-carotene	
American Spice Trade Association (1968)	Photometric method (ASTA)	
Khristova (1961)	Determination of pigments	
Santa Maria and de-Ruiz-Assin (1961)	Methods of analysis of paprika, color index (transmittance of 0.05 g/100 ml in CCl_4 at 460 mμ, 15%)	
Videki and Videki (1961)	American Spice Trade Association method, spectrophotometric method, compared with Benedek method	Lengthy and expensive although the inaccuracy is negligible. Benedek method preferred.
Serna (1962)	Photometric method measurement of transmission at 460 mμ, true color purity values	Results on 122 samples obtained with spectrophotometry and two colorimeters showed very close agreement with true color purity.

Sancho and Navarro (1962a)	Variation in color, effect of factors on carotenoids	
Sancho and Navarro (1962b)	Ultraviolet absorption spectra of carotenoids	Nine carotenoids obtained by chromatography.
Curl (1962)	Carotenoids of red bell pepper (% of different pigments given)	
Sancho (1962)	Photometric method, color measurement at 460 mμ, comparison with standard solution of $K_2Cr_2O_7$ and $CoCl_2$	
Palotas and Koneesni (1964)	Application of ASTA method (No. 10) and comparison with Benedek method	Values of one method are convertible to those of the other.
Benedek (1968)		
Rosebrook *et al.* (1968a)	Improved method for the determination of extractable color	
Lantz (1946)	Effect of slicing or slitting on time of dehydration and color	
Lease and Lease (1956a)	Effect of stage of ripening on the retention of color	
Lease and Lease (1956b)	Effect of fat-soluble antioxidants	
Lease and Lease (1962)	Effect of drying conditions	
Daoud and Luh (1967)	Method and nature of packaging, laminate and aluminum film pouches	
Chen and Gujmanis (1968)	Autoxidation of extractable color	
De La Mar and Francis (1969)	Carotenoid degradation in bleached paprika	
Phillip and Francis (1971a)	Nature of fatty acids and capsanthin esters in paprika	
Phillip and Francis (1971b)	Isolation and chemical properties of capsanthin and derivatives	
Rosebrook (1971)	Collaborative study of a method for extractable color in paprika and paprika oleoresin	

TABLE XXXIV COMPOSITE COLOR GRADING DATA ON DOMESTIC PAPRIKA SAMPLES[a]

Sample no.	Visual score[b]	Lovibond red[c]	C units[d]	Absorbancy at 462 mμ[e]	Order or grade[f]
1	24.7	17.7	44.6	1.024	1
2	23.9	16.8	41.0	0.950	2
3	21.7	15.6	36.5	0.870	3
4	19.3	15.0	34.0	0.830	4
5	16.1	14.6	32.6	0.752	5
6	12.2	13.7	29.5	0.676	6
7	11.8	13.3	27.7	0.620	7
8	8.2	11.9	23.8	0.540	8
9	6.0	10.8	20.7	0.485	9
10	3.9	9.6	17.4	0.415	10
11	2.2	8.2	14.0	0.342	11
12	1.0	7.0	11.0	0.277	12

[a] From Shuster and Lockhart (1954).

[b] Average of ten observers.

[c] Two-inch cell. Lovibond yellow units constant at 20–27; concentration 1:500; ethanolic extract.

[d] Two-inch cell. From Lovibond red units.

[e] Hardy recording spectrophotometer (cell path 1 cm; bandwidth 10 mμ; concentration 1:1000; acetone extract).

[f] An order of 1 signifies the reddest sample. The order was the same for the visual scores, the absorbancies, and the C units.

lent to that of 0.05% $K_2Cr_2O_7$ solution, when viewed visually or colorimetrically. Because it is subjective, the paper chromatographic method is being considered by national and international standards organizations such as the International Organization for Standardization. Biffoli (1955) reported a spectrophotometric method, and Munjal *et al.* (1971) described an elaborate method involving absorption spectra and TLC techniques for evaluation of quality with special reference to color.

d. Coloring Power of Turmeric. Turmeric *(Curcuma longa)* is a normal constituent of condiments, curry powders, and prepared mustards for coloring and flavoring them. It has been used in the past for dyeing of wool, silk, and unmordanted cotton. Curcumin, the major crystalline coloring pigment in turmeric, has been widely used in analytical techniques for boron, arsenic, etc. Its applications have been reviewed by Janaki and Bose (1967), who have also reported a simpler method for the isolation and estimation of curcumin, which is claimed to be superior to all the earlier methods. In the author's view, it is still time consuming and cannot be recommended for routine quality control. The American Spice Trade Association (1968) has described a simple method for determining color power (curcumin content) by extraction with 95% ethanol

followed by colorimetric or spectrophotometric measurement of the total extractable color as a measure of quality. This method is under consideration of ISO, with suitable modifications if necessary.

Mitra *et al.* (1956) have reported a modified wool-dyeing technique followed by spot tests and confirmation of added coal-tar dyes in turmeric by the circular paper chromatographic method.

4. *Glycoalkaloids in Pepper*

Broda *et al.* (1966) reported the determination of glycoalkaloids in seeds of Spanish pepper *(Capsicum annuum)*. The content of glycoalkaloids calculated as solanine was 1.8 to 2.1%.

5. *Morphine in Poppy*

Buchi and Huber (1961) discussed the possible methods of separation and determination of the principal alkaloids of opium, and the methods of determination previously used were summarized and partially examined. A method for the determination of the secondary alkaloids codeine, thebaine, papaverine, and nar-

TABLE XXXV FRACTIONS OF PEPPER PIGMENT OF 1950 PEPPERS AS PERCENTAGE OF TOTAL COLOR OF THE HEXANE EXTRACT[a]

Variety	Free xanthophyll (%)	Ester xanthophyll (%)	Carotene, nonsaponifiable (%)	β-Carotene (mg/g dry pepper)
Louisiana sports				
New	10.5	81.3	8.1	8.0
Aged (70.8%)[b]	4.9	61.3	10.8	2.9
Long cayenne 69A				
New	8.8	78.6	12.7	5.0
Aged (44.2%)[b]	6.6	67.8	10.4	1.2
"Seedless" cayenne				
New	3.9	87.8	8.3	9.2
Aged (35.9%)[b]	6.6	60.6	7.5	1.5
"Stock" cayenne				
New	7.4	85.4	7.2	4.2
Aged (65%)[b]	4.9	66.0	8.6	1.8
Paprika Carolina				
New	18.5	74.1	7.4	5.7
Aged (35.2%)[b]	11.1	64.7	7.1	0.97
Paprika 1-M-1				
New	12.1	77.2	10.7	13.0
Aged (30.8%)[b]	7.9	51.5	7.2	0.97

[a] From Lease and Lease (1956a).

[b] Percentage of total color left after 11½ months of aging at 25°C.

cotine is given. Danos and Sarkany (1961) reported the novel results of the poppy hybrid experiments directed toward increasing morphine production. Zoschke (1962) described a gravimetric method for the determination of morphine and studied genetically caused variability in the morphine content of poppy. Sakurai and Umeda (1961) reported the determination of main alkaloids in opium. Van Pinxteren and Verloop (1962) reported that opium and its derivatives interfered with the determination of morphine. This could be overcome by extraction of the eluate with $CHCl_3$ in the presence of $CHCl_2COOH$. Morphine remained in the aqueous layer, and the other alkaloids were extracted into the $CHCl_3$. Schnekenburger (1964) proposed a new, simple, precise gravimetric method for the determination of morphine representing a modified Mannich method. Trojanek *et al.* (1965) described isolation of morphine from opium poppy capsules.

6. Phenols in Nutmeg and Mace

The American Spice Trade Association (1968) has described a spectrophotometric method for distinguishing between nutmegs and maces grown in the West Indies and those grown in the East Indies; the oils from the former have absorbances of less than 0.020, whereas the oils from the latter normally have absorbances between 0.060 and 0.150.

7. Sulfur Compounds

Sastry (1957) described a photometric method for the determination of organic sulfur in asafetida. The procedure involves the reduction of an aqueous emulsion of the sample with zinc and sulfuric acid and subsequent reaction of the hydrogen sulfide formed with *p*-aminodimethylaniline hydrochloride to form methylene blue in the presence of ferric ammonium sulfate. Raghavan *et al.* (1974) determined the chemical composition of several varieties of asafetida, including sulfur content.

Markov (1964) reported a micromethod for the determination of sulfur in glucosides of garlic.

Parekh *et al.* (1958) reported the estimation of apparent SO_2 in fresh and pickled onions. Saghir *et al.* (1964) described a rapid and sensitive method of gas–liquid chromatographic separation and identification of sulfides from allium vapors. The vapors were withdrawn from small samples of chopped tissues. Data are given on retention volumes and on relative proportions of symmetric and asymmetric methyl propyl and allyl disulfide and of allyl monosulfide separated from vapors of *Allium ampeloprasum, A. cepa, A. bakeri, A. fistulosum, A. sativum, A. schoenoprasum,* and *A. tuberosum*. The relation of these compounds to allicin and to the odors of *Allium* species is discussed.

Delonca *et al.* (1963) modified the official French method of estimating allyl isothiocyanate in two ways: (1) Mustard flour was macerated for 1 hour in distilled H_2O at 40°C with stirring (instead of 1 hour at 20° to 30°C), and (2) after

distillation and addition of 0.1 *N* $AgNO_3$, the mixture was stirred for 1 hour at 80° to 85°C on a water bath instead of being kept in darkness for 12 hours. Diemair and Bauierh (1964) reported the interference in the determination of sulfurous acid in mustard oil-containing products. Excessive SO_2 values in the determination of SO_2 in the presence of mustard oil were studied with regard to the amount of SO_2 developed (from the allyl thiourea) from mustard seeds, the relation of the essential oil content to the SO_2 developed, the influence of the age of the seed, and the SO_2 developed from the mustard oil. The studies show that SO_2 develops from the essential oils through overheating and localized heating (Bunsen flame) during distillation. Distillation of SO_2 from a MeOH solution of the sample reduces the error in the determination.

Barothy and Neukom (1965) reported the synthesis of *p*-hydroxybenzyl isothiocyanate and its isolation from white mustard seeds.

8. Vanillin in Vanilla

Sharp (1951) reviewed the methods available for the assay of vanillin and proposed a modified gravimetric procedure that gives more accurate results than the previous method. A new volumetric method is described involving the titration of the acids produced by oxidation with hydrogen peroxide in alkaline solution. In addition to the colorimetric method, the interference of sugars in the reaction and the application of the method to processed goods containing vanillin have been discussed by Bains *et al.* (1958). Sherwood (1961) discussed the production of vanillin. Joppine (1961) described the detection and determination of vanillin and ethyl vanillin in a number of foods. Krummel (1961) made a comparison of two methods for determining vanillin mixed with sucrose. Pen (1966) described a colorimetric determination of vanillin with 2,4-dinitrophenylhydrazine. The mean relative error was 0.85%. Seniko and Reznikov (1967) developed a micromethod for the simultaneous determination of vanillin and vanillic acid in alkaline nitrobenzene oxidation products by employing paper chromatography and ultraviolet spectrophotometry. The relative error for vanillin was 1.5 to 2%, and that for vanillic acid was 5 to 7%. There are numerous other references on the subject, which have been reviewed by Guenther and his associates (1959, 1961, 1963, 1965, 1966, 1967, 1969, 1971, 1973, 1975, 1977). Bonnet (1968) reported the gas and TLC separation of compounds (natural and added) in vanilla extract, including the determination of vanillin.

III. DETECTION OF ADULTERATION

The available literature on adulteration in a few spices is found scattered in the form of reviews on asafetida (Subrahmanyan and Srinivasan, 1955; Sastry *et al.*, 1955b), cardamom (Kulkarni and Pruthi, 1967), cinnamon (Lawrence, 1967), pepper (Pruthi, 1968a), cardamom, ginger, pepper, and turmeric (Srivas *et al.*,

1963b), saffron (Fernandez Pizarro and Samper, 1953; Fungairifio *et al.*, 1954; Sastry *et al.*, 1955a; Madan *et al.*, 1966), and vanilla (Horst and McGlumphy, 1962; Guenther *et al.*, 1959, 1961, 1963, 1965, 1966, 1967, 1969, 1971, 1973, 1975, 1977). It is also partly covered in some standard books on food analysis such as those by Liverseage (1932), Leach and Winton (1941), Woodman (1941), Winton and Winton (1945) Jacobs (1958), Cox and Pearson (1962), the American Spice Trade Association (1968), and the Association of Official Analytical Chemists (1970, 1975). They cover the nature and extent of adulteration and methods of detection of adulteration, including physicochemical and microscopic examination. The available recent literature on some of the important spices is summarized below.

A. Asafetida

Sastry *et al.* (1955b) discussed the main differences in derivation, properties, and uses, as described in the literature, of the two major varieties of asafetida known to the trade as Hing and Hingra. Asafetida used in India as condiment is Hing in its many forms; Hingra is apparently the drug of European commerce.

The apparent woody residue associated with asafetida samples is not necessarily considered an indication of low quality.

Mere analysis (alcohol-soluble matter or sulfur content) does not reveal varietal differences that are physical and organoleptic in character. Chemical tests, some known and some evolved by the authors (Sastry *et al.*, 1955b) on the basis of certain functional groups in asafetida, have been investigated with a view to adopting them as a means of distinguishing between varieties, previously determined by smell. Classification of *Kabulidana* and *Shabandi* (varieties of Hing) by chemical tests is borne out by classification made on the basis of price. The need for a revision of the current standards for asafetida to include a clause relating to its organoleptic quality is pointed out.

Mitra *et al.* (1966b) described a method for the detection of colophony resin in asafetida.

Kartha and Sharma (1967) developed a test for the detection of colophony in asafetida (Hing). The test is a modification of the Lieberman–Storch test, which fails to detect colophony in the presence of incidental impurities. Admixture of 0.25% of colophony can be clearly detected in all cases.

B. Caraway, Coriander, Dill, Ajowan, and Fennel

The number of spots, their relative position, and the colors produced with various reagents on thin-layer chromatographic plates were used by Atal and Shah (1964) to detect adulterants in caraway, coriander, dill, and fennel. This technique could not, however, be used with ajowan. According to Betts (1964),

unknown umbelliferous fruits and powders can be identified by thin-layer chromatography of petroleum extracts on silicic acid-containing fluorescein. Constituents of essential oils thus extracted are usually determined. This method can distinguish Indian dill from European dill and will detect 1 part of the former in 4 parts of the latter.

By means of thin-layer chromatography in the system ether–benzene (1 : 1) saturated with 10% HOAC, an inventory was given by Horhammer *et al.* (1966) of cumarin and furocoumarin derivatives of some plant roots including spices possessing therapeutic value.

C. Cinnamon and Cassia

By capillary analysis (absorption of alcoholic tincture of $CHCl_3$ extraction on a strip of filter paper) and comparison with an authentic standard strip under ultraviolet light, Ceylon cinnamon can be distinguished from other spices of cinnamon bark, and various adulterants can also be detected (Berisso, 1940).

Betts (1965b) devised a direct thin-layer chromatographic method for the evaluation of cinnamon and its substitutes. Only cinnamon contained both eugenol and cinnamaldehyde, while the substitutes lacked one or the other, as well as varying in other respects, such as fiber structure or content of calcium oxalate. Stahl *et al.* (1967) identified cassia and cinnamon by TLC.

Components of powdered beechnut husks, aromatized with cinnamic aldehyde, and marketed as "pure powdered cinnamon," are described and illustrated by Czaja (1949). A microchemical reaction using phloroglucin hydrochloride can be applied for the detection of cinnamic aldehyde in the powder.

Deal (1967) tried the determination of volatile oils in cinnamon and cassia as a means for detecting and proving the substitution of a cheaper spice or volatile oil or other adulteration.

Voelker *et al.* (1967) developed a method for the determination of the geographical origin of cassia and cinnamon by TLC. The technique is designed primarily as a rapid quality control tool. The technique permits the determination of the composition of unknown mixtures of ground cinnamon with 20% of one type in a mixture.

Dutta (1961) reported that the contents of mucilage solution in 5% AcOH and precipitated by absolute alcohol, and of its ash, were, respectively, 1.9 to 3.7% (average 2.6%) and 17.2 to 24.6% (average 19.7%) for fourteen *C. zeylanicum* samples, and 8.0 to 11.5% (average 9.5%) and 6.1 to 7.8% (average 7.2%) for *C. cassia* samples.

A method is proposed by Eisenberg (1961) using direct flotation in 60% isopropyl alcohol, which gave cleaner papers and better recoveries of insect fragments. It was collaboratively compared with the present method and that of Schwartzman (1955).

Seideman (1962a) compared the starch of cassia bark powder with wheat starch by microscopy. Stahl *et al.* (1969a) reported a simple method for the differentiation of certain types of cassias and cinnamons by measurement of their mucilaginous character.

Buzas and Sqendrei (1964) reported the identification of plant drugs by ultraviolet fluorescence. An effective method for identifying more than one hundred crude drugs was developed. After drying, the pattern is observed with ultraviolet light, and the colors of the various zones (up to six) radiating from the center are noted. The method is illustrated with twenty-seven drugs.

Esses and Schelton (1956) in a series of six papers on food microscopy have given descriptions and photographs (including microphotographs) of thirty-five spices including cinnamon, cassia, nutmeg, mace, ginger, clove, aniseed, caraway, coriander, dill, fennel, cardamom, poppy, star anise, angelica, vanilla, turmeric, saffron, bay leaves, mint, parsley, sage, thyme, basil, marjoram, origanum, rosemary, savory, garlic, horseradish, celery, tamarind, parsley, caper, and tarragon. Quality, identity tests, and adulterants are discussed.

Ceylon artificial cinnamon was prepared by Schmal (1940) by mixing 3.4% of a mixture of 96% cinnamaldehyde and 4% eugenol (sometimes with the addition of 0.2% Ceylon oil of cinnamon) with a carrier such as powdered hazelnut or almond shells and coloring the mixture with yellow-brown dye. Cassia artificial cinnamon was prepared by varying the porportions of the ingredients slightly and adding cassia oil. Samples of ground cinnamon containing an excessive amount of reddish ash were first suspected of being adulterated with hematite, but investigation indicated that ocher had been added. Results for the ash of five commercial insect powders are given.

Villanua *et al.* (1954) reported the chemical and micrographic methods of analysis applicable to the following: aniseed, saffron, cinnamon, clove, cumin seed, bay leaf, mustard, red pepper, pepper, thyme, and vanilla. Notes on adulteration and on Spanish legislation are included. Richard (1921), Rosenthaler (1926), and Ferrara (1958) have also reported different methods of detection of adulteration in cinnamon.

D. Cloves

In twenty-five samples taken at Stockholm, an average of 5.06% of foreign matter was found by Harald (1919), including stems and fruits of cloves, rice fruit, white pepper, sand, earth, and extracted or colored cloves. The method of floating cloves in water at 15° to 20°C does not differentiate inferior cloves from sound cloves, especially with reference to the removal of oil. Wilhelm (1921) reported the presence of magnesium salt in cloves and its detection.

E. Chilies and Paprika

Konecsni (1956–1957) studied the microscopic determination of added food-paprika in milled spice-paprika.

Mitra *et al.* (1970) made a comparative study of estimation of starch in twenty samples of chilies by acid hydrolysis and diastase methods separately. They suggest that the Prevention of Food Adulteration Act (PFA) (Government of India, 1955) specification for the maximum limit of starch content (by the diastase method) in ground chili may be safely fixed at 4% instead of 1.5% as at present.

Navarao *et al.* (1965) have reported the thin-layer chromatographic determination of synthetic fat-soluble azo dyes in foods, particularly paprika adulterants.

Stelzer (1963) reported the identification of synthetic coloring in paprika by TLC. Pure paprika gave very low R_f values. Sudan II colored paprika gave R_f values of 0.78.

Maria and Da-Assin (1961) made a critical study of the methods of analysis of paprika and their application in the standardization of commercial products and detection of adulteration.

Schwien and Miller (1967) have reported a method for the detection and identification of dehydrated red beets in capsicum spices. Dried red beet pulp used as an adulterant in capsicum spices may be detected by removing the capsicum oil with petroleum ether and isolating the red beet particles from the decolorized spice. The beet particles are identified and confirmed by microscopic examination, paper chromatography, and spectrophotometric analysis.

Todd (1958) described a method for the detection of foreign pungent compounds in oleoresin capsicum, ground capsicum, and chili.

Sacchetta (1960) and Mitra *et al.* (1961) reported the detection of added colors to chili or capsicum powder by paper chromatography. Datta and Susi (1961) reported the quantitative differentiation between the natural pungent principle (capsaicin) and vanilyl-*n*-nonanamide in capsicum by infrared spectrophotometric procedures.

F. Pepper, Black and White

Pruthi (1968a) reviewed the detection of adulteration in black and white pepper and concluded that there was still further scope for research.

By determination of the temperature of swelling and conversion to paste of starch (swelling range) with the microheating stage of Boetius, it was possible to differentiate the starches of black and white pepper from those of long pepper, rice, oats, buckwheat, corn, and wheat (Seideman, 1962a). The swelling range for black and white pepper lies between 78.0° and 88°C, whereas the range for other starches lies below this point, with the exception of rice and rye meal. The

distinction of these latter starches from pepper starches was not possible with dyes. Polarized light also was of not much help in their differentiation.

Detection of rice flour in ground spices (Wagenaar, 1928), detection of adulterants by microscopical methods (Grunsteidi and Stobicke, 1936), and detection of olive kernels by using a solution of phloroglucinol in HCl (Paris, 1941), of a mixture of "arancini" and pimento in pepper by chemical methods (Lendner, 1919; Smith *et al.*, 1926), and of piperine and piperonol by biological tests (Piedelievre and Derobert, 1942, 1943) and chemical tests (Paris, 1941) have been reported. The most sensitive method of detection of piperine is by microsublimation and testing of the sublimate with cadmium chloride in strongly acidic solution (Paris, 1941).

The adulteration of powered black pepper with pepper hulls can be detected by the "counting method" or the "comparison number" method, which consists in examining the powder under the microscope and comparing the number of particles of a well-defined kind with another kind; the "normal number" is obtained by comparing the number of particles of a given kind with the weight of the sample (Grunsteidl and Stobicke, 1936). Many samples of commercial black pepper were examined by this method and analyzed quantitatively. In general, the sample was adulterated if the "comparison number" found was less than 2.75, as an average of five determinations of the "normal number" was over 10.7.

Smith *et al.* (1926) analyzed forty-five authentic samples of different varieties of whole black pepper, pepper shells, and pepper siftings, on the basis of which they suggested crude fiber, *d*-glucose, MgO, MgO:*d*-glucose ration, and MgO crude fiber as the most valuable criteria for detecting the adulteration of ground black pepper with added pepper shells.

Mitra and Roy (1955) reported the analysis of different varieties of genuine black pepper and some inferior or adulterated products and papaw seed, while Czaja (1953) reported the detection of roller-dried potato meal in spice mixes. Wick and Cairncross (1956) detected the off-flavor in pepper oleoresin owing to the presence of synthetic pepper bite compound.

Adulteration of pepper with palm-kernal meal can readily be detected in preparations cleared with chloral hydrate by the characteristic spotted appearance (illustrated by photomicrographs) shown by the thickened cell walls of the palm-kernel tissues (Czaja, 1962).

Bhatnagar and Gupta (1965) have described the macroscopic and microscopic characters of papaya seeds. The presence of fat globules, characteristic suberized cells of the outer region of the seed coat, and the large fiber cells of the inner portion of the seed coat are diagnostic features in microscopic detection of these seeds.

Mitra *et al.* (1966a) analyzed black pepper samples containing different amounts of light berries for ash, acid-insoluble ash, nonvolatile ether extract, and

starch. Only the starch content varied with the amount of light pepper present so that the starch concentration could be used to estimate the amount of light pepper present.

Pruthi and Kulkarni (1969) developed a simple technique for the detection of papaya seeds in black pepper berries by employing the flotation test, visual and microscopic examination of the floaters, and confirmatory tests.

Guenther *et al.* (1966) discussed the composition of ashes of black and white pepper. Hardly any basic differences exist. White pepper prepared from black pepper by decortication or water treatment is a falsification. The potassium content in unripe decorticated white pepper is higher than that in ripe commercial white pepper. Black pepper has a strongly increased ash and sand content as compared with ripe white pepper. The alkalinity number (K:Na) enables rapid differentiation between black and ripe white pepper and an identification of any falsified product prepared by decortication of black pepper.

In spite of the considerable amount of work done on the constituents of black pepper oil, it has not been possible to prove conclusively the addition of common and cheap adulterants, like the readily accessible terpenes and sesquiterpenes—namely, phellandrene, dipentene and caryophyllene—which are the natural components of the oil (Pruthi, 1968a). It may, however, be possible to assess the genuineness of the oil by determining the relative concentrations of these components and comparing them with those of standard samples of guaranteed purity. This aspect needs further detailed investigation. This may be possible through gas–liquid phase chromatography.

G. Saffron

Fungairifio *et al.* (1954) have given methods for proximate analysis, determination of flavoring principles, and detection of adulteration in condiments including saffron. Sastry *et al.* (1955a) reviewed the tests for detection of adulteration in saffron (Table XXXVI).

Netien and Mery (1962) have reported the chromatography of saffron adulterations. The circular paper chromatographic method is claimed to be sufficiently accurate to disclose mixtures of tinctures of saffrons. The R_f values for adulterated saffron tinctures (10% mixture) and color produced are given.

A new adulterant of saffron amounting to 73% of the drug has been illustrated and described by Turham (1950).

Sacchetta (1960) used paper chromatography of red paprika (*Capsicum frutescens)* powders. A method was given for the detection of alterations or artificial coloring of commercial paprika powder also adaptable to detecting fraudulent additions of paprika powders to saffron by a monodimensional ascending partition chromatography.

TABLE XXXVI DETECTION OF ADULTERANTS IN SAFFRON BY COLOR REACTIONS[a]

Adulterant	Reagent of treatment	Color	References
Nil (pure saffron)	H_2SO_4	Blue or bluish violet coloration with slight reddish tinge	Allen (1948)
	Alumina	Dull, orange lake	Barrington De Puyster (1919)
	Hot Fehling's solution (diluted 1:3)	Brick red	Parry and Bird (1925)
	Treated with 20 ml of water and shaken with 10 ml of amyl alcohol; evaporated residue treated with concentrated H_2SO_4, HNO_3, HCl or NH_4OH	Indigo blue	Aloy and Valdiguie (1925)
Brazilwood or sandalwood	Petroleum ether	Citron yellow	Allen (1948)
Infusion of poppy	Ammonia nitric acid	Greyish green Bright red	Allen (1948) Allen (1948)
Annato	C_6H_6, $C_6H_5CH_3$, $CHCl_3$, $C_2H_2Cl_4$, or CS_2	Green-blue	Bretin *et al.* (1931)
Artificial dyes	Organic solvents	Pink or red coloration	U.S. Department of Agriculture (1921)

[a] From Sastry *et al.* (1955a).

H. Turmeric

According to Mitra *et al.* (1956), the detection of coal-tar dye in turmeric has been made much easier by the use of modified wool dyeing and spot tests and circular paper chromatography.

Mitra and Roy (1957b) reported a paper chromatographic method for the detection of small amounts of turmeric in other spices.

Roy (1959) reported the detection of artificial fat-soluble coloring substances in turmeric and chili powders.

I. Vanilla

The paper chromatographic method proposed by Fitelson (1961) for the detection of foreign plant materials in vanilla extract was made "official" after a collaborative study.

Fitelson (1967) conducted further collaborative studies on methods for vanillin

and ethyl vanillin in flavoring material. Both the modified ultraviolet method and the paper chromatographic procedure are recommended for adoption as official final action.

Earlier, Fitelson (1962a) reported simple paper chromatography of the organic acids of vanilla extracts. All authentic extracts gave the same pattern of organic acids. Acids added to increase lead number could be detected. The method can detect foreign plant material having an organic acid pattern different from that of vanilla extract, and can also detect significant deficiencies in vanilla content.

Horst and McGlumphy (1962) described analytical methods (chiefly paper chromatographic) with brief comments for the detection of adulteration in vanilla extracts.

The official AOAC thin-layer chromatographic method for the detection of flavor additives in vanilla extracts was subjected to further study by Kahan and Fitleson (1964, 1965). Samples contained courmarin, veratraldehyde, piperonal vanitrope, and ethyl vanillin, singly or in combination. There were variations in the activity of the silica gel G used to make the thin-layer plates.

Metais *et al.* (1967) examined several types of foods to determine the substance responsible for the aroma of vanilla. Various methods of treatment are described for different foods, which made it feasible to extract the essences with toluene at pH 3. The extracts were subjected to the TLC technique. In all the foods tested, vanillin was the material most often responsible for the aroma of vanilla. In some cases, ethyl vanillin was found, and in some, both, A few samples were examined that apparently had neither.

Stahl *et al.* (1961) developed a two-dimensional technique for the analysis of pure and adulterated vanilla extracts. Final judgment is greatly strengthened by the additional use of results for organic and amino acids, vanillin, lead number, and resins. Stahl *et al.* (1960) determined organic acids in vanilla extracts.

Stall and Prat (1960) described a paper chromatographic method for separating acidic degradation products from vanilla beans, natural extracts, and products with added vanillin. It is suggested that it may be utilized to indicate spoilage.

Synodinos *et al.* (1964) employed TLC and paper chromatography for vanilla extracts. Analysis of commercial products in Greece showed a general use of synthetic vanillin. Adulteration with ethyl vanillin was observed in natural vanillin extracts and in chocolate in all cases.

Thaler (1959) reported that there was no basis for assuming the natural presence of ethyl vanillin in fermented vanilla capsules with eight different types of vanilla. The probability of formation of ethyl vanillin during fermentation is considered very unlikely. It was therefore considered a foreign ingredient according to German food law.

Wendt (1963) presented historical AOAC data on four FEMA (Flavoring Extract Manufacturers Association) samples of vanilla extracts. Results by ele-

ven collaborators employing the Association of Official Analytical Chemists (1970) methods on one genuine, pure (single-fold) extract and three prepared adulterated extracts to determine alcohol, lead number, resins, and vanillin show that these determinations are not adequate to detect a well-prepared adulterated vanilla extract.

J. Other Spices

Detection of adulteration in other spices such as ginger, mace, mustard, and poppy are discussed by Liverseage (1932) and Cox and Pearson (1962). Pruthi (1964) has reviewed detection of adulteration in and quality standards of curry powders. Miller (1967) reported a rapid method for the separation of extraneous materials, filth, rodent excreta, etc., and Kukkarni and Pruthi (1967) reported a simple, rapid test for the detection of adulteration in cardamom seeds.

Grunsteidl (1933) conducted an examination of spices by means of luminescence microscopy of fruit spices. The luminescence characteristics of pepper, paprika, pimento, nutmeg, mace, cardamom, vanilla, star anise, juniper, fennel, caraway, anise, cumin, mustard, and dill, and of their adulterants, are described.

Hanssen *et al.* (1957) have discussed microscopical detection of spices and other plant elements in foods. The value of microscopical and histological analysis for the detection of plant elements in foods is discussed.

Mitra and Roy (1957a) have discussed the analysis and detection of adulterants in some spices, particularly in black pepper, turmeric, coriander, and cumin.

Miller (1967) and Roaf and Brickey (1968) reported a rapid method for the separation of extraneous materials from whole and degerminated corn meal, prepared mustard, soy flour, and ground cinnamon. The proposed method improves recoveries of insect fragments by 13 to 34% and rodent hair recoveries by 25 to 54%. The analyst's time required for actual assay is reduced by at least one-third. The method deserves a collaborative study.

Sen *et al.* (1973b) have reported a quick and reliable TLC method for the detection of small amounts of mineral oil (a prohibited adulterant because of its carcenogenic effect). The method can detect mineral oil at levels below 0.1%.

Sen and Roy (1975) have discussed the difficulty in identifying the barks of cinnamon from those of cassia. After reviewing the limitations of the different visual, chemical, and chromatographic tests to distinguish between the two species, the authors suggest the use of their lesser known essential oil constituents to provide a means for solving the problem of identification. Sen *et al.* (1973a) have stressed the importance of volatile oil and cold-water extract estimation in analysis of cumins. Bose (1974) has suggested a spot test for the detection of *Argemone mexicana* seeds in mustard seeds. Divakar *et al.* (1974) have reported the detection of ragi *(Eleusine coracana)* in mustard seeds *(Brassica nigra)*. Sen

Gupta *et al.* (1973) have reported a simple TLC method for the detection of coumarin (a prohibited adulterant) in vanilla-flavored foods. The method is claimed to give better results than the AOAC method. Even traces of coumarin are detected easily. Martin *et al.* (1975) have described a method for differentiating between a true vanilla extract and one to which vanillin has been added. Bhagya (1977) detected capsaicin adulteration in ginger oleoresin. Sen *et al.* (1977) detected *Amomum subulatum* in *Elettaria cardamomum* by the TLC technique.

Rhyu (1979a) made a comprehensive comparison among the various types of sages commercially available in Albania, Greece, Italy, Turkey, and Yugoslavia during 1971–1977, based on the gas chromatographic characterization of steam-distilled essential oils extracted. The GC profiles thus obtained suggested that Greek, Italian, and Turkish sages are strikingly similar and are believed to be of *Salvia triloba* L. Though both the Yugoslavian and Albanian are *S. officinalis* L., they display substantial differences. The same author (Rhyu, 1979b) also reported gas chromatographic characterization of oregano and several other selected spices of the labiate family (peppermint, spearmint, rosemary, sage, savoury, sweet basil, sweet marjoram, and thyme). Gas chromatographic profiles show no differences in qualitative composition of monoterpenes, but only in their quantitative distribution. Differentiation between Greek and Mexican oregano has been demonstrated by use of the linear discriminant function (Rhyu, 1979b). Rajapakse-Arambewala and Wijesekera (1979) conducted a GLC study of steam-distilled essential oil of wild cardamom of Sri Lanka, which showed several significant differences from oil expressed from other varieties of cardamom. Thus, cold-pressed wild cardamom oil contained no *p*-cymene, while steam-distilled oil contained a high percentage. Thujene and *cis*-*p*-2-menthene-1-ol are the constituents reported for the first time. Earlier, Wijesekera and Fonseka (1974) used infrared spectroscopy in the analysis of cinnamon leaf, stem, and bark oils. The results obtained are in close agreement with those obtained by GLC.

Rhyu (1978) has suggested an improvement in precision in the estimation of threshold values by the Kaerber and Scoville methods.

The International Organization for Standardization (1979) has made a draft recommendation for the determination of trace residual solvents in oleoresins by gas chromatographic headspace analysis.

4

Chemical Composition

I. PROXIMATE COMPOSITION

The authorities concerned with the formulation and implementation of national and international standards have recognized the dearth of reliable data on the physicochemical characteristics of spices grown in different countries and in different regions within the same country, since, like other agricultural commodities, their composition varies from region to region. Some of the available information on proximate composition has been summarized in the older standard books on food analysis cited earlier. More recently, Pruthi *et al.* (1962) reported the physiochemical composition of twenty-one spices and condiments: ajowan, aniseed, cardamom (green), cardamom (amomum), celery seed, coriander seed, cinnamon, cloves, curry powder, fennel seed, fenugreek seed, garlic powder, garlic salt, ginger, mustard powder, onion powder, mace, pepper (black and white), chilies (red), and turmeric (Table XXXVII). Some of the reviews on individual spices mentioned in the Introduction also cover information on the subject. Dwarkanath *et al.* (1958, 1959, 1961; Dwarkanath and Ramachandra Rao, 1963) in a series of papers have reported the physiochemical composition of different varieties of pepper, their stalks, husks, etc. Pruthi and Misra (1963a) and Pruthi (1964) reviewed curry powders. Kulkarni and Pruthi (1967) reviewed cardamoms, and Pruthi (1968a) reviewed the available information on pepper.

From time to time, spice processors have been questioned by nutritionists on the mineral content of spices, with special reference to sodium content. Information on the subject is rather scanty (Elvehjem and Burns, 1952; Wenkam *et al.*,

1961). These articles contain no information on potassium, and yet the potassium levels of therapeutic diets are receiving increased attention. Christensen *et al.* (1968) employed direct-reading omission spectroscopy for the analysis of thirty spices and herbs for fourteen element minerals (Table XXXVIII). The reliability of the emission spectroscopy method was tested by comparing its results with those obtained on the same samples both by atomic absorption spectrophotometry and by conventional wet chemical methods (Table XXXIX). In general, the agreement between methods is good. Some limitations of emission spectroscopy are briefly discussed. This latest information on the subject may be of considerable value to dietitians, nutritionists, food scientists, medical authorities, and even agronomists.

II. CHEMICAL CONSTITUENTS

A. Carbohydrates

Jones and Thomas (1961) studied the structure of the gum asafetida polysaccharide. The oleogum resin of asafetida was extracted with hot methanol. A polysaccharide was precipitated when the extract was added to acidified EtOH. Acidic hydrolysis of the polysaccharide yielded *D*-galactose, *l*-arabinose, *l*-rhamnose, and glucuronic acid and its 4-O-Me derivative (5:3:trace:1).

Tookey *et al.* (1961) evaluated the seed galactomannans from legumes, including *Cassia emanginata* and *Trigonella foenun-graecum* (fenugreek), as wet-end additives for softwood, kraft, and sulfide finishes, and found them quite satisfactory. All the galactomannans tested as surface sizes were as effective as guar or locust bean gums in improving the strength characteristics of a corrosion coating base.

According to Harihara Iyer and Sastri (1933), the mucilage from fenugreek is usually associated with a nitrogenous impurity, which can be removed either by repeated dissolution in water and reprecipitation with alcohol or by saturation with magnesium sulfate. The purified mucilage is mannogalactan. Hassan and Morgan (1964) reported a direct semimicromethod for the determination of fenugreek mucilage and observed that dry, oil-free flour contained about 33% mucilage. Andrews *et al.* (1952) also studied the chemistry of galactomannans of fenugreek from North Africa, Asia Minor, India, and Pakistan.

Srinivasan and Bhatia (1954) reported that polyfructosan from garlic contains combined glucose (according to chromatographic evidence). In the light of this evidence, the molecular configuration proposed by Kihara (1937) for the polyfructosan from garlic, with fructose as the only structural unit, would need revision. Srinivasan *et al.* (1953) also observed that the pattern of carbohydrate makeup in garlic and onion is similar to that in *Agave veracruz* and reported the

TABLE XXXVII PHYSIOCHEMICAL AND MICROBIOLOGICAL EXAMINATION OF SPICES AND CONDIMENTS[a]

Spice	Moisture (%)	Volatile oil (%)	Total ash (%)	Water-soluble ash (%)	Alkalinity of water-soluble ash (%)	Ash insoluble in HCl (%)	Protein ($N \times 6.25$) (%)	Alcohol extract (%)
Ajowan	9.01	3.01	2.75	6.80	—	—	16.20	—
Aniseed	11.43	2.40	3.46	2.22	2.0	0.25	20.68	23.36
Cardamom (Green)	10.46	4.80	4.54	2.53	1.0	0.31	6.00	10.85
Cardamom (amomum)	8.49	2.80	4.01	2.15	0.90	0.42	7.38	7.02
Celery seed	11.44	Nil	6.50	3.49	0.70	0.35	18.62	21.90
Coriander seed	7.52	Nil	4.31	1.78	1.60	0.19	15.93	22.16
Cinnamon	9.49	0.70	5.41	2.80	1.4	0.88	25.57	8.18
Clove	7.72	13.20	5.26	3.17	3.0	0.24	2.19	5.64
Curry powder	7.73	1.20	3.21	2.20	2.0	0.62	15.06	23.8
Fennel seed	10.56	0.80	7.21	3.24	2.6	0.26	16.19	18.14
Fenugreek seed	8.36	Nil	3.31	0.89	1.6	0.64	28.56	15.58
Garlic powder	6.10	0.04	3.46	3.40	0.76	Nil	12.75	7.22
Garlic salt	10.02	Nil	82.23	78.43	0.33	1.04	3.0	4.54
Ginger	7.23	1.60	3.57	2.03	2.2	0.56	8.75	5.44
Mustard powder	8.29	Nil	3.78	0.62	0.36	0.27	27.75	11.05
Onion powder	6.21	0.04	3.31	2.44	0.51	Nil	23.15	10.1
Mace	8.43	11.0	1.64	1.12	1.5	0.84	8.44	30.04
Pepper (black)	9.73	1.6	4.55	2.82	3.3	0.08	14.25	10.8
Pepper (white)	8.87	1.72	2.06	0.64	2.1	0.08	15.12	18.08
Red pepper (chilies)	9.51	0.10	6.17	4.85	4.3	0.62	13.44	20.01
Turmeric	12.00	2.80	9.01	5.93	2.6	1.63	15.25	8.23

Spice	Total ether extract (%)	Non-volatile ether extract (%)	Volatile ether extract (%)	Crude fiber (%)	Starch (%)	Granulation test[b]		Total bacterial count/g
						Coarse particles	Fine particles	
Ajowan	18.59	—	—	11.20	35.20	—	—	1.4×10^4
Aniseed	36.06	34.05	2.01	8.69	23.15	6.0	4.8	1.7×10^5
Cardamom (Green)	6.17	2.97	3.19	18.70	45.0	3.0	5.5	1.0×10^4
Cardamom (amomum)	5.31	2.31	3.00	22.00	43.21	5.5	4.0	5.0×10^4
Celery seed	37.81	26.88	10.93	48.34	Nil	6.0	3.4	4.0×10^4
Coriander seed	22.57	22.21	0.30	32.02	17.99	6.4	2.6	6.0×10^4
Cinnamon	3.71	0.82	2.9	31.44	25.37	2.0	8.0	5.0×10^4
Clove	22.98	15.56	7.42	70.60	16.37	7.0	2.0	1.0×10^5
Curry powder	23.15	20.83	2.32	18.68	21.62	4.0	4.5	8.8×10^6
Fennel seed	21.66	6.53	15.13	70.53	Nil	6.5	3.0	4.0×10^4
Fenugreek seed	6.13	6.03	0.10	25.56	24.33	7.5	1.0	4.0×10^4
Garlic powder	6.31	6.20	0.10	2.40	Nil	2.0	3.0	5.0×10^4
Garlic salt	5.81	5.69	0.19	2.00	Nil	2.5	5.5	1.0×10^4
Ginger	0.89	0.32	0.57	4.10	43.2	6.5	3.5	8.6×10^4
Mustard powder	35.16	31.62	3.54	3.12	22.35	3.0	5.9	8.0×10^4
Onion powder	0.62	0.53	0.09	9.80	25.2	7.0	4.5	3.0×10^4
Mace	45.22	40.22	5.00	14.95	22.99	7.5	1.5	1.0×10^4
Pepper (black)	7.5	4.40	3.10	14.90	40.6	6.5	3.0	5.6×10^6
Pepper (white)	15.2	8.40	6.80	8.10	48.87	3.0	6.5	9.0×10^5
Red pepper (chilies)	17.51	16.36	1.15	29.92	16.0	7.0	2.0	Very high
Turmeric	21.41	7.73	16.68	11.90	24.41	Nil	8.8	6.3×10^6

[a] From Pruthi *et al.* (1962).

[b] On ground spices (30 mesh) only.

[c] N.B.: All values are calculated on a moisture-free basis.

TABLE XXXVIII ELEMENTAL CONTENT OF SPICES AND HERBS DETERMINED BY EMISSION SPECTROSCOPY[a]

		Percent					Parts per million								
Spice	Sample	Ca	P	K	Na	Mg	Al	Ba	Fe	Sr	B	Cu	Zn	Mn	Cr
Allspice	A	0.69	0.10	1.0	0.058	0.10	51	<2.0	60	3.5	14	3.3	11	5.0	<3.0
	B	0.93	0.13	1.0	0.087	0.16	101	17	81	15	9.5	3.5	7.0	65	<5.0
	C	0.76	0.11	1.3	0.086	0.11	88	2.0	85	<1.0	22	6.6	19	6.3	<3.0
	D				0.120										
Basil	A	1.7	0.48	3.7	0.032	0.38	167	19	225	66	25	13	70	25	<3.0
	B	2.1	0.43	3.9	0.055	0.44	450	27	550	100	18	10	38	39	<5.0
	C	2.4	0.49	3.4	0.042	0.41	355	31	510	85	31	16	67	31	<3.0
	D				0.016										
Bay	A	0.71	0.10	0.65	0.021	0.090	142	2.5	175	4.2	11	3.3	34	45	<3.0
	B	1.2	0.13	0.47	<0.010	0.16	730	<10	855	15	9.2	4.6	28	105	7.1
	C	1.0	0.11	0.65	0.020	0.11	550	7.1	570	1.8	16	4.6	49	95	4.2
	D				0.027										
Cardamom	A	0.59	0.18	3.0	0.022	0.44	200	56	200	23	14	5.6	87	300	<3.0
	B	0.22	0.24	<0.15	<0.01	0.24	23	<10	70	3.8	1.8	3.1	58	240	7.0
	C	0.18	0.20	0.60	<0.01	0.16	25	2.0	77	<1.0	12	2.8	79	300	<3.0
	D				0.011										
Celery seed	A	1.4	0.60	1.5	0.15	0.41	322	16	397	82	43	14	73	66	3.1
	B	2.0	0.36	1.4	0.23	0.48	730	20	620	<100	61	11	42	91	5.5
	C	1.9	0.68	1.3	0.14	0.43	200	18	330	63	50	16	93	70	<3.0
	D				0.14										
Cinnamon	A	1.4	0.055	0.62	0.009	0.055	81	60	47	72	15	3.3	26	123	<3.0
	B	1.8	0.072	0.15	<0.010	0.060	115	48	46	90	7.5	2.4	11	185	<5.0
	C	1.6	<0.025	0.50	<0.010	0.043	48	48	29	69	13	1.3	17	192	<3.0
	D				0.021										
Clove	A	0.69	0.10	1.4	0.21	0.27	81	33	57	43	25	3.1	11	176	<3.0
	B	0.71	0.11	1.0	0.28	0.28	160	22	85	57	15	2.7	4.0	315	<5.0
	C	0.83	0.11	1.1	0.27	0.26	116	29	143	39	31	4.6	21	410	<3.0
	D				0.021										

Coriander	B	0.75	0.44	1.1	0.020	0.38	27	10	39	12	9.4	6.5	35	15	<5.0
	C	0.89	0.44	1.4	0.022	0.26	36	4.9	79	14	29	13	59	23	<3.0
	D				0.040										
Cumin	A	0.86	0.44	2.1	0.12	0.34	452	8.7	>500	>100	34	9.1	51	40	3.7
	B	1.0	0.41	2.2	0.22	0.45	570	<10	525	>100	34	8.1	37	33	5.5
	C	1.0	0.49	1.9	0.14	0.38	273	7.1	410	190	50	8.8	56	27	<3.0
	D				0.014										
Dill seed	A	1.4	0.22	1.4	0.016	0.25	142	7.5	165	69	41	9.1	62	19	<3.0
	B	1.5	0.21	0.87	<0.010	0.26	110	15	110	85	28	5.5	35	22	<5.0
	C	1.9	0.20	1.1	<0.010	0.23	48	11	78	85	50	8.8	59	14	<3.0
	D				0.018										
Dill weed	B	1.5	0.36	5.0	0.11	0.33	90	14	130	73	11	3.2	20	38	<5.0
	C	2.0	0.40	4.7	0.11	0.34	88	19	150	55	30	6.6	46	41	<3.0
Fennel	A	1.3	0.39	1.9	0.12	0.31	81	6.7	117	>100	33	12	34	66	<3.0
	B	1.4	0.46	1.6	0.090	0.33	75	<10	120	140	25	7.0	25	66	<5.0
	C	1.2	0.59	1.6	0.075	0.30	45	4.3	95	95	36	13	52	64	<3.0
	D				0.052										
Garlic	A	0.074	0.39	1.2	0.014	0.055	<5.0	2.1	37	3.1	6.1	1.4	23	4.9	<3.0
	B	0.085	0.46	1.0	<0.010	0.077	<10	<10	28	7.0	2.5	2.8	18	6.0	<5.0
	C	<0.050	0.40	1.1	0.022	0.043	<10	<2.0	39	<1.0	4.5	.2	31	<2.0	<3.0
	D				0.022										
Ginger	A	0.074	0.13	1.4	0.021	0.15	73	16	60	4.2	2.2	2.1	44	265	<3.0
	B	0.12	0.13	1.1	0.045	0.20	200	23	150	7.5	1.0	3.2	48	235	<5.0
	C	0.085	0.20	1.8	0.022	0.18	102	32	128	<1.0	4.5	4.2	42	280	<3.0
	D				0.042										
Marjoram	A	2.3	0.22	1.6	0.12	0.31	>500	37	500	80	37	12	43	50	5.4
	B	2.8	0.27	1.5	0.13	0.38	1000	46	860	120	28	10	35	65	8.8
	C	2.4	0.20	1.1	0.083	0.30	740	30	820	23	48	11	46	48	<3.0
	D				0.032										
Mace	A	0.20	0.10	0.69	0.061	0.17	182	2.1	155	9.8	11	23	26	7.0	<3.0
	B	0.24	0.13	0.28	0.090	0.16	50	<10	64	15	3.6	23	11	12	<5.0
	C	0.27	0.11	0.60	0.53	0.16	102	4.9	120	6.5	13	28	22	10	<3.0
	D				0.032										
Mustard	A	0.34	0.66	0.80	0.005	0.28	<5.0	2.5	65	3.4	11	4.4	65	16	<3.0
	B	0.25	0.95	0.47	<0.010	0.37	<10	<10	90	9.0	<1.0	3.3	83	22	<5.0

(*continued*)

TABLE XXXVIII (*Continued*)

Spice	Sample	Percent					Parts per million								
		Ca	P	K	Na	Mg	Al	Ba	Fe	Sr	B	Cu	Zn	Mn	Cr
	C	0.21	0.77	0.72	0.019	0.30	<10	2.0	95	<1.0	6.4	4.6	110	14	<3.0
	D				0.010										
Nutmeg	A	0.17	0.20	0.60	0.011	0.17	<5.0	4.6	19	3.1	8.6	11	20	33	<3.0
	B	0.26	0.21	0.25	<0.010	0.23	11	<10	21	7.5	3.6	8.8	14	36	<5.0
	C	0.18	0.20	0.39	<0.010	0.15	<10	<2.0	26	<1.0	11	11	21	18	<3.0
	D				0.024										
Onion	A	0.19	0.24	1.1	0.031	0.081	<5.0	1.0	18	8.2	8.6	0.50	15	5.2	<3.0
	B	0.54	0.43	0.80	0.050	0.15	10	<10	22	70	6.2	3.5	28	7.0	<5.0
	C	0.30	0.20	1.1	0.032	0.095	<10	11	26	16	11	1.3	17	10	<3.0
	D				0.093										
Oregano	A	1.7	0.24	1.8	0.019	0.29	>500	14	>500	28	35	14	46	53	<3.0
	B	1.6	0.16	1.5	<0.010	0.26	700	15	490	31	21	5.5	35	46	<5.0
	C	1.9	0.20	1.7	0.019	0.26	590	19	610	23	41	8.8	52	41	<3.0
	D				0.031										
Paprika	A	0.090	0.27	2.1	0.019	0.11	49	2.5	127	0.85	9.9	4.4	70	5.2	<3.0
	B	0.17	0.23	2.5	0.010	0.26	700	<10	170	7.5	4.0	5.0	34	13	<5.0
	C	0.21	0.30	2.7	0.022	0.11	160	2.0	395	1.8	17	8.8	45	7.1	<3.0
	D				0.071										
Parsley	B	1.1	0.27	3.0	0.64	0.24	120	13	170	73	33	4.5	14	80	<5.0
	C	1.2	0.35	4.2	0.45	0.18	74	27	120	83	41	8.3	42	130	<3.0
	D				0.49										
Pepper,	A	0.34	0.17	1.5	0.005	0.15	237	35	270	19	8.6	14	14	30	<3.0
black	B	0.43	0.16	1.3	<0.010	0.20	130	50	200	36	4.0	11	5.2	75	<5.0
	C	0.40	0.16	0.85	<0.010	0.095	48	2.0	39	1.8	13	8.8	12	21	<3.0
	D				0.019										
Pepper,	A	0.058	0.29	2.2	<0.005	0.13	44	1.0	67	<0.50	11	3.3	17	16	<3.0
red	B	0.11	0.33	2.1	<0.010	0.18	67	<10	110	3.7	3.6	3.3	18	23	<5.0
	C	0.085	0.33	2.0	<0.010	0.14	48	<2.0	120	<1.0	13	4.6	21	14	<3.0
	D				0.019										

Pepper, white	A	0.19	0.16	0.085	<0.005	0.055	51	11	100	5.3	3.3	7.8	12	40	<3.0
	B	0.24	0.19	<0.10	<0.010	0.096	45	10	68	13	<1.0	7.5	4.8	53	<5.0
	C	0.18	0.11	<0.10	<0.010	0.043									
	D				0.009		25	4.9	39	1.8	4.5	8.8	9.0	31	<3.0
Poppyseed	A	1.5	0.78	0.84	0.011	0.31									
	B	1.7	0.95	0.60	<0.010	0.37	50	2.5	112	19	33	18	82	45	<3.0
	C	1.7	0.91	0.85	0.022	0.30	26	<10	90	27	15	13	100	80	<5.0
	D				0.011		22	4.9	95	10	30	18	125	80	<3.0
Rosemary	A	1.5	0.10	1.0	0.042	0.21									
	B	1.4	0.080	0.85	0.045	0.23	>500	11	500	69	39	4.4	36	20	<3.0
	C	1.5	0.030	1.1	0.040	0.22	425	10	270	18	22	5.5	28	22	5.0
	D				0.048		320	7.1	220	6.5	39	6.6	33	14	<3.0
Sage	A	2.1	0.10	1.0	<0.005	0.49									
	B	1.5	0.10	0.85	<0.010	0.43	307	14	255	11	33	8.7	62	40	<3.0
	C	1.7	0.070	1.1	<0.010	0.37	500	14	315	12	25	5.0	35	28	<5.0
	D				0.014		380	15	250	4.5	41	6.6	62	26	<3.0
Savory	A	1.5	0.17	1.2	0.021	0.39									
	B	2.6	0.14	0.93	0.020	0.40	>500	48	>500	15	37	7.5	30	113	4.8
	C	2.6	0.11	1.1	0.020	0.34	650	25	375	30	25	9.1	27	39	8.8
	D				0.054		400	29	260	11	34	8.8	72	31	4.2
Sesame seed	A	0.042	0.64	0.42	0.021	0.31									
	B	0.067	1.0	0.30	0.037	0.43	<5.0	9.6	60	2.3	11	18	95	13	<3.0
	C	0.050	0.73	0.50	0.040	0.30	<10	10	67	10	4.0	16	>100	20	7.2
	D				0.060		<10	7.1	59	<1.0	13	8.8	110	10	<3.0
Tarragon leaves	A	1.3	0.29	2.6	0.12	0.39									
	B	1.1	0.33	4.7	<0.010	0.22	412	9.6	432	30	47	9.1	67	106	<3.0
	C	1.5	0.32	2.3	0.090	0.43	175	<10	220	52	20	2.4	13	23	<5.0
Thyme	A	2.3	0.29	1.1	0.086	0.33	360	4.9	420	23	64	8.8	37	110	<3.0
	B	2.0	0.16	0.87	0.11	0.23	>500	61	1650	60	48	12	82	126	11
	C	1.9	0.16	0.85	0.040	0.26	>1000	73	1350	93	34	5.0	52	70	6.0
	D				0.086		1000	64	1050	50	45	8.8	58	90	4.9
Turmeric	A	0.15	0.24	2.4	<0.005	0.18									
	B	0.19	0.30	2.7	0.025	0.22	>500	25	>500	6.7	6.1	9.1	26	72	<3.0
	C	0.16	0.25	2.3	<0.010	0.18	500	20	485	15	<1.0	2.4	4.3	60	<5.0
	D				0.066		640	37	440	6.5	4.5	6.6	29	103	<3.0

[a] From Christensen *et al.* (1968).

TABLE XXXIX COMPARISONS OF SPECTROMETRIC ATOMIC ABSORPTION AND WET CHEMICAL METHOD[a]

		Percent					Parts per million			
Spice	Method[b]	Ca	P	K	Na	Mg	Fe	Cu	Zn	Mn
Allspice	Spec.	0.69	0.10	1.0	0.058	0.10	60	3.3	11	5.0
	A.A.	0.71	—	1.25	0.074	0.13	69	6.5	10	—
	Wet	0.74	0.113	0.96	0.092	0.13	69	5.2	9.3	5.0
Basil	Spec.	1.7	0.48	3.7	0.032	0.38	225	13	70	25
	A.A.	2.1	—	3.9	0.028	0.44	295	15	62	—
	Wet	2.3	0.56	3.6	0.030	0.44	380	15	54	19
Ginger	Spec.	0.074	0.13	1.4	0.021	0.15	60	2.1	44	265
	A.A.	0.080	—	1.6	0.039	0.19	88	7.0	38	—
	Wet	0.10	0.13	1.5	0.034	0.20	85	4.7	35	310
Marjoram	Spec.	2.3	0.22	1.6	0.12	0.31	500	12	43	50
	A.A.	2.6	—	1.3	0.10	0.34	637	13	30	—
	Wet	2.6	0.21	1.1	0.13	0.34	720	13	26	32
Pepper white	Spec.	0.19	0.16	0.085	0.005	0.055	100	7.8	12	40
	A.A.	0.19	—	0.070	0.002	0.070	100	11	9.7	—
	Wet	0.21	0.14	0.068	0.002	0.070	130	11	8.1	44
Sage	Spec.	2.1	0.10	1.0	0.005	0.49	255	8.7	62	40
	A.A.	1.6	—	1.5	0.005	0.42	189	11	41	—
	Wet	1.7	0.095	1.5	0.006	0.43	218	10	35	31

[a] From Christensen *et al.* (1968).

[b] Spec., spectrometric method; A.A., atomic absorption method; Wet, wet chemical method.

presence of fructose, glucose, sucrose, and three polyfructosans. The fact that they are free from starches seems to be significant for the understanding of the synthesis of polyfructosans, the status of which as a reserve plant food is now fairly well established. Bose and Srivastava (1961) studied soluble carbohydrates in onion and reported that onion juice contained fructose, glucose, and six oligosaccharides.

Constabel (1963) made quantitative studies of the extracellular hydrolysis of carbohydrates by tissue culture of juniper. Starch, sucrose, maltose, and raffinose were hydrolyzed to their constituent monosaccharides.

Kihara (1937) studied the carbohydrates of the bulbs of *Allium*. The bulbs of *Allium nipponicum* contained much scorodose. Scorodose could not be isolated from the white stalk of *Allium fistilosum,* the bulbs of *Allium cepa,* or the seeds and leaves of *Allium odorum*. The white stalk of *Allium fistilosum* was rich in the alcohol-soluble sugars, and sucrose was isolated from the alcohol extract. In a kind of pickle prepared from the bulbs of *Allium bukeri,* a part of scorodose remained unhydrolyzed.

Bazarova *et al.* (1964) reported the presence of five oligosaccharides in on-

ions. Singh and Bhatia (1972) reported that onion bulbs contain a polyfructosan, along with fructose, glucose, sucrose, and five oligosaccharides, characterized as F2G, F3G, F4G, F5G, and F6G, which were shown to be of the inulin type.

Wali and Hassan (1965) examined horticultural crops (eighty-two types—thirty-seven fruit and thirty-five vegetable varieties, and ten medicinal and aromatic plants) belonging to thirty-one plant families for total soluble solids, acidity, and pH, and also tested chromatographically for different sugars. In cut flowers, and in medicinal, aromatic, and condiment plants, rhamnose, maltose, xylose, and mannose were not detected in any of the plants. Sucrose was present in fair quantities in parsley *(Petroselinum crispum)* and in trace amounts in marjoram *(Majorana hortensis),* but was absent in dahlia, costmary, and *Mentha spicata.* Sucrose, arabinose, glucose, and fructose were absent in costmary and *M. spicata.*

B. Organic Acids

According to Soldatenkov *et al.* (1960), fresh and dehydrated onions contained malic, succinic, and citric acids—in bulbs 61.60%, 28.08%, and 10.32%, respectively, and in leaves 50%, 42%, and 8%. Some volatile and nonvolatile acids, which formed lactones, were also present in onion leaves.

By paper chromatography of organic acids in storage roots of some umbelliferae, Schramm (1961) found tartaric, malic, citric, isocitric, and traces of succinic and formic acids in celery and parsley. Quinic, shikimic, and uronic acids were also probably present.

Lewis and Neelakantan (1960, 1964) and Lewis *et al.* (1961) found that the leaves and fruits of the tamarind contain dominantly in all stages of ripeness, only as optically active *d*-tartaric acid. About 2% of the other acids are also present, chiefly malic acid. Almost half of the tartaric acid occurs in a combined form, chiefly as potassium bitartarate, and to a small extent as calcium tartrate.

Fitelson (1964) reported collaborative results on the determination of organic acids in vanilla extract by the gradient elution and paper chromatographic methods.

C. Pectic Substances

There appears to be limited published information on pectic substances in spices. Sen and Rao (1966) studied the pectic substances in onion by chromatography of H_2SO_4 hydrolyzate polysaccharides isolated therefrom and showed the presence of *d*-galactose, *N*-(*p*-nitrophenyl)-*d*-galactosylamine-*l*-arabinose, and *N*- (*p*- nitrophenyl) -*l*-arabinosylamine. Quantitative paper chromatography of the neutralized hydrolyzates gave the following estimates for polysaccharides A and B, respectively: 42.1% and 45.1%, anhydrouronic acid, 450 and 416 equiva-

lent weights, and galactose-to-arabinose ratios of 1.8:1 and 1.65:1. Both A and B also contained traces of xylose and rhamnose. Following further purification of a 2% aqueous solution of polysaccharide B by crystallization with 5% $CaCl_2$, this polysaccharide had an anhydrouronic acid content of 76%. After pectinate degradation of polysaccharide A, it had an equivalent weight of 2030 and a galatose-to-arabinose ratio of 6:1.

D. Nitrogenous Substances

Narasimhamurthy and Ranganathan (1937) found that dried black pepper contained about 2% nitrogen; the water-soluble fraction was small and consisted mostly of nonprotein substances. The proteins contained appreciable amounts of all the important amino acids. Wolter (1964) studied the differences in the occurrence of free amino acids in some diploid and polyploid plants, including cumin, by employing two-dimensional paper chromatography.

Hasan and Basha (1932) presented a preliminary note on a protamin isolated from fenugreek seed, which, like gliadins, was soluble in boiling 70% EtOH, but unlike the gliadins from wheat and maize, it was insoluble in cold 70% EtOH, Sreenivasa Rao and Sreenivasaya (1932) isolated and analyzed two proteins from fenugreek. The globulin (fraction A) is characterized by a surprisingly high content of histidine, which is about five times the average amount (2.27% of protein) contained in an average of ten typical globulins. The excellent tonic properties of the seed may be partly due to this fact, histidine being an essential constituent of hemoglobin. Fraction B is albuminous and is an interesting protein, since it appears to contain both phosphorus and sulfur. Sreenivasa Rao *et al.* (1933) characterized the prolamine (alcohol-soluble protein) from fenugreek as having a low basic nitrogen content and high percentages of cystine and tryptophan; thus, it resembles the alcohol-soluble protein of Ragi. Kolousek and Goulson (1955) determined the alkali-insoluble nitrogen, alkali-soluble nitrogen, and protein nitrogen, after isolating the proteins from seeds and thallus of Galanga and the seeds of fenugreek. The approximate quantitative amino acid composition of proteins was determined by means of rapid paper chromatography and was found to be comparable to that of the seed proteins of dicotyledonous plants.

Surinder Kaur and Vijayaraghavan (1961) isolated crude proteins from leaves of various plants including methi *(Trigonella foenum-graecum)* and analyzed them colorimetrically for methionine content. Maymone and Bergonzini (1964) studied the essential amino acid content and biological value of proteins of forage plants, including fenugreek, in which the limiting factor was methionine.

Ripa (1966) determined the protein content and amino acid composition of the proteins in fenugreek *(Trigonella coerulea)*. The nitrogen content was 2.06 to

2.6% (on a dry-weight basis); 88.8 to 92.3% of the total nitrogen represented protein. The proteins contained all the essential amino acids.

Srivas *et al.* (1963a) found that the total protein contents of freeze-dried ginger rhizomes and the spent residue left after the extraction of oleoresin were 10.46% and 11.26%, respectively. In fresh ginger rhizomes, the albumin, globulin, prolamine, and glutelins constituted 22.95%, 10.92%, 7.10%, and 11.48%, respectively, of the total nitrogen (1.83%), and nonprotein nitrogen content was 35.51%, with 12.01% being not extractable. The spent residue (200 mesh) contained albumin 22.06%, globulin 11.76%, prolamine 8.08%, glutelins 14.71%, nonprotein nitrogen 37.51%, and the nonrecoverable nitrogen 7.35% of the total nitrogen (1.36%).

The same authors also reported that the rhizomes as well as the hydrolyzate of the protein contained the following free amino acids: alanine (traces), aspartic acid (+ +), glutamic acid (+), asparagine (traces), glycine (+), lysine (traces), serine (+), valine (traces), arginine (+), cystine (+), histidine (traces), leucine (+), methionine (+), threonine (+), tryptophan (traces), and proline (++). The hydrolyzate did not, however, contain tryptophan. The concentrations of amino acids were different in the two materials.

Srivas *et al.* (1963a) also found that, electrophoretically, the ginger protein isolate (from the alkali extract) at pH 4.5 was homogeneous in character, comprising a single component (probably albumin), which migrated to the anode, indicating that it was negatively charged. Ginger flour (200 mesh) contains a fair amount of protein, unlike tuber flours.

The Japanese workers Murakami *et al.* (1965) have described the procedure for the recovery of asparagine (500 mg) and picoline acid (320 mg) from ginger rhizome.

Klimenko (1959) presented a tabulation of the distribution of various fractions of nitrogen in the seeds of radish, horseradish, and several varieties of cabbage. The total nitrogen in ground seeds was substantially the same for all the species examined. The nitrogen content was the lowest in horseradish and highest in cabbage. The globulin level was the same in all the species examined. Histidine and cystine were the lowest in horseradish. Methionine levels were quite similar in all seeds, although radish appeared to be richer.

Arai *et al.* (1957) reported that the eight *Allium* species contained large amounts of leucine, isoleucine, valine, and alanine, lesser amounts of asparagine, tyrosine, glutamic acid, and aspartic acid, and traces of lysine, arginine, and histidine. Takagi (1962) reported sulfur-containing amino acids in onions.

Atal and Sethi (1961) determined semiquantitatively alliin in *A. ampeloprasum* 0.4%, *A. sativum* (white) 0.8%, *A. sativum* (red) 1.0%, and *Allium* species (pran) trace. Paper chromatographic analysis of a 75% alcohol extract

showed the presence of alanine, arginine, aspartic acid, asparagine, histidine, leucine, methionine, phenylalanine, proline, serine, threonine, tryptophan, and valine in most of the species. An unidentified amino acid was found in a species containing little or no alliin. Virtanen and Mattila (1961a) reported that the elementary composition of the new peptide isolated from *A. sativum* in crystalline form is $C_{11}H_{18}O_5N_2S$, m. 156° to 158.5°C (decomposition); $[\alpha]_D^{23}$ 17.1 (H_2O). The structure is $HO_2CCH(NH_2)CH_2CH_2CO$ $NNCH(CO_2H)CH_2SCH_2$-$CH{:}CH_2$.

Virtanen and Mattila (1961b) also stated that at least nine glutamyl peptides are likely to be present in garlic.

Sugii *et al.* (1964) reported that the roots of garlic showed a high synthesis of sulfur-containing amino acids, and the earlier major labeled amino acids were crysteine and methionine. After cultivation for 24 hours, the formation of *S*-allyl-*l*-cysteine sulfoxide and *S*-methyl-*l*-cysteine sulfoxide was observed. In addition to these characteristic sulfur-containing amino acids, many unidentified sulfur-containing amino compounds were also detected.

Takagi (1962) reported the presence of a sulfur-containing amino acid in onion, believed to be $C_6H_{11}O_3NS$.

Virtanen and Matikkala (1960) reported the presence of neue-a-glutamyl peptide in onion. Dakshinamurthy (1962) investigated the methionine content of some South Indian foods, including onions, which contained 0.2% nitrogen and 0.014 ± 0.002% methionine. By means of paper chromatography, methionine content was found to be 0.011 ± 0.005%. Kuon and Bernhard (1963) studied twenty-two free amino acids of four varieties of onions by two-dimensional ascending paper chromatography. The more abundant amino acids were arginine, glutamic acid, phenylalanine, leucine, tyrosine, lysines, and methionine sulfoxide. Matikkala and Virtanen (1967) observed that both qualitative and quantitative differences in amino acids and peptides occurred in the varieties studied when examinations were made both directly on EtOH extracts and on neutral alkaline and acidic amino acid fractions of onions. It was not clear to what extent the differences were due to the variety or to the growth circumstances (for example, differences in sulfur content of the soil). Typical results obtained with EtOH extracts and nonacidic (neutral and alcoholic) fractions in micrograms of amino acid or peptide per gram of onion (parenthetical values represent the acid fraction), were arginine 1442 and 1465 (15.0), asparagine 391 and 407 (1.8), glutamic acid 346 and 10.1 (360), serine 166 and 178 (1.4), tyrosine 162 and 168 (24.6), and aspartic acid 131 and 6.5 (127), respectively. The highest values found were 1927 and 1922 (1965), respectively, associated with *S*-(prop-*l*-enyl)cysteine sulfonide for the first two groups and with γ-glutamyl-*S*-(prop-*l*-enyl)cysteine sulfoxide for the acid fraction.

Kholodova (1964) studied the composition of proteins of poppyseeds in relation to their physiological function. Poppyseeds contained water, salt, alkali

proteins, and nonextractable nitrogen compounds, which were chiefly proteinaceous substances. The nonextractable nitrogen fraction decreased during seed development, and globulins and albumins, which were up to 90% of the protein of isolated aleurone grains, accumulated. One major and three minor globulin fractions were found by paper electrophoresis, and seven albumin fractions were detected by chromatography on DEAE-Sephadex. The heterogeneity of the globulin and albumin fractions, their appearance in seeds prior to the formation of aleurone grains, and the absence of proportionality between enrichment and purification during extraction of aleurone grains indicated that some components of these compounds are not reserve proteins. A specificity of amino acid content of the protein fractions, low glutamic acid in nonextractable protein, and absence of methionine in albumin and glutelins were found.

Yoshio Hirose *et al.* (1962) found the following substances in saffron *(Crocus sativus)* by paper chromatography: glucose, aspartic acid, glutamic acid, cystine, serine, glycine, threonine, tyrosine, alanine, arginine, and histidine. A saponin combined with oleonolic acid as aglycon and a steroid saponin were also identified.

Endo and Tanji (1960) isolated asparagine, isoleucine, valine, aminobutyric acid, *d*-glucose, mannose, and glucose from the rhizome extracts of sweet flag by cellulose column chromatography. Proline, histidine, alanine, glycine, aspartic acid, and fructose were detected by paper chromatography. Mannitol was discovered as a reserve substance during winter.

Stahl *et al.* (1962) reported the amino acid content of vanilla extracts. The data permit differentiation between extracts containing significantly different proportions of Bourlion, Mexican, and Tahiti vanilla beans. Babayan *et al.* (1978) studied amino acid composition of *Nigella sativa* seeds.

E. Lipids

Dranitsyna (1961) examined two oil plants of the umbelliferae family from the Central Sayan Mountains and found that the fruits of *Angelica archangelica* contained 18% crude oil and compounds of the coumarin series. The mixed fatty acids of refined oil contained saturated acids 8.4%, oleic acid plus petroselinic acid 49.07%, and linoleic acid 42.53%.

Rakuzin and Starobina (1924) determined the fat content of the fruits of the most important umbelliferae. The fat content of the fruits of various umbelliferous plants—aniseed, coriander, dill, fennel, and caraway—may be deterined by extraction of the finely ground oven-dried material with $CHCl_3$ or CCL_4, after removal of essential oil by steam treatment of coarsely ground air-dried seeds. Fruits of these plants contain 15 to 28% fat.

Fefer (1959) determined the composition of the liquid fraction of the edible oil from fennel seeds. It contained 4.7% unsaponifiables. The constituents of the

glycerides were as follows: oleic acid 40.2%, linoleic acid 24.8%, saturated acids 5.2%, and petroselinic acid 29.8%.

Dublyanskaya (1964) presented tabulated data on the chemical composition of the fruits of *Pimpinella anisum, P. aniselum, Coriandrum sativum, Carum carvi, Foeniculum officinale, Trachyspermum capticum,* and *Salvia sclarea* seeds, and their fatty and essential oil physicochemical constants.

Karting (1966) also examined the lipid fractions extracted from the fruits of *A. graveolens* and *C. sativum* by employing standard methods. The former contained saturated acids with 16, 18, 22, 24, 26, and 28 carbon atoms and alcohols containing 24, 26, 28, 30 and 32 carbon atoms, but no unsaturated acids or alcohols. Coumarins (bergapten and umbelliprenin) were also found, as well as γ-sitosterol and an unidentified fraction with absorption maxima at 209, 263, 330, and 350 mμ and C=C valence stretching bands at 1600 and 1735 cm^{-1}. The same fraction was obtained from *C. sativum,* as well as sitosterol, umbelliferone, and scopoletin. Salzer (1975) reported on the fatty acid composition of lipids of spices.

Vidyarthi and Dasa Rao (1939) studied the fatty acids and glycerides of the fat from the seeds of *Garcinia indica* (kokum butter). By the usual method of lead salt precipitation and ester fractionation, the component fatty acids were found to be myristic acid (1.2%), palmitic acid (5.3%), stearic acid (52.0%), and oleic acid (41.5%). The component glycerides, in round numbers, are tristearin (2%), oleodistearin (59%), dioleostearin (21%), oleopalmitic stearin (14%), oleodipalmitin (2%), and palmitodiolein (2%).

Morozov (1959) determined the fat and allyl oil contents of yellow mustard. The fat content of the seeds of yellow mustard depended on the water supply of the plant. During three crop years, the average fatty oil content of the seed was 43.5%, 37.7%, and 38.4%, and the average allyl oil content was 0.43%, 0.78%, and 0.75%.

Goldovskii (1962) examined the principal types of quantitative interrelationships for the fatty acids in vegetable oils. The percentage distribution data for 292 varieties of seed oils plotted on the basis of their iodine and saponification values together with the graphs showing the percentage of individual fatty acids in some seed oils including nutmeg oil are given.

Vereschchagin (1962) studied the composition of higher fatty acids and their triglycerides in poppyseed oil by reverse-phase paper chromatography. The composition in mole percent was dipalmitoolein 0.06, stearopalmitolinolein 0.18, palmitodiolein 0.89, stearooleolinolein 1.22, triolein 1.15, dipalmitolinolein 6.35, palmitodilinolein 19.53, oleodilinolein 16.83, and trilinolein 48.31. Results do not agree with the theory of random distribution. The possible channels of triglyceride synthesis in higher plants are discussed.

Fischer (1965) presented a literature report on oxidative fat spoilage through aging, and on the influence of radiation on linseed and poppyseed fats.

F. Polyphenols

Heintze (1964), while studying polyphenols in vegetables, reported that red peppers contained polyphenols in amounts ranging from 350 to 450 mg per 100 g.

Harborne (1965) studied the characterization of plant polyphenols and flavonoid glycosides by acidic and enzymatic hydrolyses of onion and found the 3,4′- and 7,4′-diglucosides of querecetin in *Allium cepa* bulb scales.

Eisissi *et al.* (1965), while examining local plants as potential sources of tannins and studying the isolation of their free and combined sugars, found that *Mangifera indica* varieties contained 9.2 to 14% tannins, which were of the condensed type and contained flavonoid nuclei.

G. Coloring Pigments

The isolation and identification of pigments in important spices is briefly discussed here.

The red coloring of red-ripe capsicums, chilies, and paprika is known to comprise a complex mixture of carotenoids, xanthophylls, etc. Sharma and Seshadri (1955) reported for the first time the unusual occurrence of an anthocyanin (petinidin diglycoside) mostly confined to the pericap of four varieties of chilies grown at Indian Agricultural Research Institute, New Delhi. It is rather unusual that the unripe fruits of these varieties are deep purple and contain high anthocyanin content, wheras the ripe fruits become red and contain much less anthocyanin.

Cholnoky *et al.* (1958) determined quantitatively the pigments in yellow paprika *(Capsicum annum)* by chromatographic separation, absorption maxima, and chemical properties. Unripe fruit of yellow paprika contained β-carotene, xanthophyll, violaxanthin, and certain cis isomers of these compounds, traces of β-carotene monoepoxide, α-cryptoxanthin, antheraxanthin, and foliaxanthin (an epoxide of unknown structure). Ripe yellow fruit contained β-carotene, α-carotene, α-cryptoxanthin, *p*-cryptoxanthin, xanthophyll, violaxanthin, and cis isomers of these compounds. These results were compared with other investigations of paprika, and the following conclusions were drawn. Carotenoids act as oxygen transporters in chlorophyll-containing organs, operating by three systems: (A) zeaxanthin monoepoxide, antheraxanthin, xanthophyll; (B) β-carotene, β-carotene monoepoxide, α-carotene; and (C) *p*-cryptoxanthin, β-cryptoxanthin monoepoxide, α-cryptoxanthin. System A is the main system, systems B and C serve as reserves. System C increases in ripened fruits. Ripe fruits of red paprika contain pigments of β-structure epoxides and polyene oxyketones, but no pigments of α structure. Ripe fruits of yellow paprika contain α and β structures and an epoxide of β structure, but no polyene oxyketones. Possible explanations for this have been given by the authors.

Benedek (1959b) studied the after-ripening chemical changes in Hungarian red paprika with special reference to coloring matter. In Hungary, two kinds of red pepper are grown—hot and nonhot types. Trials were conducted accordingly with these two kinds of red pepper. The difference between hot and nonhot (nonpungent and sweet) red pepper consists not only in the capsaicin content of the former, but also in the chemical composition, which is disadvantageous for the latter.

The deep color of pepper harvested when red-ripe is due to, beside desiccation, the increased content of coloring matter (total carotenoids) in the pericarp, compared to total dry matter (Benedek, 1959). This increase in coloring matter is rather considerable in hot pepper (nearly 120%), whereas in nonhot peppers it is much less, about 65%. In the hot varieties, the increase in coloring matter content comes to a standstill about 5 to 6 weeks after harvesting, whereas in the nonhot varieties it is observed earlier, in about 3 to 4 weeks. The mean coloring matter content of fruits not subject to after-ripening, but left on the plant until fully red-ripe, is about the same in the two types; the higher coloring matter content that develops in the hot varieties occurs during the course of after-ripening. The increase in coloring matter content is accompanied by a decrease in total sugar content; this phenomenon is particularly striking in hot peppers. The decline in sugar content of the hot red pepper varieties comes to an end at a later date, in about 7 to 8 weeks after harvesting, whereas in the nonhot pepper it is earlier, in about 5 to 6 weeks. In this period, hot pepper loses on an average 32% of its sugar content established at harvesting time, whereas in nonhot varieties this value is 25%. The rate of desiccation is higher in nonhot red pepper than in the hot type (Benedek, 1959a).

Cholnoky and Szabolcs (1960a,b), while studying the structure of capsorubin, suggested that the methyl groups of capsorubin were in positions of 1, 1, 5 (1, 1, 5) and the OH group was in position 3 (3). Capsanthin and cryptocapsin could have analogous structures. These writers also discussed and illustrated the structure of capsanthin and capsorubin, as supported by ultraviolet data.

Zitko and Ondreicka (1960), in studying the biochemistry of the various kinds of natural wilting in spicy paprika, determined the amounts of solids, coloring matter, and reducing sugars and the activity of paroxidase and ascorbase during natural wilting in three varieties of Slovak spicy paprika (TS-18, TZK Fekete, and Agronomka) in wreaths or in loose bunches. Wilting occurs in wreaths at a much slower rate; after 9 weeks, total solids were 59% higher, and reducing sugars, peroxidase, and ascorbase activity were 8.8%, 9.3%, and 33% lower, respectively, than in the bunches. The coloring matter in TS-18 and Agronomka was 16% lower, and in TZK Feketa it was the same. The optimum for wreaths and bunches was 6 and 9 weeks, respectively. The higher the peroxidase activity in fresh fruit, the lower was the amount of coloring matter formed.

Zitko and Durigova (1961), continuing the study of the biochemistry of the

production of spice paprika, reported the presence of hydroxycinnamic acids and four flavonoids in paprika, by employing chromatography. Chlorogenic acid was identified from its R_f value. Capsaicin was separated from the carotenoids and identified. Another phenolic compound with an R_f value of 0.96 was also found.

Curl (1962) studied the carotenoids in red bell peppers. Capsanthin accounted for about 35% of the total carotenoids, with β-carotene and violaxanthin about 10% each, cryptoxanthin and capsorabin about 8% each, and cryptocapsin about 4%. Numerous other carotenoids were present in amounts of 2% or less, including at least eight apparently undescribed previously and two others not found previously in Nature. All ten of these pigments appear to contain cyclopentane rings, as do capsanthin, capsorubin, and apparently cryptocapsin. In addition to capsanthin and capsorubin, five other constituents were found to contain keto groups. Capsanthin is distinctly different from the deep-red pigment (reticulataxanthin) in tangerine and orange peels. Later, the same author (Curl, 1964) reported that the most abundant carotenoid in green bell peppers was lutein, with β-carotene, violaxanthin, and neoxanthin also as major pigments; minor pigments included phytoene, phytofluene, α-carotene, and γ-carotene. No ketocarotenoids such as capsanthin or capsorubin were found, nor was capsolutein, a lutein-like pigment occurring in place of lutein in the red-ripe fruit.

Atal *et al.* (1966) reported the occurrence of sesamin, originally obtained from *Sesamum indicum,* in long pepper (*Piper longum*). Kurte (1968) studied the pigments of red sweet pepper *(Capsicum annuum).* All the xanthophylls in the pericarp are combined with palmitic, myristic, and lauric acids. Unsaturated fatty acids, such as oleic, found in the fruits were not incorporated into the pigment esters. More than sixty-six compounds were derived from fourteen xanthophylls and three fatty acids.

Pink discoloration in ginger during its sun-drying and dehydration was attributed by J. S. Pruthi (unpublished work, 1962) to the presence of a leucoanthocyanin.

Pruidze (1965), while studying the physical and chemical properties of dry laurel leaves and stalks *(Lauris nobilis),* reported that the color of leaves (the content of chlorophyll) could not be used as a fundamental criterion of the quality of dry bay leaves. For the storage of dry leaves with 10 to 14% moisture content, they recommended a relative humidity of 75 to 80%.

Sivshchuk *et al.* (1968) obtained water-soluble chlorophyllins from dry mint. Kato (1965) reported the occurrence of capsanthin, violaxanthin, and β-carotene in mustard.

Koeppin and Vander Spuy (1961) investigated the microbial hydrolysis of flavonols of the inner scales of the brown-skinned onion *(Allium cepa)* varieties by analysis of an onion juice extract. The major compound was isolated and identified as quercetin 4-monoglucoside. The presence of a small amount of quercetin 3′-diglucoside and traces of quercetin and another glycoside, presum-

ably a quercetin 5-glucoside, were also demonstrated by paper chromatography. The inoculation of the onion juice with *Torulopsis holmii* and incubation for 6 weeks at 28°C resulted in complete hydrolysis of quercetin 4′-monoglucoside to quercetin and partial hydrolysis of quercetin 3-glucoside to isoquercetin. The sugar produced on hydrolysis was metabolized by the yeast.

Schwimmer (1967) reported that onion juice contains a bitter substance derived enzymatically from a nonbitter precursor. This bitter substance is nonvolatile, nonionic, and soluble in ether. It is not directly derived from the system responsible for onion odor or pungency. It may be a triterpenoid or a flavonoid, but it is not quercetin, the glycosides of which are found in the scales of yellow onions. Formation of bitterness in onion juice can be prevented without affecting odor intensity by a temporary acidification of the juice to below pH 3.9.

Shannon *et al.* (1967a) investigated the reactions involving and the conditions affecting the formation of pink pigments from precursors isolated from purées of white onion. The latter compound then reacts with H_2CO or naturally occurring carbonyls to form the pigment. The final pigment-forming reaction proceeds at about seven times the rate of the first reaction and has an optimum near pH 4.8. Pigment formation is inhibited by the SH group of cysteine. Studies with isotopes indicate that amino acids and H_2CO are incorporated in the pigment molecules. The rate and extent of pigment formation and the color of pigment formed were affected by the kinds of amino acids and carbonyls. A preparation of allinase (allin alkylsulfenate lyase) from garlic and a system of pyridoxal and copper, which simulates the catalytic action of alliinase, caused the formation of pigment precursors in the amino acid fractions from onions. The precursors formed were an ether-soluble, ultraviolet-absorbing compound and one or more unidentified carbonyl compounds. Reactions for pigment formation are proposed (*Shannon et al.*, 1967b).

Biffoli (1955) reported the absorption spectra of saffron extract in cold C_6H_6. Saffron had a maximum at 440 mμ, whereas Indian saffron had a maximum at 418 mμ with a minimum at 430 mμ.

Beauquesne and Delelis (1967) studied the flavonoids in shallot *(A. ascalonicum)*. An alcohol extract of the external brown paper-like layers of the bulb of the shallot was chromatographed on paper, and by thin-layer chromatography on polyamide, quercetol and its glucosides—spireoside, the 3,4′-diglucoside, and the 7,4′-diglucoside—were found. More quercetol and spireoside were found in *A. ascalonicum,* but 3,4′-diglucoside and 7,4′-diglucoside were more abundant in onion (*A. cepa*). The total amount of flavonoids was highest in the outer layers and decreased toward the center of the bulb (from 20% to 1%). The roots did not contain flavonic derivatives. Fresh leaves contained 1% flavonoids, but only spireoside and 3,4′-diglucoside were present. Starke and Herrmann (1976a,b) reported on flavones and flavonols in alliums.

According to Srinivasan (1953), the chromatographic resolution of the color-

ing matter of *Curcuma longa* on silica gel from C_6H_6 extracts of the powdered rhizome showed spearation into three major and three minor zones with C_6H_6 equilibrated with H_2O. Zones 1, 2, and 4 yielded curcumin, m. 182°C, R_f 0.27; *p*-hydroxycinnamoylferuloylmethane, m. 168°C, R_f 0.14; and bis-(*p*-hydroxycinnamoyl)-methane, m. 224°C, R_f 0.09, respectively. Zones 3, 5, and 6 contained geometric isomers of the three main constituents. Color reactions with $FeCl_3$, H_3BO_3, H_3BO_3-$C_2H_2O_4$, 1% NaOH, and concentrated H_2SO_4 are described. The maximum absorption and $E^{1\%}_{1cm}$ in EtOH and C_6H_6, respecitvely, are curcumin 420 mμ and 1640, and 410 mμ and 1640. The material in zone 3 showed 370 mμ and 620 in EtOH, and the substances in zones 3, 5, and 6, respectively, showed the following maximum absorption in C_6H_6: 365, 345, and 325 mμ. The purely diketonic forms of isocurcumins are not found in the natural product.

Du *et al.* (1974) isolated a new cyanadin-3-diglucoside from red onions and identified it by conventional chromatographic and spectral methods. Du and Francis (1975) reported the isolation and partial identification of the major pigments (anthocyanins) in garlic. Seven pigments (three major and four minor) have been noted.

Du *et al.* (1975) reported the anthocyanins in the seed coats of pomegranate (flesh) as 3,5-glucoside and 3-glucoside of cyanidin, delphinidin, and pelargonidin. Delphinidin pigments were not present in the peel, but the other four pigments were present.

H. Saponins and Sapogenins

Balansard and Flandrin (1945) described a method using NaCl for salting out the saponin from fenugreek, which gave a yield of 1%. Marker *et al.* (1947) found that the seeds of fenugreek contain 1 g of diosgenin, 0.1 g of gitogenin, and a trace of trigogenin per kilogram of dry seed. According to these authors, this is the only instance in which all three sapogenins appear in the plant. Bedour *et al.* (1964) also studied the steroid sapogenins in fenugreek. From the saponin-rich concentrate, diosgenin, trigogenin, and gitogenin were isolated along with the fourth product, m. 162° to 163°C, $[\alpha]_D - 179°$, which will be identified as 25D-spirosta-3,5-diene. No trigonellagenin was found. 25D-Spirosta-3,5-diene was an artifact derived from diosgenin. According to Varshney and Sharma (1966), EtOH extraction of the seeds of fenugreek yielded a mixture of two flavonoid glycosides, two flavonoid aglycons, and two steroidal saponins. Quercetin was isolated from the flavonoid fraction, whereas luteolin was identified by paper chromatography. The saponins on hydrolysis gave diosgenin and gitogenin in a 9:1 ratio.

Gladkikh *et al.* (1965) reported the presence of saponins in 553 species of plants of Turkmenia, which were studied by a hemolytic method. Saponins were

found in plants of ninety-five species belonging to twenty-two families. Saponin-containing plants were found the most often in the following families: Liliaceae, Iridaceae, Caryophyllaceae, Ranunculacere, Umbelliferae, and Primulacae. A high hemolytic index was determined in the following plants: *Allium capsium, A. regelli,* and *A. sabulosum.*

I. Alkaloids

Tomita *et al.* (1963) described the isolation of different alkaloids present in the bark of *Laurus nobilis.*

Voskerusa (1960) studied the morphine content of opium poppy capsules. He found that gray seed color and high morphine content were not related, as had been claimed earlier. Pfiefer and Heydenreich (1961) found significant amounts of the baine, but could detect only traces or very small amounts of morphine, codeine, narcotoline, and narcotine in germinating poppy seeds. According to Maturova *et al.* (1962), most *Papaver* species studied contained protopine, rhoeadine, muramine, reanthopetaline, and an alkaloid, m. 367°C. The alkaloid was present in all species studied except *P. alpinum burseri.* Kuehn *et al.* (1964) isolated mecambrine, protopine, a new alkaloid called "miltanthine," and six others from the nonphenolic bases. Armepavine and three other phenolic bases, coptisin, and palmatine were also found. Hanssen *et al.* (1965) reported the isolation and characterization of codamine in opium. Pfiefer and Banerjee (1965) isolated the red-colored alkaloids of the genus *Papaver*—namely, papaverubins A and E. Zsadon (1966) reported the preparation of narcotoline and morphine from poppy capsules, and Leterrier and Viossat (1967) reported the production of some poppy alkaloid free radicals by chemical means.

J. Sulfur Compounds

Peyron (1961) studied the precursors of certain sulfur compounds encountered with essential oils, including those of peppermint and cumin. To determine whether the sulfur compounds found in essential oils are natural or artifacts, alcohol extracts were chromatographed. Methionine sulfoxide was indicated in *Carum carvi* fruits. Methyl sulfonium methionine was found in the leaves of *Mentha piperita.* No sulfur amino acids were found in *M. arvenis* leaves. The essential oil of *M. piperita* contained dimethyl sulfide.

Nagasawa and Soga (1961) investigated the odorous substances (including sulfur compounds) in mint plants. *Mentha arvensis, M. piperita, M. spicata,* and their varieties were distilled with steam and chromatographed. H_2S, some ammoniacal compounds, and carbonyl compounds existed in the gas produced by steam distillation. San-Bir (a variety of *M. arvensis*) especially contained large amounts of H_2S. In addition, the existence of dimethyl sulfide was assumed. In

the volatile water of San-Bi, NH_3 and Me_3N were present, and the presence of Me_2NH and $MeNH_2$ was also probable. Some of the unpleasant odor of San-Bi was due to the presence of the ammoniacal compounds such as Me_3N.

Sugii *et al.* (1963) studied the biosynthesis of *S*-methyl-L-cysteine and *S*-methyl-L-cysteine sulfoxide from methionine in garlic. In addition to these sulfur compounds, cycloaliin and aliin were also isolated. Saghir *et al.* (1964) determined the aliphatic mono- and disulfides in *Allium* species by gas chromatography and their distribution in common food species. They also discussed the relation of these compounds to allicin and to the odors of *Allium* species. Markov (1964) developed a micromethod for the quantitative determination of sulfur in glucosides of garlic. Wannamaker (1966) reported the results of a study on the influence of the addition of sulfur to the soil on the magnesium, potassium, sodium, sulfur, phosphorus, copper, zinc, boron, and iron content of the onion.

Pruthi and associates (1958, 1959b–f, 1960b) and Singh *et al.* (1959a,b,c) studied the stability of allicin (allyl sulfoxide) during the manufacture and storage of garlic powder. Shankaranarayana *et al.* (1974b), in a critical and exhaustive review on volatile sulfur compounds in food flavors, also discussed the occurrence and nature of sulfur-containing volatile compounds in some spices, notably in *Allium* species (chives, garlic, leek, and onion), asafetida, celery, ginger, horseradish, mustard, and parsley. They are summarized in Table XL.

K. Enzymes

Kaneko (1960a) conducted biogenetic studies relating to the biosynthesis of anethole by fennel *(Foeniculum vulgare)*. The origin of the side-chain carbon atoms and the MeO carbon of *p*-$MeOC_6H_4CH{:}CHMe$ (I) was determined by the use of DL-phenylalanine-2-^{14}C (II) and DL-methionine-Me-^{14}C (III). Steam distillation of the plant material and ether extraction of the steam distillate yielded 0.95 g of I, maximum 268 mμ. Compounds II and III were incorporated into the side chain and the MeO group, respectively, in the sample of I produced *in vivo* by *F. vulgare*. Kaneko (1960b) further observed that the cell-free enzyme system of *F. vulgare* synthesized anethole from phenylalanine under anaerobic conditions at pH 7.0 when the incubation mixture contained adenosine triphosphate, diphosphopyridine nucleotide, $MgCl_2$, methionine, 2-oxoglutarate, nicotinamide, and glutathione. Malate, citrate, and glutamate stimulated the synthetic activity slightly, whereas malonate inhibited it. Ascorbase stimulated the synthetic activity greatly and probably maintained a reductive state for the enzyme system. The synthesis was inhibited also by *p*-hydroxycinnamic, cinnamic, and phenylpyruvic acids, which were presumed to be intermediate in the biosynthesis. In another study, Kaneko (1961) synthesized *p*-$OHC_6H_4$14CO_2H (I) and fed it to this plant. The anethole (II) isolated was found to be labeled in the methyl group of the side

TABLE XL THE OCCURRENCE AND NATURE OF VOLATILE SULFUR COMPOUNDS IN SOME SPICES[a]

Compound	Formula	Spices
1. Thiols (mercaptans)		
Methanethiol	$CH_3—S—H$	Onion, garlic, leek
2. Sulfides (thioethers) Sym		
Dimethyl sulfide	$CH_3—S—CH_3$	Onion, garlic, leek
Diallyl sulfide	$CH_2{=}CHCH_2SCH_2CH{=}CH_2$	Onion, garlic, leek, chive
3. Disulfides Sym		
sec-Butyl propenyl disulfide	$CH_3CH_2CH(CH_3)—S—S—CH{=}CHCH_3$	Asafetida
4. Trisulfides Sym		
Dimethyl trisulfide	$CH_3—S—S—S—CH_3$	Garlic, onion
Diallyl trisulfide	$CH_2{=}CHCH_2—SSSCH_2CH{=}CH_2$	Garlic, onion
Asym		
Methyl allyl trisulfide	$CH_3SSSCH_2CH{=}CH_2$	Garlic, onion
Methyl-*n*-propyl trisulfide	$CH_3S—S—S—C_3H_7$	Onion
5. Tetrasulfides		
Diallyl tetrasulfide	$CH_2{=}CHCH_2—S—S—S—SCH_2—CH{=}CH_2$	Onion
Diallyl tetrasulfide	$CH_2{=}CHCH_2—S—S—S—SCH_2\ \ CH{=}CH_2$	Onion
Dimethyl tetrasulfide	$CH_3—S—S—S—S—CH_3$	Onion

6. Thiocyanates		
Allyl thiocyanate	$CH_2{=}CHCH_2SCN$	Horseradish, onion
7. Isothiocyanates		
Allyl isothiocyanate	$CH_2{=}CHCH_2N{=}C{=}S$	Black mustard (*Brassica nigra*)
p-Hydroxybenzyl isothiocyanate		White mustard (*Brassica alba*)
3-Butenyl isothiocyanate	$CH_2{=}CHCH_2CH_2N{=}C{=}S$	Brown mustard (*Brassica juncea*)
8. Sulfenic acid		
1-Propenyl sulfenic acid	$CH_3CH{=}CH{-}S{-}OH$	Onion
9. Thiosulfinates		
Dimethyl thiosulfinate	$CH_3{-}SOSCH_3$	Onion
Allicin (diallyl thiosulfinate)	$CH_2{=}CHCH_2{-}SOS{-}CH_2CH{=}CH_2$	Garlic
10. Thiosulfonates		
Methyl methane thiosulfonate	$CH_3SSO_2CH_3$	Onion
Propyl propane thiosulfonate	$C_3H_7SSO_2C_3H_7$	Onion
11. Miscellaneous		
Thiopropanal-*s*-oxide	$CH_3CH_2CH{=}S{-}O$	Onion
Carbon disulfide	CS_2	Parsley
Sulfur dioxide	SO_2	Onion, garlic

[a] From Shankaranarayana *et al.* (1974b).

chain, indicating that I is incorporated into II without rearrangement. Kaneko (1962) further studied the biosynthesis of anethole by *F. vulgare*.

Courtois and Percheron (1961) extracted an α-galactosidase from germinated seeds of fenugreek. This enzyme, with an optimum pH of 4.6, hydrolyzed heterosides and oligosides containing galactose and partially attacked galactomannan from fenugreek to yield galactose. Moreover, it possessed transferring activity of the α-D-galactopyranosyl group to hydroxylated acceptors.

Bhatia *et al.* (1955) studied carbohydrases in garlic bulb; they consist mainly of invertase and polyfructosidase. No definite evidence could be obtained for the presence of transfructosidase in garlic, which thus contrasts sharply with other glucofructosan-bearing plant tissues. They further observed that the high concentration of invertase masked the presence of transfructosidase in garlic. Watanabe *et al.* (1963) reported that an aqueous extract of garlic contained phosphatase in addition to allinase and an unknown substance that prevented the oxidation of vitamin C even in the presence of copper.

Capelle and Monique De Serres (1967) studied the distribution of arginase in *Allium* species. Arginase activity was determined in four growth phases of *A. sativum, A. cepa, A. ascalonicum, A. porrum,* and in three onion strains. Activity appeared at cultivation time of the bulbs. In most of the cases, the bulbs were devoid of activity. Contrary to findings with *Jacinths* and *Lilium,* chlorophyll-containing leaves were also devoid of arginase activity.

Lu and Whitaker (1974) studied some factors affecting rates of heat inactivation and reactivation of horseradish peroxidase.

Dobrynina (1958) studied the inactivation of enzymes in plants by freezing and γ-irradiation. Catalase, ascorbic acid oxidase, polyphenol oxidase, and peroxidase activity were determined in fresh, frozen, and combined frozen-irradiated celery and green onion. The degree of inactivation of the enzymes suggests that freezing, followed by irradiation, gives better enzyme inactivation than freezing alone.

Tsuno (1958a) observed that allinase from *A. bakeri* and *A. odorum* released pyruvic acid. Allinase acts best at 37° to 40°C, pH 5.6 to 6.4. Activity of 100% was measured within 3 minutes. Magnesium ions activate and KCN inhibits the reaction. Allinase is relatively stable; two weeks' standing in a cold room did not decrease its activity.

Kupiecki and Virtanen (1960) investigated the cleavage of alkyl cysteine sulfoxides by an enzyme in onion that is specific for alkyl cysteine sulfoxides. They extracted and partially purified it from *A. cepa*. The enzyme appears to require pyridoxal phosphate as a cofactor, but it shows no stimulation in activity by various metals. It shows a very low order of inhibition by various sulfhydryl group reagents. Electrophoresis shows a single protein peak at 2 pH levels.

Schwimmer *et al.* (1960) demonstrated allinase in proteinase preparation from onion.

Hewitt (1963) studied the enzymatic enhancement of flavor in onions and cabbage. Protein fractions from fresh onions were active on *S*-alkyl cysteine sulfoxide and enhanced the flavor of dried onions. Mustard and cabbage enzymes enhanced the flavor of dehydrated cabbage. Carson (1967) observed that the flavor precursors or substrates for enzymatic action in onion and garlic were sulfoxide amino acids. The most important volatile flavor compounds from onion contain sulfur and include H_2S, thiols, disulfides, trisulfides, thiosulfinates, and the elusive lachrymatory factor. Of the various gas chromatographic procedures for determining the volatile flavor of onions and garlic, the most promising is the electron-capture hydrogen-flame dual-channel method.

Benn (1962) preseneted experimental evidence to show that α-amino acids are biogenetic precursors of the mustard oil glycerides, which, by enzyme-induced Lossen-type rearrangement, produce the mustard oil by various reactions.

Mackay and Hewitt (1959) also studied the application of flavor enzymes to processed foods and compared the effect of flavor enzymes from mustard and cabbage on dehydrated cabbage.

Halbert and Weeden (1966) observed lipolytic activity in black pepper, paprika, ground cinnamon, cayenne pepper, and ground ginger, and found it to be markedly greater in black pepper than in others. The lipolytic activity in black pepper was mainly in the outer pericarp, was insoluble in water, and had an optimum temperature of 46°C. Gross and Ellis (1969) also studied lipase activity in spices and seasonings. They observed that off-flavors develop, owing to formation of free fatty acids by hydrolysis of triglycerides. A soapy taste results when this reaction is catalyzed by acid, alkali, high temperature, and lipolytic enzymes. To avert the possible development of off-flavor when spices are used along with fats and oils in processed foods such as baked goods, the use of spice extractives in place of black or white pepper and thyme is suggested.

According to Pequin (1961), etiolation increased both catalase and peroxidase activities in red pepper and eggplant, although it did not change the distribution of activity in various tissues.

Schenk *et al.* (1962) studied the enzymatic and nonenzymatic oxidation of a therapeutically active plant substance—morphine in poppy capsules *(Papaver somniferum)*. The maximum losses of morphine in fresh, crushed poppy capsules occurred in the first 8 days of storage at 20°C; after 24 days, no decrease in morphine and no further activity of phenoloxidase were discerible. In a model experiment, phenoloxidase reduced morphine by 5% in 5 hours in the extract from poppy capsules (in the absence of *p*-coumaric acid and caffetannic acid). The action of phenoloxidase in which *p*-coumaric acid and caffetannic acid served as carriers was responsible for the losses of morphine that occurred during the gathering and storage of opium and poppy capsules.

Bhat *et al.* (1954) found that asafetida had no significant effect on the activities of salivary amylase, pepsin, trypsin, renin, pancreatic amylase, and pancrea-

tic lipase. Patwardhan and Sastry (1957) also reported that asafetida had no effect on intestinal enzymes.

Bhat and co-workers (1953) found that saffron and nutmeg stimulated the activities of pepsin and renin. Thus, it is evident that some spices promote digestion by increasing the secretion of digestive juices by way of stimulating the enzymatic activities.

L. Vitamins

Considering the quantities of spices normally used in flavoring foods, spices do not contribute much to the food value as measured in terms of proteins and carbohydrates. They may, however, increase the vitamin content of some foods to some extent, as will be briefly reviewed here.

By employing chemical and microbiological methods, Brown *et al.* (1946) reported nicotinic acid in caraway, poppy, etc.

Watanabe and Goto (1954) examined free and bound biotin in several kinds of crude drugs including cinnamon by the microbiological assay method. Gosh and Rajagopal (1951) reported that coriander seeds were especially rich in riboflavin (350 μg%).

According to Covello (1943), raw, dry fenugreek seeds contain about 150 μg% of trigonelline and practically no nicotinic acid. If the seeds are roasted sufficiently to brown, then about two-thirds of the trigonelline is converted into nicotinic acid. Lopez *et al.* (1950) reported that fenugreek seed was one of the richest sources of trigonelline (0.1274%), a methyl betaine derivative of nicotinic acid, which makes it important in terms of vitamin content. Rajlakshmi *et al.* (1964) made a study of the amount of free and total niacin in grain-based foods popular in Egypt and India, and changes in available and total niacin as a result of cooking, roasting, fermentation, and germination. Roasting resulted in an increase in both free and total niacin except in the case of corn, which showed an increase only in the niacin. Fenugreek seed, which contains trigonelline and is popular in the Egyptian diet, was roasted in a shallow pan over an open fire. An increase in free and total niacin was noted. Various mixtures of the grains with and without raw, germinated, or roasted fenugreek seed were used in the typical preparation of the following Indian and Egyptian foods: dosa, dhokla, idli, and chapaties. Some foods were prepared with fermented dough. In every case, there was an increase in total and free niacin after cooking. The addition of 5% fenugreek to corn resulted in a 30 to 35% increase in free niacin in the cooked products. Fermentation resulted in a significant increase in both free and total niacin. There was some loss when the fermented dough was cooked, but the finished product still compared favorably with the unfermented dough. The effects of pickling, boiling, and storage in an acid medium were also tested on

samples of fresh and pickled lemons, and on fenugreek leaves cooked and stored in tarmarind juice. There were definite increases in available and total niacin.

Tsuno (1958b) reported total and free thiamine content of several *Allium* plants. Garlic had the highest content of total thiamine; onion had much less. Gusev and Grishina (1963) reported that vitamin C varied from 15 to 20 mg per 100 g in different varieties of garlic. Steponaviciene (1964) determined the amount of vitamin C by the hydrogen sulfide method in leaves and roots of twenty-eight herbs and condiments. Ishiguro (1963) reported that stone leek, garlic, leek, and cauliflower were rich in pantothenic acid. Suhara (1962) detected thiothiamine in onions.

Schillinger and Zimmermann (1965) reported the stability of thiamine, riboflavin, nicotinic acid, ascorbic acid, and β-carotene in freeze-dried and heat-dried vegetables, including parsley and leeks, stored *in vacuo* and in nitrogen at room temperature (relative humidity 35 to 60%) for 3 years.

III. CHEMICAL COMPOSITION OF SPICE ESSENTIAL OILS AND OLEORESINS

The literature on spice essential oils and oleoresins is too voluminous to be compressed into this review. Guenther (1948–1952) covered the subject in six volumes of his standard book on essential oils, including spice essential oils and oleoresins. Guenther and his associates (1959, 1961, 1963, 1965, 1967, 1969, 1971, 1973, 1975, 1977) have further discussed the subject in a series of systematic biennial reviews. Kulka (1962) has ably covered the chemistry of essential oils. The antimicrobial activity of spice essential oils and their volatile constituents as well as the preservative action of a number of spice essential oils, etc., has already been reviewed in Chapter 2 (Sections II and IV). Analytical methods for essential oils have been reviewed by Duquenois (1962), La Face (1962), Seher (1962), and Nigam and Kumari (1963), and their chemical aspects, etc., by Sterrett (1962b) Parczewski (1962), Baslas (1967, 1968), and Baslas and Baslas (1968a,b). Baboob and Bharmaramba (1963) and Pruthi *et al.* (1978c) conducted studies on Indian cinnamon leaf oil. For other details on essential oils, see Section VI of Chapter 6.

5

Postharvest Technology of Spices

Spices, like all other agricultural commodities, invariably contain high moisture (55–85%) at the time of harvest, which must be brought down to 8–12%. Futhermore, spices vary considerably in shape, texture, size, color, etc., as they may be fruits, barks, seeds, leaves, rhizomes, roots, unopened flower buds (cloves), and other floral parts (saffron). Hence their pretreatments, curing, and methods of processing also vary considerably. During their processing after harvest, they are subjected to different types of unit operations such as washing, peeling, curing, drying, cleaning, grading, and packaging, until they are ready for the consumer or for the market. Such postharvest processing technology should ensure proper conservation of the basic qualities for which they are valued—aroma, flavor, pungency or bite, color, etc.

It is the prime requisite that all spices be harvested at the correct stage of maturity without much physical damage, after which they are processed properly for the market. In most cases, they are sun dried at the farm and transported to an appropriate center for further processing. Processing operations, of course, vary with the individual spice concerned. Shankaracharya and Natarajan (1973c) have discussed the problems of postharvest technology of spices.

I. PRETREATMENTS

Postharvest treatment of black pepper as practiced in different parts of the world have been reviewed by Larcher (1967), who, after presenting the results of further tests, has suggested the technique of 48 hours in the shade as the best

pretreatment for the manual separation of berries and the highest yield of essential oil content (4.96% on a dry-weight basis) and piperine content. The boiling-water treatment was found to be not useful, as it alters the chemical composition of the oil.

A. Washing

Root and rhizome spices such as angelica, calamus, ginger, and turmeric, which are dug out of the soil, need washing to remove the adhering mud and dirt. Pressure-water washing may be possible, wherever facilities are available. Reduction in the microbial load will also be influenced by the efficacy of this operation.

B. Peeling

Almost all spices are dried whole (without peeling), with the exceptions of ginger, onion, and garlic. Since the skin of the ginger rhizomes constitutes a barrier to evaporation or transportation of moisture from within, a prerequisite for efficient drying of ginger hands is peeling; hand-peeling with special scraping knives is resorted to. Partial mechanical peeling of ginger has been tried with abrasive machines for 60 seconds, which is equivalent to hand-scraping with respect to loss of volatile oil, time of drying, and peeling loss of 10 to 12%. The timing of abrasive peeling is rather critical, and care should be taken; otherwise it results in heavy losses. Another method of reducing the time of drying of ginger and turmeric is to slice them before drying, but the conventional trade will have to be educated and assured about its economy and the negligible chances of adulteration in the sliced and dried product (Natarajan *et al.*, 1972; Kuppuswamy, 1974).

Cinnamon and cassia barks are peeled off the tree branches (2 to 3.5 cm thick) with the help of special curved knives, cut into convenient sizes before they are cured, and dried as quills, quillings, featherings, and chips (Pruthi, 1976, 1979; Pruthi *et al.*, 1974a,c, 1975, 1978c).

C. Pricking

Pricking the skin of chilies longitudinally helps to reduce the time of drying (Laul *et al.*, 1970).

D. Blanching

Blanching is almost universally practiced in vegetable dehydration chiefly for the inactivation of enzymes, but exceptions are celery, garlic, onion, and parsley, which are valued primarily for their specific flavors, since blanching

destroys the very enzymes involved in the generation of their respective flavors. In chilies and ginger also, blanching is not desirable (Natarajan *et al.*, 1970, 1972).

However, blanching or simmering is a commercial practice in the case of turmeric, particularly in the presence of a small quantity of alkali (Desikachar *et al.*, 1959).

E. Chemical Treatments

Many types of chemical treatments such as alkali treatments, antioxidant treatment, bleaching by liming treatment, sulfuring by sulfur fumes, and sulfitation or treament with SO_2 or hydrogen peroxide are employed by the industry for different spices—for example, alkali treatment for cardamom, liming or bleaching of ginger, bleaching of cardamom by sulfuring or sulfitation, and curing of turmeric with appropriate chemical solutions, as discussed briefly below.

1. Alkali Treatment

The attractive green color of cardamom, which is due to chlorophyll, can be stablized to a great extent by steeping the cardamoms in 2% sodium carbonate solution for 10 minutes (Natarajan *et al.*, 1967a, 1968a). Alkali treatment has also been found useful in drying of chilies, particularly in conjunction with olive oil (Laul *et al.*, 1970). Lime treatment is used for bleaching of ginger, as will be discussed briefly later.

2. Antioxidant Treatment

The attractive red color of capsicums or chilies, which is due mostly to carotenoids (notably capsanthin, capsorubin, etc.), is stablized to a great extent by treatment with a suitable antioxidant (Van Blaricum and Martin, 1951; Lease and Lease, 1956b). Van Blaricum and Martin (1945) and Lease and Lease (1956a) have also studied the effects of other factors such as initial composition, light, air, temperature, condition (whole or ground), harvesting practices, and predrying treatments on the retention of color in cayenne pepper during drying and storage.

3. Bleaching of Cardamom and Ginger

a. Cardamom. There is some demand for bleached cardamom, although mainly it is sold with its natural green color. In fact, cardamom capsules that do not have a uniform green color are less valuable, and such capsules are put through a bleaching process to produce a uniform color. Bleaching powder, sulfur dioxide, or hydrogen peroxide is generally used for bleaching. Sulfur dioxide is obtained either through burning sulfur or by wet sulfitation—that is, steeping in dilute solutions of water-soluble salts of SO_2. Bleaching is also done

by hydrogen peroxide. Bleaching has been developed into a highly scientific and paying profession in Sweden. In South India too (West Coast and Mysore State), bleaching of cardamom is carried out (Indian Council of Agricultural Research, 1967; Marketing Research Corporation of India, 1968). However, SO_2-bleached cardamom samples have shown variations in SO_2 content from 20 ppm to 3 ppm (Indian Institute of Foreign Trade, 1967). There is, therefore, need for standardization of the method of sulfuring or sulfitation.

b. Ginger. The peeled ginger hands are washed in water and kept steeped in plain water for 2 to 3 hours. Thereafter, they are taken out and steeped in about 1.5 to 2.0% lime (CaO) solution for about 6 hours. They are then drained and sun-dried on mats, barbecues, or a clean cement floor. This liming or bleaching of ginger not only improves its color, but also helps to preserve it better. Care should be taken to use the best-quality slaked lime in order to get better whiteness (Rodriguez, 1971).

The application of these chemicals requires proper control, especially in the bleaching of cardamom and ginger. It is necessary to know the extent of SO_2 pickup by cardamom during bleaching. Wet bleaching involves rehydration, and therefore careful control of the final moisture content is essential for prevention of insect infestation during storage. It is thus necessary to prescribe scientifically controlled procedures to obtain uniformity in product batches and at the same time to safeguard against the indiscriminate use of such chemicals.

F. Curing and Other Treatments

Different spices such as cinnamon and cassia, garlic, saffron, turmeric, and vanilla beans are cured by different techniques in order to generate the characteristic aroma or flavor for which they are valued.

1. Cinnamon and Cassia

The peeled slips (bark) from cinnamon and cassia branches are gathered and packed one above the other with their concave and convex surfaces in juxtaposition until the packings measure about 20 to 30 cm wide and 30 to 45 cm long. These slips are piled up within enclosures of sticks and wrapped up in mats. The packs are kept overnight in that condition for curing or for allowing so-called "fermentation." Actually no real fermenation process develops. A little softening of the bark does occur, with the result that the peels become more easy and pliable for the subsequent piping operation or the removal of the epidermis and the green cortex (Fazlullah Khan, 1967).

2. Garlic—Smoke-Curing

The better quality of smoked foods is due to (1) partial dehydration, (2) incorporation of antioxidants, (3) impregnation of antiseptic constituents, (4)

effect of heat on microorganisms, and (5) improvement in organoleptic quality. Srivastava and Mathur (1956) discussed the benefits of the smoking process of garlic. The physicochemical changes during storage of smoke-cured garlic have also been described and discussed.

3. Saffron Curing

The value of saffron depends heavily on the methods by which the stigmas are processed (Madan *et al.*, 1966). In Spain, where the process is called toasting, the stigmas are placed in sieves in layers 2–3 cm thick. These sieves are placed 15 cm above an almost spent fire for drying. By stacking them and by changing their order or position, the product is carefully dried (Arjona, 1945). Great care must be taken that the stigmas are protected against dampness as well as light, because light bleaches saffron to a dull yellow color. Drying in the sun or over smoke also bleaches the saffron or changes its color (Katyal, 1967).

4. Curing of Turmeric

Curing of raw turmeric rhizomes and fingers freshly dug out of the earth is essential for the development of the attractive yellow color (mostly due to curcumin) and aroma; without curing, it lacks both. Curing is begun 3 or 4 days after harvesting. The mother rhizomes and fingers are separated by hand. If necessary, the former are kept for seed purposes and the latter are cured for marketing by the following techniques.

a. Conventional Technique. The fingers and bulbs are boiled in water until a froth comes out and white fumes start to appear, giving the typical turmeric aroma. Earlier, cow-dung extract was added to boiling water to make it alkaline. The fingers and bulbs become softer and yield when pressed between the fingers. At this stage, they are removed from the boiling water, drained properly, colored artifically if necessary, and dried in the sun for 10 to 15 days, after which they are polished to remove any adhering rootlets and scales.

The quality of the final product, including its color and aroma, depends largely on the correctness of curing. Another improved country method has also been described (Directorate of Arecanut and Spices Development, 1970), but according to Anjaneyulu and Krishnamurthy (1968) the curing quality of turmeric is mostly a varietal character, although other factors such as high moisture content, maturity, and seasonal conditions influence the percentage of recovery (the ratio of the cured turmeric to raw turmeric) to some extent. They further claim that short-duration Kasturi turmeric *(Curcuma aromatica)* types recorded the highest percentage (24 to 26%), medium-duration Kesari turmeric *(C. longa)* types gave the lowest values (14 to 20%), and long-duration *(C. longa)* types showed medium values (21 to 24%). Mother rhizomes recorded a higher percentage than the corresponding fingers. The curing percentage increased with increasing maturity of the rhizomes. Raw rhizomes with higher initial moisture content yielded both a poor-quality product and a lower curing percentage.

b. Improved Scientific Method. Desikachar *et al.* (1959) developed a new scientific method according to which the washed raw tubers are boiled in dilute lime water, sodium carbonate, or sodium bicarbonate solution instead of in water. For imparting the characteristic yellow color to the tubers, a solution containing 20 g of sodium bisulfite and 20 g of concentrated hydrochloric acid per 150 pounds (or about 70 kg) of boiled tubers has been recommended in place of the Chemichrome solution used earlier in the country practice, which by now is no longer popular.

5. Curing of Vanilla Beans or Pods

At the time of harvest, in the vanilla fruit or pods, the normal components forming the characteristic vanilla flavor do not exist in their final form, but appear only following a curing (fermentation) process involving enzymatic actions on glucosides, the most important of which is glucovanillin, which produces vanillin (the main aromatic component of vanilla) and sugar as a result of the action of β-glucosidase. Similarly, aldehydes, protocatechoic acid, benzoic and vanillic acids, anisic alcohol, etc., are also formed. These different compounds impart subtlety to the fragrance of natural vanilla, enabling it to be distinguished from synthetic vanillin (Theodose, 1972).

Different curing methods are used in different producing countries such as Madgascar, Mexico, Tahiti, Guadeloupe, Bourbone, and India. All these methods of preparation used are characterized by the following different stages:

1. Cessation of the vegetative life of the bean to allow the onset of enzymatic reactions.
2. Raising of the temperature to promote this action and to stimulate at first quite rapid drying, thereby preventing the appearance of harmful fermentation.
3. Slower drying followed by the production of different fragrance components.
4. Conditioning of the product, during which the operations geared to commercial presentation and to obtain good preservation are carried out.

Theodose (1972) has systematically reviewed and discussed these methods and, on the basis of his own research, has described an improved method that cuts labor by 80% and furthermore gives a 6 to 8% higher yield with higher vanillin content and better quality. The improved method consists in sorting or grading, scalding, autoclaving, loading, and mechanically drying in a tunnel dryer, followed by drying in the shade.

The following new improvement is further designed by Theodose (1972) to give excellent results.

After scalding and autoclaving, the vanilla beans are chopped into 2- or 3-cm pieces. This leads to an acceleration of the oxidative and enzymatic reactions.

The "cuts" are put through the dryer at a temperature of 65°C each day. As soon as they come out, they are put into an isothermal chest for the next 24 hours, their temperature being maintained at around 50°C. These operations are repeated for 12 days, after which a product containing 20 to 25% moisture is obtained, with a yield of about 4.5 to 1.0. The transfer to the isothermal chests after the tunnel drying is vital, since, in addition to accelerating the enzymatic action, it allows the various cuts, some of which have a tendency to dry quickly, to equiliberate their moisture content; otherwise, with an acceleration of drying, a product with poorly developed aroma and a smell almost of prunes is obtained. The only drawback of the method is its likely social repercussion, as it eliminates 80% of the labor required in the conventional techniques. Otherwise, this new process is capable of producing 3 tons of vanilla per day, and thus it constitutes a significant breakthrough, especially for vanilla-producing countries with heavy rainfall. Plans for installations and comparative costs of production by different techniques have also been discussed.

Kannan and Pillai (1966), after conducting some trials in India on different methods, suggest the Mexican process of alternatively sweating and drying, according to which the shriveled vanilla beans, 3 to 4 days after harvest, are immersed in hot water (60°C) for 1 minute and spread out on woolen blankets for sun drying. When the beans become too hot to hold in the hand, the blanket is folded over them and kept there for the rest of the day. At night, the beans are kept in "sweating boxes" lined with blankets. The next day, the beans are again put on the blankets and dried in the sun. The process is repeated for about 8 to 10 days, depending on weather conditions. By this time, the beans have lost most of their moisture, acquire a dark chocolate brown color, and develop a typical vanilla aroma.

Nair and Mathew (1969), after conducting their experimental trials on different methods of curing of vanilla as adopted in different vanilla-producing countries, also found the above Mexican process the best.

However, the Mexican process requires a series of sheds or buildings for "sunnings" and "sweatings" and "mahagony sweating boxes" or "aging boxes." The total process takes about 5 to 6 months, including aging. Finally, the beans are graded, bundled, and packed in special tins lined with waxed paper, as reviewed by Shankaracharya and Natarajan (1973b).

II. DRYING OF SPICES

A. Sun Drying

Drying constitutes the most important processing step in postharvest technology. The main object of drying is to reduce the moisture naturally present at the

time of harvest to a safe limit of, say, 8 to 10%. At this moisture level, the chances of insect and mold infestation are minimized, and thus the commodity (spice) can keep well. For mainly economic considerations, most of the spices are sun dried in the producing and developing countries, although admittedly the sun-drying operation has the disadvantage of possible contamination by microorganisms from the soil. Also, the quality and sometimes the color and flavor of sun-dried spices are not very uniform. However, if sun drying is carried out on raised platforms, or on barbecues or racks suitably designed to expose the products to the sun better, at least microbial contamination through the soil can be considerably reduced. On these raised platforms or racks, the drying rate is also better, owing to the draft of air passing through the bottom and sides.

1. Sun Drying of Cardamom

Vijayan (1974) reported the results of his trials on sun drying of cardamom treated with 2% sodium carbonate in a special type of glass chamber, which could exclude rain and mist and at the same time provide necessary ventilation. The product was compared with that obtained by the conventional techniques employing smokeless flues and direct sun drying. No significant difference was found among the three methods of drying, although a slight tarnishing of green color was noticed in solar drying in spite of the usual alkali treatment. Loss of moisture was also slightly more in flue drying. A major saving in labor could be obtained by using the solar-drying technique. However, there is need for development of still better treatment for the preservation of the natural green color.

2. Sun Drying of Chilies

In Japan, sun drying of chilies is followed by mechanical drying, dressing, and again sun drying before packing. The entire stem or plant with the fruits is cut and hung from bars and then exposed to the sun for partial sun drying. After the removal of 80% moisture, the parts are further dried in a dryer to the critical moisture level. The dried parts are removed with the help of a dressing machine and finally again dried in the sun before packing. This efficient combination of sun drying and dehydration may work well in such cases. The main problem is the heterogeneity in the product with special reference to size, color, and quality. Grading may help in getting better results. Improved agronomic practices are also required to get a more uniform product at picking time.

Sun drying of chilies has also been studied in some detail in India. Whole chilies spread on perforated rectangular aluminum trays (5 kg/m^2) took about 15 days to dry at a room temperature of 20 to 25°C and relative humidity of 34 to 50%. Pricking the chilies longitudinally reduced the drying time to 12 days, and blanching reduced it to 7 days. In the case of "checking," the drying time was 7 days after dipping in 2.5% potassium carbonate solution, and it was further reduced to 6 days in the presence of deodorized olive oil (Laul *et al.*, 1970).

3. Sun Drying of Garlic

Pruthi *et al.* (1959d) observed that in sun drying of garlic (flaked), the losses in allyl sulfide, total sulfur, antibacterial activity, and aroma were the highest compared with losses in four different methods of dehydration (artificial drying).

4. Sun Drying of Ginger

In the author's experiments, hand-peeled and abrasive-peeled ginger lots with and without liming treatment were dried in the sun on raised platforms, which yielded satisfactory products (bleached and unbleached ginger) in 7 to 9 days with a final moisture content of 7.8 to 8.8%. Raina *et al.* (1978) conducted studies on the sun drying and dehydration of ginger grown in Himachal Prudesh (India) and concluded that it was slightly inferior to Kerala ginger in respect of volatile oil and oleoresin content, but superior to that from other hilly areas.

5. Sun Drying of Pimento

At present, sun drying of Jamaican pimento or allspice berries is done on a concrete barbecue. Because of frequent shifting of the berries in and out of the sheds during rainy days, many berries break. Hence, mechanical drying is now preferable, and it is also more economical than sun drying (Breag *et al.*, 1972).

6. Sun Drying of Other Spices

Saffron discolors during sun drying (Katyal, 1967). Photosensitive spices like cardamom, and to some extent turmeric, bleach in the sun owing to the oxidation of chlorophyll and carotenoids naturally present in them. During the rainy season, however, it is essential to resort to mechanical or artificial drying, which is briefly discussed below.

B. Mechanical Drying

In order to avoid dependence on the vagaries of weather and also to reduce microbial contamination, natural convection dryers or forced-draft dryers can advantageously be used to get a better quality product. An air temperature of 60° to 70°C gives a drying time of about 5 to 9 hours, depending on the commodity, particle size, and method of drying (Ramanathan and Srinivasa Rao, 1974).

1. Factors Affecting Quality of Dehydrated Product

The quality of the dehydrated product is affected by a number of factors, including the quality or nature of the raw material, its method of preparation or processing treatments, the density of loading, the time, the temperature, and the method of drying or dehydration (Pruthi *et al.*, 1959c,d). Of these factors, the temperature of the air used during dehydration greatly affects not only the time required for dehydration, but also the quality of the finished product. In order to

secure large capacity and minimum operating costs, it is essential to use the highest temperature that will not materially injure the product. The optimum drying temperature and also the critical temperature of a product vary with the nature of the product and its moisture content (Pruthi *et al.*, 1959b).

a. Critical Temperature of Dehydration. The critical temperature is defined as the temperature at which a nearly dry product is seriously injured when exposed for a certain period of time. At temperatures above the critical temperature, the dried product is likely to scorch, the sugars present are likely to caramelize, and the color, flavor, and aroma of the product are likely to be adversely affected. To determine precisely the critical temperature of a product, the stability of its most desirable character(s) is followed up during heating at different temperatures.

In the absence of any published information, Pruthi *et al.* (1959b) systematically studied the critical temperature of dehydration of garlic. Based on the data on the retention of antibacterial activity, allicin, allyl sulfide, color, and flavor in fresh and dried garlic flakes under different temperatures of dehydration from 30° to 70°C, the critical temperature for dehydration of garlic was found to be 60°C. J. S. Pruthi and R. Susheela (unpublished work, CFTRI, Mysore, 1960–1962) observed pink discoloration in ginger from Assam when it was heated beyond the critical temperature of 60°C. Work carried out in Australia by Richardson (1966) has shown that 135°F is the highest temperature at which ginger for the spice market should be dehydrated, but if the ginger is to be used for extraction purposes, then temperatures up to 180°F are satisfactory. On the other hand, Natarajan *et al.* (1972) found a perceptible darkening of ginger at temperatures above 60°C (140°F). There was no significant difference in the color of samples dried at temperatures between 50° and 60°C, indicating thereby that the critical temperature of ginger was 60°C. Raina *et al.* (1978) also confirmed that the critical temperature for dehydration of Himachal ginger was 60°C.

Reports on the critical temperature of dehydration for chilies are rather conflicting. According to Murthy *et al.* (1968), the critical temperature for the dehydration of chilies is 46°C. In the author's view, 46°C is too low a temperature for economic dehydration. It takes 40 to 42 hours for the chilies to dry at this temperature in an oven, and even with optimal airflow the drying time is 26 to 28 hours (Kuppuswamy, 1974). There is no appreciable darkening of chilies at temperatures up to 140° to 150°F (Natarajan, 1974). These observations are in line with the findings reported from the South Carolina Experiment Station (Lease and Lease, 1962).

b. Methods of Dehydration. The method of drying or dehydration is another important factor affecting the quality of a dehydrated product. Natural convection dryers or forced-draft dryers can advantageously be used. Pruthi *et al.* (1959d) studied the comparative effects of four mechanical methods of dehydration—namely, hot-air drying by cross-flow or by through-flow sepa-

rately, freeze drying, and vacuum-shelf drying—versus sun drying on allyl sulfide, total sulfur, volatile reducing substances, antibacterial activity, color, and flavor of garlic and recommended through-flow dehydration for reasons of economics and quality.

For further drying of sun-dried black pepper, some of the progressive traders have installed cascade-type dryers using kerosene oil or gas as fuel. The moisture in the partially sun-dried black pepper is brought down from 25% to within 10% in two stages in a countercurrent hot-air flow system. Pepper is introduced at the top, while hot air is introduced from the bottom. The reason for drying in two phases is that in the first drying the surface of the pepper dries quite fast, but the moisture from within the core requires some time to diffuse out to the surface. Thus, after one pass the pepper is stored for 24 to 28 hours, after which it is dried again as above down to 10% moisture.

The author has also seen the working of a mechanical dryer imported from Holland, in which 30 to 40 tons of pepper can be dried per 8-hour shift. It has proved to be more economical than sun drying, with the added advantage of better retention of quality. It also has a winnowing facility, but it has one social repercussion because the of drastic cut it would bring in human labor normally engaged for sun-drying operations.

c. Other Factors. Several other factors, such as the method of preparation, pretreatments like sulfuring, sulfitation, or treatment with antioxidants (in chilies) or other chemicals, and density of loading affect the quality of the finished product, as discussed by Pruthi *et al.* (1959c,d,e) and in standard textbooks on the dehydration of foods.

2. Dehydration of Cardamom

For drying cardamom, a natural convection drying system is used in two ways. In large plantations, drying is done in an enclosed room in which the capsules are heated by a system of pipes running along the length and height of the room, through which hot air or flues are passed from an external furnace. The cardamom placed on the racks inside the heated room gets dried out slowly. In small holdings, cardamoms are dried on an open platform heated from below and sheltered by a roof open on all sides. Sometimes, drying is also done on beaten ground or on a mat in the sun, but color preservation is better with the above two methods. The Directorate of Marketing and Inspection (1965a) and Abraham (1965) have described in some detail the technique of drying cardamom capsules in a heated chamber, along with the precautions to be observed. Great care is to be exercised in controlling the temperature of drying as well as the drying rate. The dry-bulb temperature should not exceed 130° to 135°F in the stove. Drying should not be too rapid. At no time during the drying process should the cardamom be exposed to strong light, which bleaches out the green color (chlorophyll).

There is scope for improving the performance of the existing stoves by providing ventilators or exhaust fans at the top near the roof to facilitate the occasional removal of the air loaded with high moisture in the initial stages. Mechanical drying under controlled conditions is possible wherever electricity is available. Recently, a number of progressive planters in India have tried cross-flow dryers with success.

3. Dehydration of Garlic

Pruthi *et al.* (1959c,d,e) conducted a systematic study of several factors including method of preparation, pretreatments such as sulfuring and sulfitation, density of loading, and spraying of garlic with antioxidants, on the quality of the final product. The optimum tray load was found to be 1.14 lb/ft^2, or 4 lb per tray of 16 × 32 inches. Both sulfuring and sulfitation of garlic prior to through-flow dehydration improved the color of the product, but they significantly reduced its flavor and completely destroyed antibacterial activity. Hence sulfuring and sulfitation of garlic are unnecessary. Application of *l*-ascorbic acid at 200 mg per 100 g also had no beneficial effect on the retention of flavor and antibacterial activity. In fact, it appeared to enhance browning during dehydration.

4. Dehydration of Ginger

In Austrialia, the rotary dryer appears to be preferred for ginger dehydration because of lower labor costs (Sharpnell, 1967). However, unless the ginger is sliced fairly thin, its dehydration is essentially a slow process. It is controlled for the most part by moisture diffusion, which is not hastened by agitation, with the result that the extra power consumption involved in working the rotary dryer for long periods is not justified (Natarajan *et al.*, 1970; Kuppuswamy, 1974). The rotary dryer is not suitable for sliced ginger, since the wet material packed like sawdust forms an impermeable mass through which the air cannot penetrate (Sharpnell, 1967). Even with the development of newer drying techniques, most vegetables (including spices) in a wide range of shapes and sizes are still dried on belts or trays—the simplest and the most economical method of dehydration. Raina *et al.* (1978), using a cross-flow dryer, concluded that it took only 5–6 hours to dry sliced, scraped ginger, as compared with 16 to 18 hours for dehydration of scraped, whole ginger hands.

5. Dehydration of Onions

Dehydrated onion is produced by removing about 95% of the original moisture—that is, it is dried down to about 5% moisture—from peeled, derooted, and destemmed sliced onions. Pruthi (1960–1962, unpublished) and Pruthi and Lal (1960) studied the batch dehydration of onion and packaging and storage of onion powder. Havinghorst (1955) has described a novel continuous process for

the dehydration of onion and garlic. Details of this process are covered later in Chapter 6, Section II.

6. Dehydration of Pimento Berries

Breag *et al.* (1972) conducted systematic studies to determine the precise conditions for mechanical drying of pimento berries. On the basis of evaluations of drying rates conducted both in London and in Jamaica, analysis, GC pattern, and trade assessment, they concluded that green pimento berries could be artificially dried at an air-flow rate between 25 and 30 cm per second and a drying temperature of 70° to 75°C more economically than by sun drying, with an acceptable brown color and appearance and without significant loss of oil. It was also found that periods of up to 2 days of fermentation facilitate the drying process. An indirectly oil-fired tray dryer for the average-sized pimento estate has been recommended.

Theodose (1972) also has demonstrated the economic feasibility of his improved process of mechanical (tunnel) drying of cut pieces of vanilla beans, which reduces labor costs by 80% and is more efficient and convenient than sun drying.

III. PRECLEANING AND GRADING

Before packing, the dried seed spices are cleaned to get rid of extraneous matter such as dirt, grit, stones, stalks, and leaves, and also graded. Pneumatic separators equipped with magnetic separators are used to remove debris; their operation is based on the relative specific gravity of the particles. Magnetic separators are used to remove metallic contamination such as iron filings and stray nails. A combination of air classification and vibratory conveying, using inclined docks, is very efficient for destoning of spices. These units are fast, thorough in cleaning, compact, easy to install, and require a minimum of maintenance (Ramanathan and Srinivasa Rao, 1974).

On a small scale, the dried spices are generally cleaned manually by winnowing or by use of blowers, etc., so that the heavier and bolder berries of pepper separate out from the dust, stalk, and pinheads, which, being lighter, are blown away.

In well-known spice processing houses, pepper is cleaned and graded with the help of a multiple-sieve-cum-air-classifier type of machine whereby the following fractions are conveniently and quickly obtained: dust, stalks, pinheads, hollows, immature pepper, ripe pepper, and extra-bold pepper. The separated pepper is then washed and dried in order to make it free of any adhering fungus and other extraneous matter. Alternatively, mechanical brushing between two rotating brushes can be used both to clean the pepper and to impart a nice shine to it. To sum up, the above techniques of processing of pepper, technically, consist

first in four steps—(1) drying, (2) separation of various fractions, (3) size segregation or grading, and (4) physical cleaning—and finally packaging. It should be possible to cover the first four steps in one operation by using a pneumatic conveyer-cum-dryer with an arrangement for trapping the different cuts or segregations at different points or stages (Marivala, 1974).

Such mechanical cleaners-cum-graders should be installed near the producing areas or in the assembling or regulated markets so that only properly cleaned and graded spices are offered for auction. Likewise, destoners, air classifiers, gravity separators, color graders, and sieve graders of proper capacity should be installed for the benefit of the growers, sellers, and buyers. A single mechanical machine composed of clippers, air classifier, polisher, specific gravity or size grader, and inspection table, with a capacity of 3 to 5 tons of minor seed spices per shift of 8 hours, is available and is being used in some exporting and importing countries. Mechanical size graders of different capacities for onions and garlic are already in use, and their greater use should be encouraged in spice-producing countries.

Well-defined trade grades and quality characteristics of major spices such as pepper, cardamom, ginger, chilies, and turmeric, ground spices, curry powder, and eighteen other minor spices have been established by the Department of Marketing and Inspection, Government of India, according to the preshipment inspection scheme under the Compulsory Quality Control Act. These standards are being implemented rigorously under the Agmark Grading Scheme, particularly for exports from India (Directorate of Marketing and Inspection, 1957, 1965a,b, 1966a,b, 1968a,b, 1970a,b, 1971).

Dried cardamoms are cleaned by rubbing them gently over a coarse surface of wire mesh or bamboo trays in order to remove the stalks and all dried remains of floral parts. This is best done when the material is still hot (Abraham, 1965). The cleaned or sorted cardamoms are then graded into longs, mediums, and shorts.

Cassia and cinnamon barks are also sorted out and graded into quills, quillings, featherings, and chips, based on their length, unit weight, color, taste, texture, thickness etc.

Dried, unbleached ginger rhizomes are gently rubbed by hand in order to remove the last bits of the skin or any adhering extraneous matter. They are then sorted into suitable grades prescribed under the various national standards such as those of the U. S. Department of Agriculture, the Food and Drug Administration, National Standards Institutions, etc., which vary somewhat from country to country.

In the United States, most onions are graded in central packing sheds and warehouses, which are usually located along railroad sidings, where the bagged onions can be loaded directly into refrigerator cars or trucks. Onions are run over power-operated mechanical graders that blow and screen out any dust and loose scales, and grade the onions into different sizes. As the bulbs move along the grading table, undesirable ones are removed. Usually the several size grades are packed separately to make a more uniform and attractive product. Portable onion

graders, which allow cleaning, grading, and sacking in the field, are also used in some areas.

The adhering scales and root bases or hairy roots are removed from cured and dried turmeric rhizomes (bulbs and fingers) by rubbing with the help of a coarse cloth or burlap, followed by winnowing. The cleaned product is then graded into fingers, bulbs, brokens, etc.

Vanilla beans, after curing, are graded according to length and appearance. However, the system of grading of vanilla beans does not appear to be consistent. According to one system, there are four grades:

1. *Top class beans* are up to 20 cm in length, very dark, oily in appearance, show no defects, and have a good, powerful odor.
2. *Second Grade*. The lower grades are short, thinner, and lighter in color, and the odor is not so pronounced.
3. *Splits*. When the beans open at the end during the curing process, they are graded as "splits." Very thin pods are often combined with these.
4. *Cuts*. If the beans have developed mod, then the mouldy portion is cut away and the rest is sold as cuts.

IV. PACKAGING, PRESTORAGE PROCESSING, AND STORAGE

With the exception of delicate spices such as saffron and vanilla, which are packed in airtight tins lined with waxed paper or butter paper, most of the other spices are packed in airtight or double burlap bags with or without internal polyethylene linings. Vanilla beans are also packed in airtight wooden boxes lined with waxed paper. Another photosensitive and costly spice—cardamom—is sometimes packed in wooden or plywood chests lined with kraft paper or a suitable polyethylene lining and wrapped with burlap sometimes coated with coal tar as an insect repellent.

According to Mahadeviah *et al.* (1976), under tropical conditions, 200 gauge low- and high-density polythene films are suitable for retail packaging of whole chilies in units of 250 g each. Moisture content higher than 15% is critical with respect to mold growth. The discoloration of the red pigment of chili during storage is greatly influenced by moisture, temperature, and light. Detailed aspects of packaging and storage have been reviewed (Pruthi, 1970d) and are covered in Chapter 8.

6

Spice Products Technology

World demand has been increasing for processed spices and spice products such as sterilized ground spices, spice blends or seasonings, spice oils, oleoresins, essences, emulsions, decoctions, dispersed spices, encapsulated spices, and soluble or instant spices. No regular world statistics are available for many of them. However, the available technical information for most of these subjects is reviewed and briefly discussed below.

I. MANUFACTURE OF WHITE PEPPER

Black pepper *(Piper nigrum)* is by far the most important spice of the world, accounting for about 35% of the total world trade in spices. India alone contributes about 25 to 40% of the total world supply and thus occupies a unique position in the international marketing of pepper. Pepper's supremacy in the world of spices goes unchallenged (American Spice Trade Association, 1964).

Black and white pepper are prepared from the berries of the same plant or species, *P. nigrum*. If the end product is to be black pepper, the mature green or greenish yellow berries are separated from the spikes and dried in the sun, whereupon they turn black and become shriveled. They are cleaned, graded, packed, and sold as black pepper,

White pepper is prepared by removing the pericarp (rind) of the pepper berries, either before or after drying. White pepper is used primarily in food products in which dark particles would be undesirable, such as light-colored sauces and salad dressings, mayonnaise, and cream soups. In most of the European countries,

white pepper is used traditionally and preferred over black pepper (American Spice Trade Association, 1964). Considerable quantities of white pepper are also consumed in the United States, Canada, Australia, and New Zealand in the belief that it is milder in pungency than black pepper.

It is estimated that, of the total annual world production of 70,000 to 75,000 tons of pepper, about 25% is in the form of white pepper (Lewis *et al.*, 1969a); white pepper is thus of considerable economic as well as international importance.

A. Techniques of Manufacture of White Pepper

White pepper can be manufactured by any one of the following techniques:

1. Water steeping and rotting technique
   (a) From ripening fresh berries
   (b) From dried berries
2. Steaming or boiling technique (Lewis *et al.*, 1968, 1969a)
3. Chemical technique (Joshi, 1962)
4. Decortication technique

1. Water Steeping Technique

a. From Ripening Fresh Berries. According to the old, indigenous method, pepper spikes are harvested in stages when one or two berries in each spike start turning yellow or red. The berries are separated from the rachis, packed in burlap bags, or as such steeped in tanks or under running water for 7 to 10 days. The pinheads and light berries that come to surface are separated, dried, and sold separately. The remaining mass is stirred two or three times daily. On the eleventh day they are heaped on a tarpaulin and rubbed by hand or trampled to remove the softened skin. The deskinned berries are then washed and put in a galvanized iron vessel containing a solution of bleaching powder. Berries are kept immersed there for 2 days. They are then removed, drained, and dried.

b. From Dried Berries. Pepper berries dried for 7 to 10 days after harvesting are also packed in burlap bags and steeped in water for 1 to 2 weeks, after which the berries are removed, rubbed, washed, and dried in the sun for 3 to 4 days.

2. Steaming or Boiling Technique

This improved technique was developed at the Central Food Technological Research Institute, Mysore, India, by Lewis *et al.* (1969a), who have also discussed other processes and their merits and demerits. This process, in brief, consists in steaming or boiling of mature, ripening green berries for 10 to 15 minutes. The boiled berries are then passed through a motorized fruit-pulping machine to remove the skin. The deskinned berries are then washed and treated

with sulfur dioxide or bleaching powder solution, after which they are drained and dried in the sun or mechanically (Lewis *et al.*, 1968).

The skin of the boiled berries is collected separately from the machine and immediately utilized for steam distillation for the recovery of pepper oil, which is quite costly and constitutes an important by-product, thus giving this improved process an edge over other indigenous techniques (Lewis *et al.*, 1968, 1969a).

3. Chemical Process

Joshi (1962) patented a chemical process for white pepper, which consists of (1) steeping whole, dried black pepper with about five times its weight of water for 4 days; (2) treating it with 4% NaOH solution and boiling the mixture; (3) removing the skins by agitation with a stirrer at 1600 rpm; (4) washing the product with water; (5) bleaching with 2.5% v/v H_2O_2 solution; and (6) drying the white berries at 125°F. Use of other chemicals and bleaching agents gave similar results. This process has not been tried commercially, however.

4. Decortication Process

White pepper is also obtained by decorticating the whole, dried black pepper berries in special decorticating machines, details of which are not available. The major problem in decortication is too much breakage of berries, and lack of uniformity in shape, surface, etc.

B. Comparative Techniques in Southeast Asian Pepper-Producing Countries—Their Advantages and Disadvantages

On the mainland of South Sumatra (Lampong), only black pepper is produced, whereas in Bangka, an island of South Sumatra, the main production is white pepper. In Sarawak, both black and white pepper are produced, but the quantity of the two kinds produced in a year depends on the price that each kind brings. In Thailand, Cambodia, Malaya, and Ceylon, both white and black pepper are produced (Kurup, 1959).

The percentage of white pepper in relation to the ripe, fresh berries is about 25, while the percentage of black pepper from fresh pepper is about 33. Generally, the price of white pepper is 65 to 75% more than that of black pepper in Southeast Asian countries, but in certain years, owing to an increased demand for white pepper, the price of white pepper may be far higher. On other occasions, the difference may not be much. Actually, the production of white and black pepper fluctuates according to the price each kind fetches.

For producing white pepper, in Bangka, Sarawak, Thailand, and Malaya almost all the berries on the spikes are allowed to become ripe. Thus, harvesting is done in different stages. In Indonesia, Sarawak, Thailand, and Malaya, even for making black pepper, harvesting is done when almost all the berries on the

spikes are ripe. In Cambodia, the harvesting is done when a few berries on the spikes become ripe. Thus, harvesting has to be spread over a period of about three months. In Ceylon, the spikes in the lower half of the vines are harvested first and used for making black pepper. For making white pepper, the spikes from the upper portion of the vines are used, and these are harvested in one stage. The spikes from the upper half of the vines are harvested about a week or two after the spikes from the lower half are harvested. Generally, the spikes in the upper portion of the vines mature earlier than those in the lower portion, and therefore a large percentage of the berries in the upper portion of vines will have become ripe. In Ceylon, white pepper is produced only to a very limited extent in a few European-owned cocoa plantations, where pepper is grown as an intercrop trained to the shade trees.

In Cambodia, the entire spikes are dried for about 50 days and the berries are separated by trampling with feet. The dried berries are winnowed, and the heavier fraction is separated. This heavier fraction is put in burlap bags and steeped in water for about 10 days to produce white pepper; the lighter fraction is disposed of as black pepper (Kurup, 1959).

In Ceylon, the berries from the upper portion of the vine that are almost ripe are heaped up and covered for 3 days for withering. They are then steeped in a cement tank filled with running water from a pipe for about two weeks. When the rind has become soft and black, it is removed by trampling with feet in small tubs or buckets. The separated rind is removed by further washing with water, the water is drained, and the white pepper is dried on a cement floor or on mats (Kurup, 1959).

In India, at present, there is little production of white pepper, except in very small quantities in some households in Kerala for medicinal and domestic purposes (Directorate of Marketing and Inspection, 1971).

Allowing the spikes to become fully ripe on the vine is not considered advisable by the growers in India, since such practice reduces the yields and is believed to weaken the vines and cause their premature death. Allowing almost all the berries on the spikes to become fully ripe on the vine itself certainly will reduce the yield, owing to the removal of the ripe berries by birds and dropping off of the ripe berries naturally and in the process of harvesting. The belief that allowing the spikes to become fully ripe will reduce the life of the vines is supported by the fact that the life of the vines in Indonesia, Sarawak, Thailand, and Malaya, where this practice is prevalent, is much shorter than that in Cambodia, Ceylon, and India, where this practice is not followed. If the spikes are to be harvested only when almost all the berries have become fully ripe, then harvesting cannot be done in one stage, but will necessarily have to be done in stages and will therefore spread over a few weeks at least. In Indonesia, Sarawak, etc., the harvesting is done by the grower himself at weekly intervals, whereas in

India, and also in Ceylon, the harvesting is done by hired laborers mostly. Therefore, if the harvesting is done in stages, the harvesting cost will be very high.

C. The Comparative Composition of White and Black Pepper

The comparative composition of black and white pepper with regard to important constituents is summed up in Table XLI. The United States Standards for black and white pepper are given in Table XLII. The significant difference between black and white is in starch and fiber content. Some differences in composition due to geographiclal origin also are indicated. Brazilian white pepper is less pungent than that from Muntok. The belief that whie pepper is milder in flavor than black pepper does not seem to be confirmed by the scientific data (Table XLI).

TABLE XLI ANALYSIS OF WHITE PEPPER VARIETIES FROM DIFFERENT COUNTRIES[a]

Country of origin	Volatile oil (%)	Nonvolatile ether extract (%)	Piperine in nonvolatile ether extract (%)	Piperine in pepper (%)
India	2.51	7.60	60.7	4.61
Sarawak	1.97	8.72	54.0	4.71
Brazil	1.96	8.68	50.0	4.34
Muntok	2.03	8.47	57.0	4.83
India (black pepper, garbled)	2.32	8.53	50.0	4.20
Malaya	2.22	8.09	—	—

[a] From Lewis *et al.* (1969a).

TABLE XLII UNITED STATES STANDARDS FOR BLACK AND WHITE PEPPER

Constituents	Black pepper (%)	White pepper (%)
Ash (max)	7.0	3.5
Acid-insoluble ash (max)	1.5	0.3
Crude fiber (max)	15	5
Nonvolatile ether extract (min)	6.75	7
Starch (min)	30	52
Foreign organic matter (max)	2	2

II. DEHYDRATED PRODUCTS

Of late, there has been a growing demand for dehydrated spices such as onions, garlic, paprika, and bell peppers. The greatest demand is for dehydrated onions, which are used mostly for seasoning of a number of food products such as soups, sausages, and dry soup mixes. Dehydrated soup is one of the most popular products. There are a number of reports on dehydrated garlic, paprika, and bell pepper, which are briefly reviewed below.

A. Dehydrated Onions

Of all the spice bulb crops, onion *(Allium cepa)* is commercially by far the most important. It is both a vegetable and a spice or condiment, when dehydrated. The annual average world production of onions is estimated at around 7 million tons. The United States is the leading producer. The United Arab Republic, Japan, Brazil, Bulgaria, Spain, India, and Italy are other major onion-producing countries. In international trade, the United Arab Republic, The Netherlands, Spain, India, the United States, and Italy play an important part in exporting onions, contributing, respectively, 20.9%, 19.8%, 10.2%, 9.6%, 7.4%, and 6.7% (together accounting for 74.6%) of the total exports (Directorate of Marketing and Inspection, 1965b). Dehydrated onion is a highly competitive product. Dehydrated onions are available in different shapes or forms—sliced, chopped, minced, flaked, kibbled, granulated, and powdered. Dehydrated onion is produced by removing about 95% of the original moisture—that is, it is dried down to 4 to 5% moisture—from peeled, derooted, and destemmed sliced onions. Generally, 10 pounds of onions yield 1 pound of dehydrated onion. This dehydration ratio (10:1) varies with the variety, quality, size of onion, processing conditions, etc. The rehydration ratio also varies but is generally of the order of 1:8.

The important prerequisites of onions suitable for dehydration are (1) white flesh, (2) full globe to tall globe shape, (3) medium size (5 to 6 cm in diameter), (4) high solid content, about 15 to 20%, (5) high degree of pungency, (6) good yield, (7) good storage life of two to three months, (8) freedom from joints, and (9) freedom from fungal and bacterial contamination (Jones and Mann, 1963). Bhatti and Asghar (1965) evaluated six Pakistan varieties of onions for dehydration purposes. Sethi *et al.* (1973) screened as many as sixteen Indian varieties of onions and recommended cultivar No. 36-1-3-4 grown at IARI, New Delhi, for dehydration purposes.

A novel, continuous process for the dehydration of onion and garlic has been reported by Havinghorst (1955), in which a photocell-fed flame peeler, automatically controlled flow, and continuous belt-type dehydrator replace the conventional batch-type, tray-tunnel method in the production of dried onions and

garlic. This technique increases output to 75,000 pounds of onions of 86% moisture per 24 hours, cuts down labor, and upgrades the quality of slices through highly effective quality control on the production line.

The various steps involved in processing are flame peeling, ashing, trimming, second washing, final trimming, surge hopper, elevator, belt scale recorder delivering uniform recorded flow, slicer, and dehydrator. A photoelectric cell regulates the feed from the hopper to the elevator leading to a flame peeler, which is actually a rectangular-shaped, refractory shell furnace. It is fired by forty-nine high-velocity natural gas burner nozzles. Under high pressure, these develop a temperature in excess of 2000°F. It takes only 10 to 12 seconds to flame peel each batch of 3000 pounds. The process is very efficient.

The outstanding features of this unit are: (1) continuous instead of batch-type flow; (2) elimination of foreign matter; (3) increased capacity of 75,000 pounds per 24 hours for onions containing 96% moisture; (4) a more uniform, higher quality product; (5) highly effective quality control; and (6) saving in labor (much less needed than in tunnel operation).

B. Dehydrated Garlic

Pruthi *et al.* (1958) patented a novel, simple, and comparatively inexpensive yet efficient batch process for the manufacture of dehydrated garlic and powder, which cuts down the labor cost, reduces the time of dehydration, and results in significantly improved garlic powder of better color, flavor, and antibacterial activity. This process is being used by the industry in India. Pruthi and Subrahmanyan (1962) have described this process along with its advantages and economic feasibility.

Casetti (1962) also patented a process in France for the manufacture of garlic powder. Preservation of color and freshness of garlic powder is obtained by vaporization of 50 mg of SO_2 for every 100 g of garlic powder. The amount of NaCl used can be increased to 40% of the weight of the powder. However, the addition of SO_2 in garlic powder is not permitted in some countries. Furthermore, sulfitation reduces the flavor and destroys the antibacterial activity of garlic, as reported by Pruthi *et al.* (1959c).

Singh *et al.* (1959a) presented results of a systematic study on the extent of variation in moisture, allyl sulfide, total sulfur, antibacterial activity, color, and flavor of garlic grown in three major garlic-producing regions of India—Jamnagar, Arsikere, and Mysore. Although there was no material difference with respect to physical characteristics such as percentage of recovery of cloves from garlic grown in the three regions, garlic from Jamnagar and Arsikere produced a better quality product than that from Mysore. With regard to the component parts of the garlic bulb, the outer, papery skin or husk and the tough skins of the cloves

had no antibacterial activity and negligible amounts of allyl sulfide and total sulfur. The pungency and antibacterial activity were mostly present in the peeled cloves. Considerable batch-to-batch variation was observed with respect to the effect of bulb size on pungency and antibacterial activity of garlic powder.

On the basis of data on the retention of antibacterial activity, allicin, allyl sulfide, color, and flavor in fresh and dried garlic flakes during dehydration at different temperatures ranging from 30° to 70°C, the critical temperature for dehydration of garlic was reported to be 60°C by Pruthi *et al.* (1959b), who suggested that the product temperature during through-flow dehydration of garlic should preferably be lower than 60°C.

Pruthi *et al.* (1959c), in another paper, studied the effect of three factors relating to dehydration on allyl sulfide, antibacterial activity, flavor, color, and general quality of garlic powder. They suggested optimum tray loads of 1.14 pounds of freshly prepared garlic per square foot of tray surface, or 4 pounds per solid tray (16 × 32 inches). Both sulfuring and sulfitation of garlic prior to dehydration improved the color of the garlic powder slightly, but they significantly reduced its flavor and completely destroyed its antibacterial activity. The application of *l*-ascorbic acid at the rate of 200 mg per 100 g had no beneficial effect on the retention of antibacterial activity and flavor of garlic powder. In fact, it appeared to enhance browning during the dehydration of the product.

Pruthi *et al.* (1959d) also made a comparative critical study of the effects of different methods of drying or dehydration on the quality of garlic powder. Freeze drying and vacuum-shelf drying produced a product with slightly better color than was found in products obtained by hot-air drying (through flow and cross flow) and sun drying. There was, however, no significant difference in the retention of flavor and antibacterial activity. Since freeze drying and vacuum-shelf drying are not economical, hot-air drying is preferred. Of the two hot-air drying methods, through-flow drying is recommended because of the 50% reduction in drying time.

Pruthi *et al.* (1959e) presented data on the pilot-plant scale production of garlic powder by the new and the old techniques. The yield of garlic powder (6% moisture) was about 26%. The losses in allyl sulfide and antibacterial activity were about 50%. The cost of production by the new technique was about 30% lower than that by the old technique, with the added advantage of a better-quality powder.

C. Odorless Garlic Powder

Pruthi *et al.* (1959i) were successful in producing an odorless garlic powder by thermal inactivation of allinase naturally present in garlic prior to its dehydration. Further studies revealed that flavor regeneration in odorless garlic powder could

be achieved by addition of the enzyme allinase, which, on reaction with alliin, would produce allicin, the main flavoring principle, which is also antibiotic. Two Japanese patents (Kojiya Mizaemon Co., 1966; Yamada Seijaku, 1973) on odorless garlic powder have also been reported.

Pruthi *et al.* (1959a) have reviewed the available literature on the chemistry and technology of garlic powder.

D. Freeze Drying of Onion

Hamed and Foda (1966) studied the freeze drying of onions. Slices of onion (4 to 6 mm thick) were rapidly frozen to −30°C. Drying was for 7 to 8 hours at 6.1 mm Hg pressure, while on a plate warmed to 80°C. A final moisture content of 2.00 to 2.14% was attained. The drying had no great effect on the ascorbic acid, volatile sulfur compounds, or sugar content. The process did not cause discoloration. The ability to take up water was much better in the freeze dried than in the air-dried products. Pruthi *et al.* (1959i) studied freeze drying of garlic for the preparation of odorless garlic powder.

Freeman and Whenham (1974a) studied the changes in onion flavor component resulting from some postharvest processing, including freeze drying and dehydration in hot air.

E. Pink Discoloration in Dehydrated Onion

Li *et al.* (1967) prevented the discoloration (pink color formation) of comminuted white onions by the addition of 0.05 to 0.3% by weight of cysteine and heating to 160° to 190°F to destroy pathogenic microorganisms. The cysteine may be as the free base or as the hydrochloride or sulfate. An antioxidant (citric or ascorbic acid), up to 0.3% by weight, may be added. Thus, a batch of White Globe onions was washed and comminuted, and to the mass was added 0.05 wt% L-cysteine. The mass was pasteurized at 161°F for 15 seconds, cooled to room temperature, and reduced to the desired viscosity. The onions were white even after 154 hours.

Lukes (1959) conducted an investigation into the causes for the development of a pink pigment in onions while being dehydrated, which demonstrated that the discoloration is not due to bacterial action. Actually, when the onion cells are ruptured, the native enzymes react with a substance in the juice to form a color developer. This then reacts nonenzymatically with the amino acids in the onion to form a pink pigment.

A patented process (Li *et al.*, 1967) for the prevention of pink discoloration in comminuted white onions consists in the addition of 0.05 to 0.3% by weight of cysteine and heating to 160° to 190°F.

F. Types of Dehydrated Onion Products

The American Dehydrated Onion and Garlic Association has set up standard nomenclature for the various onion particle sizes and has also established normal bulk index averages, as well as screening and other specifications. The bulk index, the volume in milliliters of 100 g of the product after tapping to constant volume, is important in figuring the net weight that may reasonably be packed within any given size of container. The following tabulation gives the American industry identification of the various sizes, approximate screening, and bulk index averages for each dehydrated onion product:

Nomenclature	Screening (Tyler)	Bulk index (ml/100 g)
Powdered onion	Through 48 on 200	Not determined
Granulated onion	Through 32 on 100	160
Ground onion	Through 14 on 48	165
Minced onion	Through 6 on 20	205
Chopped onion	Through 4 on 16	245
Large chopped onion	Through 3 on 12	390
Sliced onion	Through ⅜′ on 10	450
Large sliced onion	Through ⅝′ on 8	600

All the various sizes of dehydrated onion are available in toasted form. Toasting gives the product a sautéed-like onion flavor. Also, ground, granulated, and powdered onions are available with a small amount of calcium stearate added as an anticaking agent, desirable when packed for food service or consumer use (American Spice Trade Association, 1967).

Similarly, dehydrated garlic is prepared in the form of flaked, granulated, or garlic powder with or without anticaking agent.

G. Microbiology of Dehydrated Onion

Vaughn (1951) conducted systematic studies on the microbiology of dehydrated vegetables, including onions, in relation to the factors that influence the microbial populations of the finished products. The souring of onions during dehydration has been described in detail to show the necessity of a minimum time lag between preparation and start of the dehydration process (Table XLIII), and optimum tray loading and its relation to the finishing temperature of the dehydrated product (Table XLIV). The possible use of microorganisms as indices for quality of dehydrated vegetables (including onions) has been discussed in relation to the need for the attainable tolerances. The earlier work has also been critically reviewed, and the need for obtaining additional information has been stressed (Vaughn, 1951, 1970).

The following factors in production have been reported to influence markedly the microbial population of dehydrated vegetables:

1. The microbial quality of the fresh produce.
2. The method of pretreatment (peeling, blanching, etc.).
3. The time lag between preparation and start of the dehydration process.
4. The time of dehydration.
5. The temperature of dehydration.
6. The moisture content of the finished product.
7. The general level of sanitation in the dehydration plan.
8. Other factors, such as chemical characteristics of the raw material; the presence or absence of antibiotic compounds in the raw material; correct loading of the dryers; and the average finishing temperature of the product at the end of the dehydration process.

Although the effects of most of the above factors have been recognized in a general way, many have received little or very little consideration with respect to their effect on bacteria that may survive on dehydrated vegetables.

Johnson and Vaughn (1969) studied the kinetics of the decline of population of *Salmonella typhimurium* inoculated into freshly reconstituted dehydrated onion and garlic powders. At comparable concentrations, growing cultures of *Escherichia coli* were as susceptible to garlic but apparently more resistant to onion than were those of *S. typhimurium*.

Recently, the commercial users of dehydrated onion and garlic products have been prone to demand unattainable microbiological tolerances in their purchase specifications. As a result, considerable confusion has developed with regard to the significance and practical importance of the microbiology of these products. In an effort to respond to this problem, Vaughn (1970), Sheneman (1973), and Firstenberg *et al.* (1974) have studied the incidence of various types of microorganisms present in dehydrated onion and garlic and the possible significance of

TABLE XLIII BACTERIAL COUNT OF ONIONS SLICED AND LEFT STANDING BEFORE DEHYDRATION[a,b]

Standing time (hours)	Bacteria per gram on fresh-weight basis	
	E.M.B. agar	Yeast glucose agar
0	5,925	28,500
2	22,000	35,250
4	39,000	106,000
8.5	310,000	675,000

[a] From Vaughn (1951).

[b] Net weight of onions on car, 580 lb, equal to tray load of 1.24 lb/ft^2. Air temperature at start of experiment 40°C (104°F).

TABLE XLIV EFFECT OF CAR LOADING ON SURVIVAL OF MICROORGANISMS DURING DEHYDRATION OF ONION SLICES[a]

Car number	Car load (lb)	Tray load (lb/ft²)	Bacterial counts (dry-weight basis)[b] Before drying	After primary tunnel	After secondary tunnel[c]	After bin[c]
1	435	0.93	365,000	312,000	70,200	108,000[d]
2	630	1.35	180,000	248,000	59,700,000	13,020,000 (composite)[e]
					62,000,000	24,700,000 (wet)
3	650	1.39	299,000	245,000	3,980,000	10,000,000 (composite)[e]
					14,800,000	12,000,000 (wet)[f]

[a] From Vaughn (1951).

[b] Counts made on yeast infusion, glucose agar.

[c] The apparent increase in numbers is partly the result of sampling error, which, for obvious reasons, is quite high with onion slices, and partly the result of multiplication of the bacteria.

[d] This is a composite sample of a uniformly dry car.

[e] The composite samples include dry and wet slices that had soured.

[f] Total drying time of 24 hours. Finishing air temperature of primary parallel airflow tunnel 130°F (54.4°C); finishing air temperature of secondary countercurrent airflow tunnel 143°F (61.7°C); finishing air temperature of bin 115°F (46.1°C).

each group as an index of microbial quality of the products as related to their use in remanufacture.

Vaughn (1970) systematically investigated the incidence of different thermophilic spores known to be of importance; the coliform bacteria, the lactic acid bacteria, the pectinolytic and psychrophilic bacteria, and the standard plate count of dehydrated onions and garlic.

Sheneman (1973) conducted a survey of aerobic thermophilic bacteria and observed that the normal aerobic bacterial microflora of well-processed dehydrated onion products consisted almost entirely of spore-forming bacteria of the genus *Bacillus*. These have little significance as a health hazard but are important only in that they are responsible for the high total plate count in the finished product. No *Salmonella* or *E. coli* were found in dehydrated onion and garlic. Non-spore-forming bacteria had a low population.

Firstenberg *et al.* (1974) sought a reliable indicator for the microbial quality of dehydrated onions and also for the effect of brine treatment of onions on final microbial counts. According to these authors, enterococci were a better index than coliforms for microbial contamination in dehydrated onions. They multiplied at a slower rate on raw onions and were much more resistant to the process and in storage. In further dehydration experiments both in the laboratory and on the industrial line, it was found that dipping or fluming of onions prior to dehydration in 2 to 4% salt solutions improved their total count about 12-fold and

coliform counts about 32-fold. The fungi counts of dried onions could be reduced to treating whole onions with saturated steam prior to dehydration. Krishnaswamy *et al.* (1973, 1974a) also studied the standard plate count, types of coliform, aerobic spore-formers, yeasts, and molds in some of the spices including dehydrated onions. They stressed the need for the collection of more first-hand data for the formulation of microbiological standards in view of the growing quality consciousness among the importers of spices and specially of dehydrated onion and garlic.

H. Dehydrated Paprika, Capsicums, Bell Peppers, and Chilies

While studying the effect of canning and drying on carotene and ascorbic acid content of chili, Lantz (1946) found that slicing or slitting the pods of chili reduced drying time by half and that there was no loss of initial color on drying for 72 hours at 140° to 168°F. Lease and Lease (1956a) reported that the color retention in peppers is affected by the stage of ripeness at harvest. By the addition of the antioxidant BHA to the ground pepper, the retention of color is improved (Lease and Lease, 1956b). While studying the effect of drying conditions on initial color, color retention, and pungency of red peppers, Lease and Lease (1962) concluded the following: (1) Drying or curing of sliced pods of Carolina hot pepper at 150°F was optimum for quality. (2) BHA antioxidant, when added, markedly improved the color retention of cured pods and also of freshly harvested pods. (3) Drying at 70°F led to a lower initial color, color retention, and pungency. (4) No correlation was found between the moisture content and the above quality factors. (5) Aired peppers retained more color when stored whole than when ground following curing. However, the color of whole, cured pods was lost on grinding. (6) Another variety, Louisiana Sports, lost color, etc., faster than did the Carolina variety. (7) Autoclaving pepper before drying increased the rate of color loss and suggested that color breakdown could not be attributed directly to enzyme activities.

Daoud and Luh (1967) reported that the color of red bell peppers is preserved by freeze drying without blanching. Temperatures of less than 68°F are best for quality retention. The loss in aroma and flavor at 86°F is due to deteriorative chemical changes and enzymatic reactions—for example, the formation of brown water-soluble pigments, the formation of cysteic acid and taurine from cysteine, and the loss of amino acids and carotenoids.

Chen and Gujmanis (1968) showed that the deterioration of extractable color pigments of dehydrated ground chili peppers during storage was due to an autoxidative process having the kinetics of a second-order reaction. Consequently, the reaction rate constant, K_2, was used to evaluate the effect of a number of variables, such as moisture content, storage atmostphere, and ethoxyquin treatment. It also provided a means for comparing the relative color stability of different pepper varieties.

De La Mar and Francis (1969) studied carotenoid degradation in bleached paprika. Nearly 96% of the total extractable pigment expressed as β-carotene is lost during sunlight bleaching of paprika. There were fifty-four and thirty-seven pigments isolated from bleached and unbleached paprika, respectively, of which thirty-three and twenty-one, respectively, were definitely or tentatively identified. Sixteen known pigments were found in both samples. Pruthi (1969a) also reported the bleaching or degradation of color (capsanthin and capsorubin) in Hungarian paprika powder during storage.

Phillip and Francis (1971a,b) reported the isolation and chemical properties of capsanthin and derivatives as well as the nature of fatty acids and capsanthin esters in paprika. Rosebrook (1971) reported collaborative studies in a method for determining the extractable color in paprika and paprika oleoresin.

Prakash and Natarajan (1974) reported the composition and dehydration of curry leaves.

III. GROUND SPICES

Spice milling is an ancient industry, closely akin to the cereal milling industry, with the difference that in spice grinding there are the additional problems of the volatility of essential oils, etc.

There is a considerable volume of international trade in ground spices, among which black and white pepper powder is by far the most important. It is available in a variety of different grinds. The most common are cracked, coarse ground, table grind, and fine grind or pulverized. Generally speaking, the finer the grind, the more immediately available is the flavor, but the shorter its shelf-life will be (American Spice Trade Association, 1964). Other popular ground spices include chili or capsicum, turmeric, onion, garlic, cinnamon, and coriander. Strauss (1967) investigated nutmeg powder.

The optimum size of the grind for each spice depends on its ultimate end use. Standard ground spices are usually ground to allow them to pass through U. S. standard sieves ranging from No. 20 to No. 60 mesh. Spices are also ground to microscopic fineness with a particle diameter of 50 microns, which is one-twentieth the size of 60-mesh particles.

The proponents of this fine grind argue that extreme fineness contributes to unlocking of the natural flavors, aids quick and thorough dispersion, and permits constant control of flavor intensity. When they are used in food products, uniform distribution of microground spices avoids hot spots. Further, because of the minute particle size, no dark specks can be noticed in the finished product (Neale and Klis, 1964). Also, the coloring components are extracted into the carrier medium or the food product within a short period of cooking. Grinding improves the esthetic appeal of the product and also enables the housewife or other commercial users to effect greater economy in use, time, and labor.

The other advantages of ground spices, according to Heath (1972), are as follows: (1) slow flavor release in high-temperature processing; (2) ease of handling and accurate weighing; (3) no problems of labeling declaration. However, ground spices are known to lose a measurable fraction of their volatile oil or flavoring components owing to the heat generated during grinding (Miller, 1951; Pruthi and Misra, 1963c). Pruthi and Misra (1963c) reported total losses of 0 to 1.15% of volatile oil and 0.5 to 3.0% of moisture from different spices during grinding. They also reported that the product temperature during milling ranged between 42° and 95°C in different spices, when ground individually. Water cooling of grinding machines (Parry, 1945), use of liquid nitrogen (Miller, 1951), and use of "amulin" during grinding (Grimme, 1953, 1954a,b) are some of the means practiced in some countries to check such losses of volatile oil. According to the Griffith Lab, Inc., conditioning or chilling of spices prior to their grinding curbs the loss of flavor volatiles during both milling and storage. In spice milling, controlled storage temperature and humidity is a must! This system saves labor, time, and space. Conditioning the spices for milling also increases throughput up to 20% and minimizes shrinkage during milling. They go through the mill easily, without smearing, blinding, or coating the mill. Refrigerated storage of spices reduces loss in weight. Thus, black pepper when stored in the cooler for three months lost only 0.3% weight, whereas its counterpart stored in the conventional warehouse lost 3.6% weight.

A. Milling or Grinding Equipment

In order to reduce flavor loss in ground spices due to excessive heat produced during grinding, cooling arrangements by means of air blown through the grinders or jacketed water-cooled units are necessary. Closed-circuit grinding with vibratory screens gives a variety of particle sizes.

For size reduction or fine grinding, air-swept beaters, double rollers, cages, and hammer mills are used. Hammer mills are not very satisfactory for fine grinding. Plate mills are used for small-scale and domestic purposes, and pin mills are employed for very fine grinding and higher capacity.

The fat content of spices is generally a problem in milling. Particle size, product yield, product uniformity, freedom from contamination, economy, and dust from the operation are other factors to be considered in selection and operation of the grinders (Ramanathan and Srinivasa Rao, 1974).

B. Cryomill Process of Freeze-Grinding of Spices

1. Principle and Description of the Process

Wistreich and Schafer (1962) have described a novel "Cryomill process" of freeze grinding of spices, which is accomplished by the controlled injection of

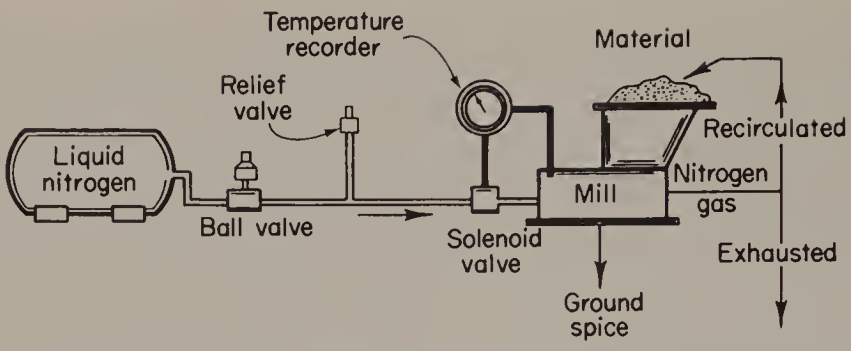


FIG. 11. Schematic drawing of a freeze-grinding (Cryomill process) installation. Liquid nitrogen is piped from large storage tank (left) to the mill's grinding zone. A temperature controller, which monitors the flow of liquid through a solenoid valve, maintains the desired product temperature. Part of the exhausted stream of cold nitrogen gas is recirculated to the spice hopper for precooking of spices. From Wistreich and Schafer (1962).

liquid nitrogen (which acts as a direct-contact refrigerant) directly into the mill's grinding zone (Fig. 11). The instantaneous evaporation of the liquid refrigerant quickly chills both the spice and the mill. It also absorbs the frictional heat of grinding. Thus, the temperatures in the grinding zone generally are well below −100°F. In contrast, conventional spice milling can raise spice temperatures to as high as 200°F (Wistreich and Schafer, 1962; Misra, 1962; Pruthi and Misra, 1963c).

2. *Advantages of the Cryomill Process*

This novel process has several advantages over conventional processes: (1) It cuts down the oxidation of spice oils because, as the liquid nitrogen evaporates in the grinding zone, it tends to expel any air in the mill. The flavor is thus much better retained. (2) It also permits extremely fine grinding because the spice oils solidify at low temperature, thereby making the spices very brittle. (3) Such finely ground spices disperse flavor uniformly throughout the final product. (4) They virtually eliminate specking problems, as in sausages. (5) In liquid preparations, the settling rate of freeze-ground spices is noticeably reduced. (6) It curbs the usual loss of spice aromatics and moisture. The ground products retain their original flavor strength and weight. On an average, 7 ounces of freeze-ground spices have the same flavoring power as 10 ounces of their conventionally milled counterparts. (7) Freeze-ground spices have proved to be considerably more stable than conventionally ground products. Possibly, the spices absorb or in some way retain some of the nitrogen. (8) Low-temperature treatment lowers the microbial load on the spices. (9) Grinding rates are increased because the mill's low-temperature operation minimizes the "gumming up" of grinding surfaces and screws. In a 25-horsepower mill refrigerated with liquid nitrogen, 1000 pounds of spices can be ground per hour. (10) The actual costs of Cryomill

spices are lower than those for conventional products when the increased flavor strength is taken into account. In addition, fine grind and greater stability are bonuses to food processors. (11) The process is applicable to a variety of other foods, such as cocoa, coffee, tea, coconut, vanilla, and dehydrated meats.

C. Disadvantages of Ground Spices

Compared with spice oleoresins, ground spices, according to Heath (1972), have the following disadvantages:

1. Variable flavor quality
2. Variable flavor strength
3. Microbial contamination
4. Possible contamination by filth
5. Easy adulteration with less valuable materials
6. Presence of lipase enzymes in some spices
7. Flavor loss and degradation on storage
8. Undesirable appearance characteristics in end products
9. Poor distribution of flavor, particularly in thin liquid products (sauces)
10. Discoloration due to tannins
11. Usually a hay-like aroma in herbs
12. Bulk handling dusty and unpleasant

Some of these objections may be overcome by the use of sterilized spices.

IV. STERILIZED SPICES

Some of the objections to grinding of spices may be overcome by the use of ground spices that have been exposed to mixtures of carbon dioxide and ethylene oxide, or some other bactericidal gas, in order to reduce the bacterial load (Heath, 1964; Gerhardt, 1969). Such spices are, however, graveyards of dead bacteria and may carry some load of bacterial toxins (Heath, 1972). The sterilizing technique involves very precise operating conditions, particularly with regard to temperature and humidity control, and the gases used seldom penetrate sufficiently to kill all the bacteria or bacterial spores carried by the original spice. Nor does the technique get rid of rat hairs, droppings, and insect parts, all of which are objectionable in food products. Most of these gases also act as solvents to a certain extent, and there is often a loss of essential oil strength after gassing—especially as the process involves vacuum treatment. It is well-recognized that one fumigant gas, ethylene oxide, in the presence of salt, forms traces of

TABLE XLV REPRESENTATIVE MICROORGANISM COUNTS ON TREATED (STERILE TREATMENT) AND UNTREATED SPICES[a]

Spice	Total bacterial count/g		Mold and yeast/g	
	Untreated	Treated	Untreated	Treated
Cinnamon	110,000	100	3800	0
Cumin	17,000	100	1000	0
Ginger	5,500,000	7100	1500	0
Marjoram	38,000	200	2600	0
Nutmeg	220,000	500	600	0
Oregano	13,000	100	1200	0
Black pepper	3,000,000	2600	2300	0

[a] Courtesy A. E. Staley Manufacturing Co., Detroit, Michigan.

chlorohydrins (Wesley *et al.*, 1965; Coretti and Inal, 1969). These are known to be toxic and, hence, undesirable—even in trace quantities—in any food product.

Using an FDA-accepted procedure that incorporates ethylene oxide as the effective sterilent, the gas sterilization unit typically reduces total plate counts to less than 10,000 per gram (Table XLV). This treatment is effective against molds, yeasts, and bacteria, including flat sours, thermophiles, mesophiles, and coliforms. Residual sterilent levels conform to maximum tolerance limits. Malter (1952) patented a process for decolorizing and sterilizing spices (clove, nutmeg, and pepper). Szabo (1968) described experiments for reducing microbial contamination of paprika.

Fortunately, spices are not favored by many insects that cause serious infestation in many other agricultural commodities. Perhaps the only insect that prefers spices is the drugstore beetle or the biscuit or bread beetle, *Stegobium paniceum* (L) (Coleoptera: Abobuidae). Muthu *et al.* (1962) and Muthu and Majumder (1974) have reviewed the subject of inspect control, which is discussed briefly in Chapter 11.

V. SPICE MIXTURES, BLENDS, OR SEASONINGS

Curry powders are mixtures or blends of numerous spices with varying formulas to suit the preparation of appetizing foods that are delicious to taste and exotic in flavor.

Curry powder is a judicious blend of different spices, farina, and salt. According to the International Organization for Standardization (ISO) specification (1971), "curry powder is the product obtained by grinding clean, dried and sound spices and condiments. The ingredients used in the preparation of curry powder should be listed on the container in descending order of proportion by

mass. The powder may contain edible starchy material, such as farinaceous matter or farina (nature to be declared). It may also contain not more than 5% (w/w) sodium chloride of food grade. The proportion of spices in the curry powder should not be less than 85%, meaning thereby that farina should not exceed 10% (w/w). Curry powder should be free from artificial coloring matter, and its flavor should be clean, fresh, and typical of the product. It should not have a rancid taste or musty smell. It should be free from coarse particles and should be of a fineness specified in the national standards. It should also be free from living insects or molds and should be practically free from dead insects, insect fragments, and rodent contamination, visible to the naked eye (corrected, if necessary, for abnormal vision with such magnification as may be necessary in any particular case). If the magnification exceeds 10×, this fact should be stated in the test report.''

''The chemical requirements of curry powder are that it shall not contain more than 10% moisture, 1% acid-insoluble ash, and 15% crude fiber. It shall contain at least 0.4% volatile oil per 100 g and 7.5% nonvolatile ether extract. Most of the national standards conform to the above ISO specification.''

The term ''curry powder'' should not lead one to think of it as something very pungent and therefore best neglected. Not all spices are hot, and therefore not all curry powders are too pungent or too hot. They should not be neglected, since they stimulate secretion of digestive fluids in the stomach. They are carminative, digestive, and antiflatulant. Thus, curry powders play a significant role in the culinary art and add flavor to otherwise dull foods.

India has been a traditional exporter of spices and curry powders. Despite the fact that there are numerous spice houses in different countries, Indian curry powder still holds its position, and 7.5 to 8.0 million rupees worth is exported to the United Kingdom, Europe, the Caribbean, the United States, Canada, Australia, the Fiji Islands, New Zealand, Hong Kong, and Japan, to name a few.

Great care must be exercised in the manufacture of curry powders. All spices should be bone dry and cleaned to zero percent refraction or foreign matter. Coriander seed, chilies, turmeric, black pepper, cumin, fenugreek, and mustard can be the basic ingredients. Other exotic spices such as cinnamon, clove, nutmeg, and mace should be added judiciously to suit the demand or product in which it is to be used (Madhusudan, 1974).

In India, there is strict quality control of curry powder under the compulsory Quality Control and Preshipment Scheme of the Government of India.

A. Types of Spice Mixtures

1. Curry Powders

In general, there are two broad types of curry powder—salted and unsalted. Within these two categeories, there are mild, medium, and strong curry powders, depending on the relative concentration of mild or strong spices.

2. *Other Spice Mixtures and Seasonings*

Although different brands of curry powders are mostly exported from India, the following types of spice mixtures are being manufactured for home consumption: sambar powder, rasam powder, chutney powder, pickling masala (spice), chicken masala (seasoning), Mutton masala (seasoning), and gram masala (hot seasoning). Their composition varies from region to region, depending on the local taste. Different receipes of a variety of curry powders and spice mixtures have been reported in the literature (Lal and Sadasivan, 1944; Gorsica, 1959; Misra, 1962). Gorsica (1959) patented the composition of a soluble, dry frankfurter seasoning stabilized against caking, loss of color, and oxidative deterioration.

B. Manufacture of Curry Powder

The chief operations in the manufacture of curry powders and other spices mixtures are (1) cleaning of the spices by shakers and blowers, (2) conditioning or drying them to the proper moisture level, (3) grinding or milling, (4) mixing the ingredients by mechanical mixers, and (5) packaging in moisture-proof or hermetically sealed containers.

Operations 1–3 have already been discussed above and in Chapter 5, and item 5 will be discussed in Chapter 8. Only the mixing operations will be mentioned here.

Horizontal trough-type mixers with paddlers are ideal for all sizes of spices. Twin shell blenders with an arrangement for addition of liquids are also widely used. Nauta mixers of the augur type are also common (Ramanathan and Srinivasa Rao, 1974).

In salted curry powders, the addition of high-quality salt of fine texture treated with tricalcium phosphate as a free-flow conditioner is useful. There is a Japanese patent on curry powder (Taiyo Chemical Industry Co., 1971).

C. Physiochemical Changes during Manufacturing Operations

Pruthi and Misra (1963c) have reported some important physiochemical and microbial changes that occur during drying, milling, and mechanical mixing operations in the manufacture of curry powder. Losses in weight, moisture, volatile oil, and volatile reducing substances (VRS) as a measure of aroma (Table XLVI) and reduction in bacterial load in spices and curry powders during both drying and milling operations were observed; these changes are attributed mostly to the rise in product temperature during drying and milling (Pruthi and Misra, 1963c; Pruthi, 1964).

TABLE XLVI CHANGES IN MOISTURE, VOLATILE OIL, AND VOLATILE REDUCING SUBSTANCES OF INDIVIDUAL SPICES DURING DRYING AND MILLING[a]

Spice	Moisture (%)		Volatile oil (%)		VRS[b] (meq $KMnO_4$/g)		Temperature attained during milling and mixing (°C)	Percentage loss in weight	
	Initial	After milling	Initial	After milling	Initial	After milling		During drying	During milling
Chili	4.00	3.00	Nil	Nil	70	20	95	8.11	1.30
Cinnamon	5.75	4.50	—	—	1590	1570	78	3.56	1.18
Clove	8.00	2.50	—	—	3530	3120	45	19.30	7.00
Coriander	5.00	4.00	0.50	0.50	260	250	80	3.31	1.13
Cumin	6.00	4.00	5.75	4.60	270	260	85	8.65	2.00
Curry powder (hand-mixed)	5.00	5.00	2.75	2.70	242	240	30	6.18	3.83
Curry powder (mixed mechanically)	4.50	4.00	2.25	2.00	242	135	42	7.32	6.18
Curry powder	5.00	4.00	1.25	1.00	150	145	65	6.75	3.39
Ginger	6.50	5.50	2.75	2.50	375	335	76	6.05	6.00
Mustard	—	4.50	—	1.00	—	195	—	3.33	
Pepper	10.50	7.50	4.00	3.30	260	115	85	3.92	4.64
Turmeric	5.00	4.50	4.50	4.00	155	110	82	4.41	2.20

[a] From Pruthi and Misra (1963c).

[b] Volatile reducing substances.

D. Types of Seasonings

Seasonings such as curry powders or spice mixtures are blends of two or more spices formulated for a particular food product or dish, such as pizza pie, chicken pot pie, and bologna. The files of a progressive seasoning manufacturer may contain as many as 10,000 different seasoning formulations especially concocted to meet the specific requirements of hundreds of food processors, and hence it would be virtually impossible even to enumerate their names. Suffice it to say that, in general, seasonsings may be divided into three categories: (1) ground spice seasoning, (2) soluble spice seasoning, and (3) a combination of ground and soluble spice seasonings.

In marketing, the seasoning manufactuer gives the food processor a warranty that the flavor will remain stable. This is extremely difficult to do when one works with ground spices only. Besides, the flavor of spice may vary from one crop year to the next, and, even with adjustments, a constant flavor is difficult to attain. However, consistency is possible with soluble seasonings, which are made by adding essential oil and oleoresin, usually to a salt or dextrose or sugar base. Essential oils can be purchased that will fall within 10% of the standard flavor level. Oleoresins can be blended to specific values, with the help of a chemist and a flavor technologist. When such seasonings are used on the salt or sugar base, a fair constancy or uniformity in flavor can be guaranteed.

Soluble seasonings have other advantages, such as (1) freedom from the color of the spice, (2) freedom from microbial count, and (3) convenience and ready availability of the desired flavor. Proper unit packaging and storage will further help to maintain flavor over longer storage periods.

E. Testing for Maintenance of Quality

A qualified chemist, an experienced flavor technologist, and a pot of soup are indispensable tools for measuring and controlling flavor in any seasoning. The soup is usually made by adding 6 ounces of wheat flour and 1½ ounces of common salt to 1 gallon of boiling water, followed by cooling to 140°F. A specified weight of the spice or seasoning is added to 8 ounces of this hot soup. After steeping for 30 minutes, the soup is tasted and compared with products of accepted standard value. Experienced experts in flavor evaluations can detect the slightest difference in flavor, if any.

Chemically, for individual hot spices like capsicums or red pepper, the quality is determined by estimation of capsaicin. Likewise, the pungency in pepper can be determined chemically by estimation of piperine. However, no reliable, accu-

rate, chemical method is yet available for the quantitative determination of pungency of ginger.

VI. SPICE ESSENTIAL OILS

Various specialized requirements of modern food processing have inspired the development of spice extractives, which may be liquid or dry. They may be composed of extracted essential oils or oleoresins, depending on the nature of the subject spice (Fig. 12). As with ground spices, the manufacturers blend the extractives to achieve the desired flavor, performance, consistency, etc. There are several types of spice extractives: (1) spice essential oils, (2) spice oleoresins, (3) spice essences, (4) spice emulsions, (5) spice decoctions, (6) "dispensed" spices or "dry soluble" spices or "instant" spices or "spray-dried" spices, (7) "encapsulated" spices on salt or dextrose as base, (8) "oil-based" or "fat-based" spices, and (9) liquid and dry seasonsings. These will be briefly discussed below.

The aroma and flavoring properties of all spices are attributed to their essential oil content. Essential oils are the volatile oils obtained from plants or from parts thereof by (1) water distillation, (2) steam distillation, (3) enzymatic action followed by steam distillation, or (4) water and steam distillation. The theory and practices of distillation as followed in different countries have been dealt within detail by Guenther (1948–1952) and need not be elaborated on here.

Most essential oils consist of mixtures of hydrocarbons (terpenes, sesquiterpenes, etc.), oxygenated compounds (alcohols, esters, aldehydes, ketones, etc.), and a small percentage of nonvolatile residues (paraffins, waxes, etc.). The oxygenated compounds are more soluble in dilute alcohol and in general are more stable against oxidizing influences. The terpenes and sesquiterpenes, because of their unsaturated character, oxidize easily under the influence of air and light or under improper storage conditions.

To improve the storage stability and other qualities of essential oils, many spice manufacturers produce concentrated terpeneless and sesquiterpeneless oils. These oils are made by countercurrent extraction with polar and nonpolar solvents and by chromatographic separation.

The use of such distilled oils certainly goes a long way in overcoming the major objection of flavor variability, as they are reasonably constant in flavor character, and, if used at a fixed rate, they give an acceptable flavor effect. However, they are but incomplete or ghosts of the real flavor, for they lack the many nonvolatile components that are present in freshly ground spice. In certain cases, pepper and ginger are good examples. The volatile oil only gives the odor of the spice; the bite principles, being nonvolatile, do not appear in the essential

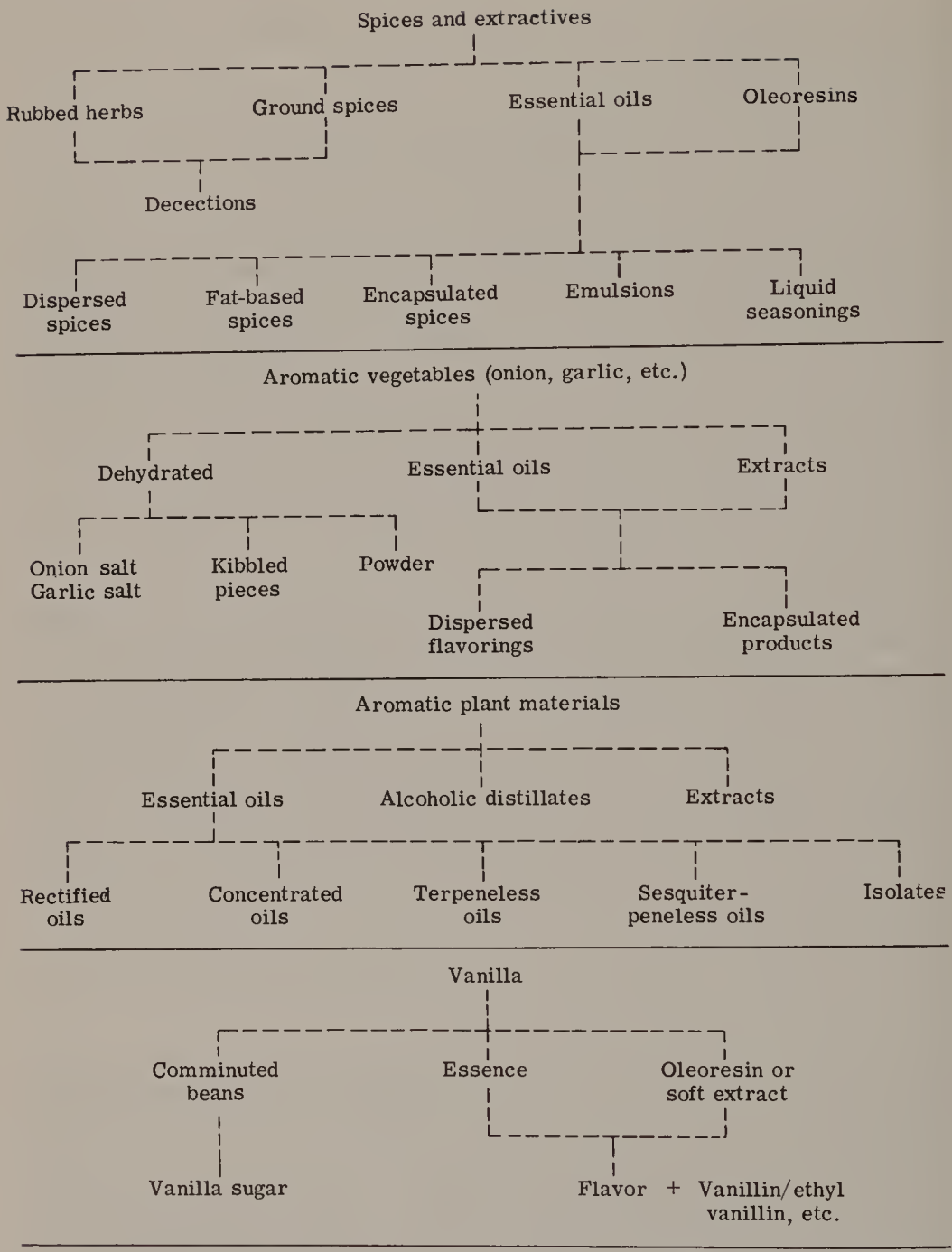


FIG. 12. Spice products technology—a schematic plan of manufacture of spice products and their interrelationship. From Heath (1972).

oil, which is, as a consequence, almost bland. The advantages and disadvantages of spice essential oils are listed below (Heath, 1972).

A. Advantages

1. They are hygienic, being free from all microorganisms.
2. They have reasonable standard flavoring strength.
3. Their flavor quality is consistent with that of the raw material.
4. They do not impart any color to the product.
5. They are free from enzymes.
6. They are free from tannins.
7. They are reasonably stable on storage under good conditions.
8. Color specks are eliminated.
9. Freight expense is decreased.
10. Less storage area is required.

B. Disadvantages

1. Their flavor is good, but incomplete.
2. The flavor is often unbalanced.
3. Some essential oils become readily oxidized.
4. No natural antioxidants are present.
5. They are readily adulterated.
6. They are very concentrated and hence difficult to handle and weigh accurately.
7. They are not readily dispersible, particularly in dry products.

According to Gildermeister and Hoffmann (1956–1961), twenty-four spices out of the seventy spices and condiments listed by the International Organization for Standardization (1968, 1972) were being used for the production of essential oils as early as the sixteenth century (Table XLVII). However, rapid strides have been made in the fields of production, processing, packaging, and utilization of spice essential oils. They are used in modern food preparations, perfumery, medical preparations, cosmetics, etc.

The available information up to 1961 has been nicely compiled and discussed by Guenther (1948–1952) and by Gildermeister and Hoffmann (1956–1961). The salient features of the various steps in the production of spice essential oils and some practical problems relating to steam distillation are summarized here.

C. Production of Spice Essential Oils

1. Preparation of Raw Material

Distillation of spice oils on an industrial scale does differ somewhat from distillation of essential oils in general. In addition to the well-known handbooks

TABLE XLVII ESSENTIAL OILS FROM DIFFERENT SPICES DISTILLED IN THE SIXTEENTH AND SEVENTEENTH CENTURIES[a]

Botanical name	English name	First time reported in the year:
Acorus calamus L.	Sweet flag	1582
Aframomum melegueta (Roscoe) K. Schum.	Madagascar cardamom	1608
Alpinia galanga (L.) Willd	Galanga	1587
Angelica archangelica L. (ex seeds)	Angelica	1592
Carum carvi L.	Caraway seed	1574
Cinnamomum zeylanicum Bl.	Cinnamon	1574
Coriandrum sativum L.	Coriander	1584
Crocus sativus L.	Saffron	1613
Cuminum cyminum L.	Cumin seed	1574
Elettaria cardamomum L.	Cardamom	1540
Eugenia caryophyllus (Spreng.) Bullock et Harrison	Clove	1564
Ferula asafoetida L.	Asafetida	1685
Foeniculum vulgare P. Miller	Fennel	1574
Juniperus communis L.	Juniper	1546
Laurus nobilis L.	Bay or laurel leaf	1540
Myristica fragrans Houttuyn	Nutmeg	1574
Ocimum basilicum L.	Sweet basil	1582
Petroselinum crispum (P. Miller) Nymann ex A. W. Hill	Parsley	1589
Pimpinella anisum L.	Aniseed	1574
Piper nigrum L.	Black pepper	1574
Rosmarinus officinalis L.	Rosemary	1488
Salvia officinalis L.	Sage	1582
Thymus vulgaris L.	Thyme	1589
Zingiber officinale Roscoe	Ginger	1672

[a] From Gildermeister and Hoffmann (1960–1961) and Pravatoroff (1972).

on essential oils by Guenther (1948–1952) and Gildermaister and Hoffmann (1956–1961), there is a comprehensive paper by Ames and Mathews (1968), to which some points have been added by Pravatoroff (1972). An important step in the production of spice oils is the proper preparation of the plant material prior to distillation.

2. Cleaning

Prior to any comminution, the spice must be freed from extraneous matter, such as dust, dirt, pebbles, and metallic objects. For this purpose, sifting machines with a magnetic screen and screens of different diameters are used.

3. Milling

For the grinding of spices, mills of different types are used. Some are equipped with rotating tooth disks, some are hammer mills, and, for some special pur-

poses, triple-roller mills are used. Both the rotating tooth disk mills and the hammer mills have a considerable airflow, which causes a certain loss of volatile oil from the ground spice. The rotating disk mills may be adjusted to the desired grain size of the milled product, but frictioned heat develops during grinding, and therefore they have to be furnished with a cooling device. Hammer mills are not practicable when the spice material is small and contains a relatively high percentage of fat, as, for instance, in seeds from some Umbellifereae. For such cases, crushing in triple-roller mills, which rotate at different velocities and have ribbed rollers, is the appropriate method of comminution.

4. Soaking

Ames and Mathews (1968) mention the desirability of soaking of some plant material—for instance, cinnamon bark—and also of grinding the spice prior to distillation.

5. Loading or Charging

Ground spice must be loaded into the still and distilled without any delay; otherwise, losses of essential oil and deterioration through oxidation are inevitable. The spice oil distiller must realize that his valuable product will someday be used in food without any further preliminary purification. Contamination of the essential oils with rust, dirt, salts of copper, or water cannot be tolerated. Therefore, the distillation apparatus should preferably be made of stainless steel. As a minimum requirement, the lid of the still, the gooseneck, the condenser, and the Florentine flask or oil receiver should be made of this material.

6. Method of Distillation

The choice of the method of distillation—whether water, water–steam, or steam distillation—depends on the kind of spice being processed, and the most economical way of processing to yield a good or excellent quality of oil must be determined. For several spices, an agitated water distillation with cohobation of the distillation waters can be used. The oils obtained in this way are of very high quality. The advantage of this method is that, by floating freely in boiling water, the plant material does not have the opportunity to rest on the heated walls or on the steam coil; a burnt odor in the oil is thus avoided (Pravatoroff, 1972).

A somewhat exceptional design of steam distillation apparatus for one particular spice (coriander) oil shown in Fig. 13 is in use in the USSR. It works continuously and is used for the distillation of coriander seeds (Sokolinikov, 1958). The crushed seeds are fed into the distillation tower by a screw charger and then exhausted by steam while floating downward. In this way, about 10,000 tons of coriander seeds are processed yearly to yield 1000 tons of coriander oil, which is used mainly as a source of linalool and only to a small extent as a spice oil.

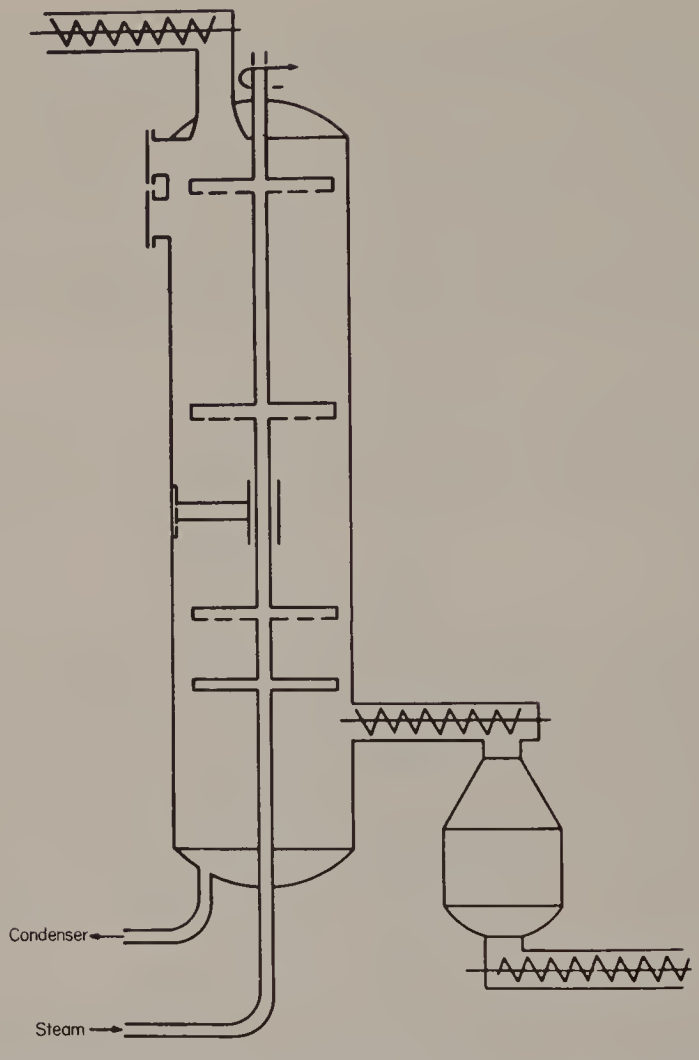


FIG. 13. A special design of continuous distillation apparatus for coriander oil in use in the USSR. Construction of Ponomarenko and Pokolenko, type 2. From Sokolinikov (1958).

7. Problems Relating to Steam Distillation

1. When utilizing steam distillation, even when a grid is used, the danger of introducing a burnt odor into the oil is considerable. The reasons for this are not only that a part of the distillation material may be "steam-dried," but also that a part of the injected steam will condense within the still, extract some water-soluble material from the spice, flow downward, and reach the more-or-less dry, hot steam coil. This danger may be reduced by using a water-sealed drain (van Oss and van Oss, 1956).

2. Concerning the "still" odor of freshly distilled oils, it should be pointed out that some oils do actually improve after a few weeks of storage, but if an oil

is really "burnt," it may be possible to improve it by flushing with nitrogen over a long period. However, this treatment partly removes the top notes of the oil, and the product will not be of high quality (Pravatoroff, 1972).

3. The problem of laboratory-scale separation of the essential oil from the distillation waters has already been mentioned. Working on an industrial scale, it seems reasonable to take the findings of Kashchenko *et al.* (1971) into account. According to this author, the separation of the oil from the distillation waters depends on the following factors: the composition of the oil; the temperature of the distillate; the flow rate of the distillate into the separator; and, finally, the design of the separator. As the temperature of the distillate increases, the specific gravity and the viscosity of the essential oil diminish considerably in comparison with those of water. The effectivity *(K)* of the separation can be represented by the following equation:

$$K = W_2/W_1$$

where W_1 = velocity with which the particles of essential oil are drawn downward at the entrance of the separator, in meters per second, and W_2 = the constant velocity of the uniform motion by which the oil particles rise to the surface, in meters per second.

The velocities W_1 and W_2 and their variation during distillation depend not only on the design of the separator and the velocity of the distillation, but also on the temperature of the distillate. There is, however, no direct proportional relation between the temperature of the distillate and the separability of the essential oil from the distillation waters. The results of the investigation of Kashchenko *et al.* (1971) may be seen in Fig. 14. This shows the percentage of oil suspended and dissolved in water in relation to the temperature of the distillate. It is obvious that the four oils investigated—(1) eugenol-basilicum, (2) coriander, (3) lavender, and (4) pine needles—show a maximum in the range of 25° to 35°C. It is further shown that in some cases it is reasonable, for optimum separation of the oil, to keep the temperature of the condensate at 45° to 60°C. Such a temperature implies that the oil separator should be fitted with a kind of reflux-condenser.

4. Generally, the oil suspended in the distillation waters should preferably be recovered by cohobation of these waters. But sometimes solvent extraction, for instance with hexane or pentane, is used for the isolation of the oil from the water. These extraction solvents must be of satisfactory purity. For preference, pentane should be used.

5. Channeling, clogging, and bumping are some of the other problems of batch-type distillation, which can be eliminated by suitable selection of the steam distributor, particle size, baffles, etc.

8. Filtration and Storage of Spice Essential Oils

This aspect is dealt with in some detail by Ames and Mathews (1968).

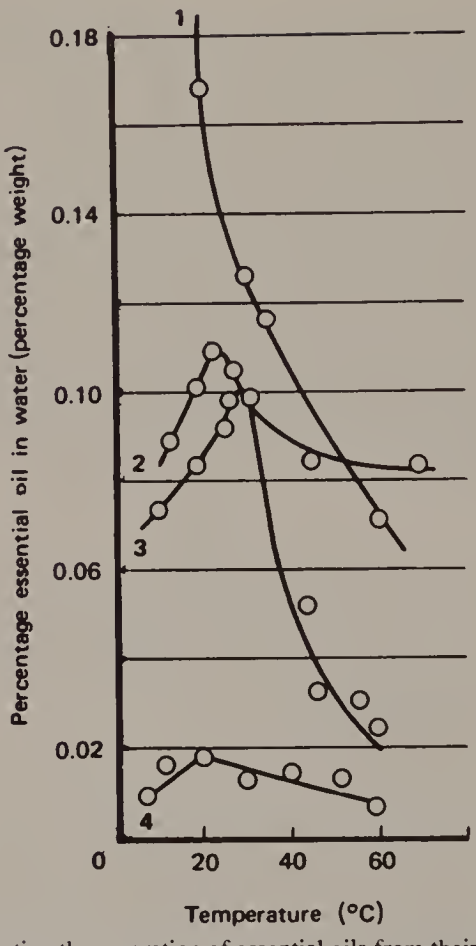


FIG. 14. Diagram depicting the separation of essential oils from their distillate waters at various temperatures of the distillates. Oils of (1) eugenol-basilicum, (2) coriander, (3) lavender, (4) pine needles. From Kashchenko *et al.* (1971).

D. Other Aspects of Spice Essential Oils

Literature references on different aspects of researches on spice essential oils such as production, composition, and volatile constituents are too numerous to be discussed here. They have been adequately covered by Guenther (1948–1952) and Gildermeister and Hoffmann (1956) and reviewed biennually by Guenther *et al.* (1959, 1961, 1963, 1965, 1967, 1969, 1971, 1973, 1975, 1977). To cite a few additional references, ajowan essential oil has been reported by Bhargava and Haskar (1959, 1962a,b) and Nigam *et al.* (1963a); angelica oil by Handa and Kapoor (1958) and Cieri (1969); aniseed oil by Porsch *et al.* (1965), Cook and Haward (1967), and Colombo and Manitto (1971); bay oil by De Martinez Mandal (1959), Nigam *et al.* (1960), Lamparsky (1963), Chow *et al.* (1965), Teisseire (1966), Putkaradze and Taudgiridz (1970), Vorontsova and

Zabolotskaya (1970), Ames *et al.* (1970), and Pruidze *et al.* (1971); sweet basil oil by Dhingra *et al.* (1954), Bruto and Dalvarano (1959), Abdel-Hafez *et al.* (1964, 1966, 1967a,b), Horhammer *et al.* (1964a), Ivanov *et al.* (1964), Pogany *et al.* (1968), Nigam *et al.* (1970), Lawrence *et al.* (1971b, 1972a), and Singh *et al.* (1971); caraway seed oil by Dhaul *et al.* (1958), El-Deeb *et al.* (1962a,b), Deryng *et al.* (1966), Anonymous (1970d), Yin *et al.* (1970), Schultz and Hansen (1971), and Razdan and Kaul (1973); cardamom oil by Ozewalla and Willms (1962), Chopra (1965), Nigam *et al.* (1965a), Lewis *et al.* (1966), Nambudiri *et al.* (1968), Brennand and Heinz (1970), Bernhard *et al.* (1971), and Baruah *et al.* (1973); Greater Indian cardamom oil or Nepal cardamom oil by Nigam and Purohit (1960a,b); cassia oil by Rosebrook *et al.* (1968b) and Heide (1972); Saigon cassia oil by Asakawa *et al.* (1971) and Wijesekera and Jayawardena (1972); celery oil by Farooq (1958), Gold and Wilson (1963a,b), Chopra *et al.* (1968b), Wilson (1969a,b, 1970), Wilson *et al.* (1969), Anonymous (1970a), Ahuja and Nigam (1970), and Baslas *et al.* (1971); cinnamon oil by Brown *et al.* (1954), Hattiangdi and Nimbalkar (1958), Kulkarni (1958), George and Pandalai (1958), Baboob (1961), Ramalanjaona and Jourdan (1968), Lawrence (1969), Wijesekera and Jayawardens (1972), and Wijesekera *et al.* (1972, 1974); cinnamon leaf oil by Wijesekera *et al.* (1974) and Pruthi *et al.* (1975, 1978c); clove oil by Anonymous (1957), Moorhouse (1962), Belcher (1965), Spinks (1969), and Walter (1972); coriander oil by Parczewski (1962), Manolov and Georgiev (1967), Redshaw *et al.* (1971), and Meerov *et al.* (1972); cumin seed oil by Gupta and Patwardhan (1957) and Varo and Heinz (1970a,b); curry leaf oil by Nigam and Purohit (1961); dill oil by Imlhoff (1956, 1957), Kapoor and Abkol (1961), Talwar *et al.* (1963), Singh and Gupta (1964), Baslas and Baslas (1968a,b), Virmani *et al.* (1968), Gulati *et al.* (1969), Virmani and Datta (1970c), Singh and Singh (1970), and Baslas *et al.* (1971); Indian dill (sowa) oil by chakravarti and Bhattacharya (1955b), Gupta *et al.* (1958), Verma (1960), Sethi *et al.* (1965), Chopra *et al.* (1968a,b), Misra and Nigam (1969), and Baslas and Baslas (1971); fennel oil by Fernandes Costa and Cardoso Do Vale (1959), Naves and Tucakov (1959), Kobayashi *et al.* (1967), Peyron *et al.* (1969), Harborne and Saleh (1971), and Lal and Sen (1971); galanga oil by R. N. Chopra *et al.* (1958) and Lawrence *et al.* (1969a); garlic oil by Jacobs (1951) and Schultz and Mohrmann (1965a); ginger oil by Pruthi *et al.* (1960a), Nigam *et al.* (1964), von Jaminet (1965, 1968), Connel (1970a,b), Tihanyi and Zienty (1970), Krishnamurthy *et al.* (1970), Connel and Jordan (1971), and Mathew *et al.* (1973); juniper oil by Handa and Kapoor (1958) and Karlsen and Svenden (1967); lovage oil by Trabaud (1958); mace oil by Forrest and Heacock (1972); marjoram oil by Ilieva and Zolotovitch (1959), Schroder (1959, 1969), Vashista *et al.* (1963), Lossner (1967), and Dayal and Purohit (1971); mint oil by Reitsema (1958), Gupta and Gupta (1959), Handa *et al.* (1964), Chopra *et al.* (1964), Baslas and Baslas (1969), Anonymous (1970b),

Baslas (1970), and Gill *et al.* (1973); mustard oil by Kirk *et al.* (1964); nutmeg oil by Westaway and Williams (1959), Jaurenguiberry and Wolff (1962), Bejnarawicz and Kirch (1963), Shulgin (1963), Shulgin and Kerlinger (1964), Wurziger (1968), and Sammy and Nawar (1968); onion oil by Brodnitz *et al.* (1969), Bandyopadhyay *et al.* (1970), Brodnitz and Pollack (1970), and Gletto (1973); paprika oil by Gerhardt (1968); parsley oil by Garnero and Chrétien Bessiere (1968), Kunz (1970), and Kasting *et al.* (1972); peppermint oil by Virmani and Datta (1970a,b), Anonymous (1970e), and Lawrence *et al.* (1972b); pepper oil by Hasselstorm (1957), Nigam and Handa (1964), Chopra (1965), Lewis *et al.* (1967, 1969b,c), Pruthi (1968b, 1969a), Russell *et al.* (1968), Jennings (1969), Pangborn *et al.* (1970), Russell and Jennings (1970), Richard *et al.* (1971), Russell and Else (1973), and Kahan and Stahl (1973); long pepper oil by Handa *et al.* (1963b) and Nigam and Radhakrishnan (1965, 1966); pimento oil by Nabney and Robinson (1972) and Veek and Russell (1973); rosemary oil by Variati and Rovesti (1960), Karawya and Wahba (1962c), Muller (1963), Abdel-Hafez *et al.* (1967c, 1968), Kalyanasundram and Venkataratnam (1965), Klein and Rojahn (1967), Favro (1968), Grenger *et al.* (1970), Rao and Rao (1971), and Muller (1971); spearmint oil by Chakravarti and Bhattacharjya (1954, 1955a) and Virmani and Datta (1968, 1970b); sage oil by Brieskorn and Dalferth (1964), De Gavina Mugica *et al.* (1969), Devetak and Cenci (1967), and Lawrence *et al.* (1971a); clary sage oil by Genova and Balinov (1970); sweet flag oil by Litchfield and Wilcoxon (1949), Gupta *et al.* (1956), Viseglio-Tomasset (1966), and Larry (1973); tarragon oil by Sinah and Baslas (1968), Thiems and Tam (1968), and Sinah and Baslas (1971); thyme oil by Fibranz *et al.* (1958), Gabel *et al.* (1962), Granger *et al.* (1963, 1965), Messerschmidt (1964), Munchow and Pohleudekfabine (1964), Tucakov (1964a,b), Miller (1965), Schratz and Qedan (1965), Hort (1968), Damjanic *et al.* (1969), Naves (1959), Rovesti (1971), and Russell and Olson (1972); turmeric oil by Burger (1958); and vanilla oil by Bohnsack (1971a,b). Reviews by Sterrett (1962a,b) and bibliographies on spices by Heath (1973) and Sangameswaran *et al.* (1974) are also suggested for further reference on the subject. Papers presented at three spice conferences and symposia held at the Tropical Products Institute in the years 1968 and 1972 and at the Central Food Technological Research Institute (CFTRI), Mysore, in 1974 also deserve the attention of interested readers. Embong *et al.* (1977a,b) have discussed sage and dill oils.

Spice essential oils and their mixtures with resins and gums are commonly found in special secretory structures. The kind or nature of these structures is one of the special characters of a family (Hardman, 1972). For instance, the following families of spices possess the special structures mentioned with them:

Zingiberaceae (cardamom) possess oil cells (Fig. 15A).
Lauraceae (cassia) possess oil cells (Fig. 15B).

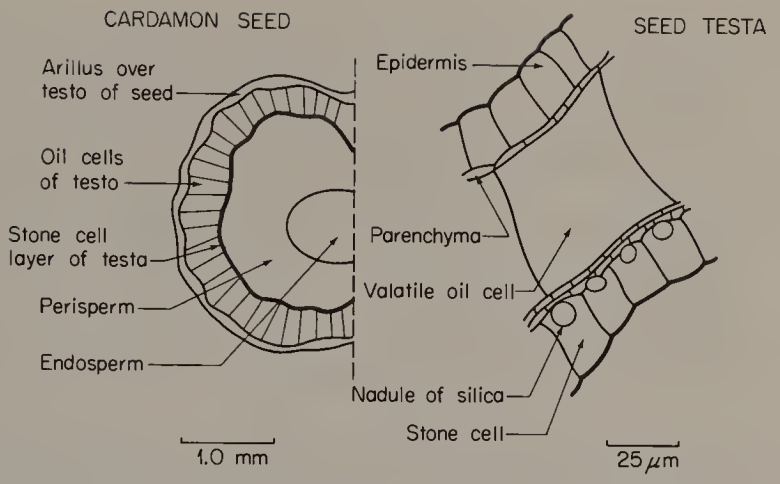


FIG. 15A. Transverse section of cardamom seed testa. From Hardman (1972).

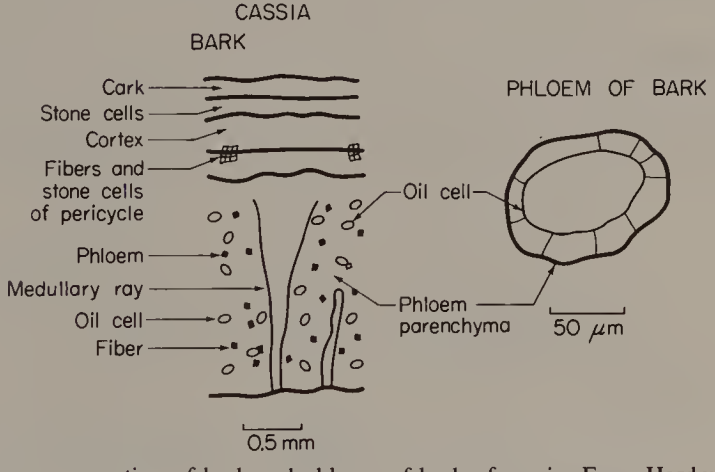


FIG. 15B. Transverse section of bark and phloem of bark of cassia. From Hardman (1972).

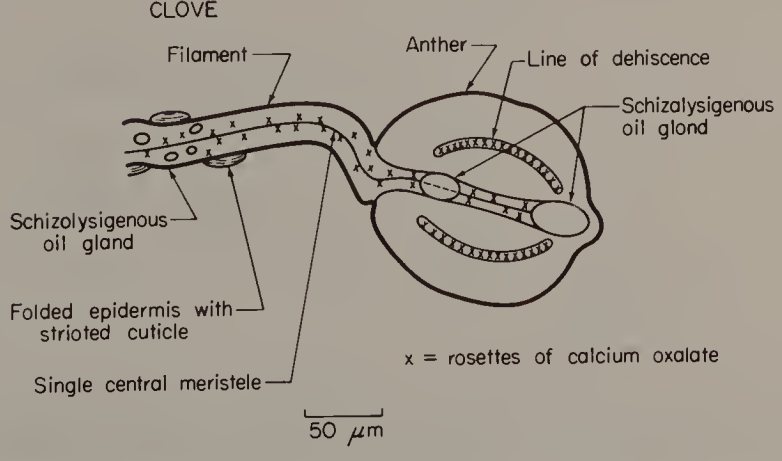


FIG. 15C. Transverse section of clove stamen. From Hardman (1972).

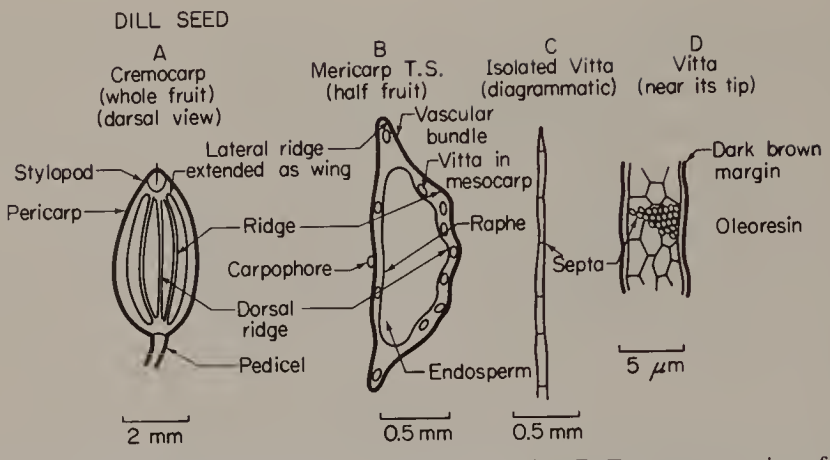


FIG. 15D. (A) Dorsal view of cremocarp (whole fruit) of dill. (B) Transverse section of mericarp (half fruit). (C) Isolated vitta (diagrammatic). (D) A view of vitta (near its tip). From Hardman (1972).

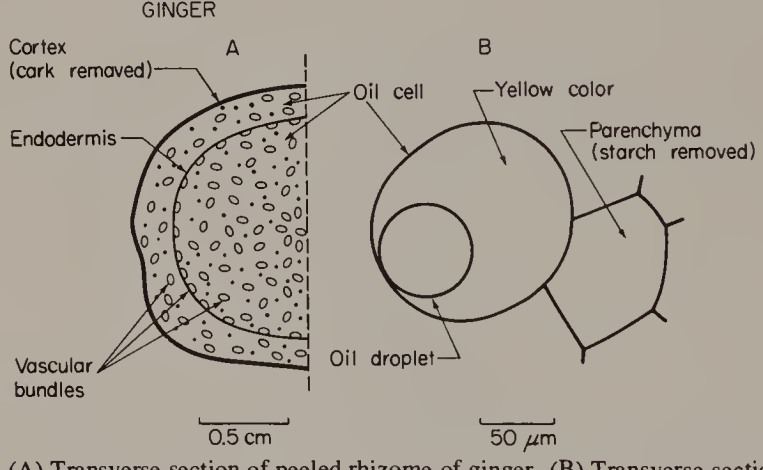


FIG. 15E. (A) Transverse section of peeled rhizome of ginger. (B) Transverse section of parenchyma with oil cell. From Hardman (1972).

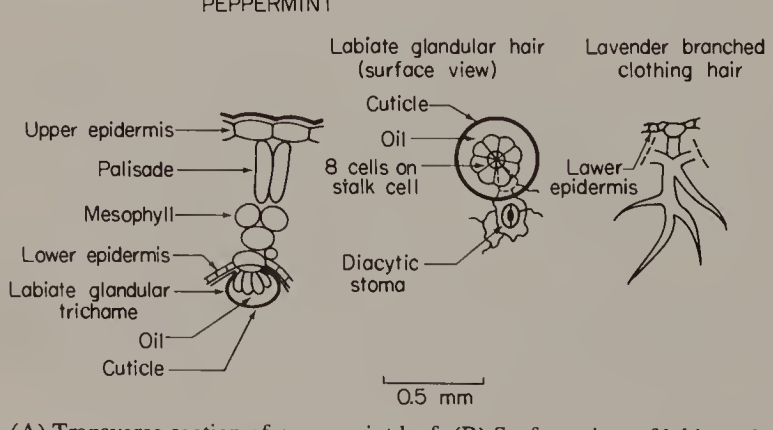


FIG. 15F. (A) Transverse section of peppermint leaf. (B) Surface view of labiate glandular hair. (C) Lavender branched clothing hair. From Hardman (1972).

Myrtaceae (clove) possess oil glands (Fig. 15C).
Umbelliferae (dill) possess oleoresin canals or vittae (Fig. 15D).
Zingiberaceae (ginger) possess oil cells (Fig. 15E).
Labiatae (peppermint) possess labiate trichome (Fig. 15F).
Myristaceae (nutmeg) possess oleoresin cells.
Compositae (tarragon) possess oil ducts.
Orchidaceae (vanilla) possess internal hairs.

Thus, steam distillation problems vary with individual spices, but by judicious selection of the steam distillation unit and technique most of the problems can be overcome.

For batch-type steam distillation, steam chests with arrangements for open steam, condensors, and oil separators are employed. A typical batch-type flow diagram of the steam distillation process for cardamom is illustrated in Fig. 16.

Although some spices have a more attractive odor when they are fresh, the oil can be distilled only from the cured, dry material. The cured spices can be stored and ground easily. In most cases, the oil is extracted easily from the ground material. This is not the case with wet material. The ground, paste-like material can be subjected only to water distillation. The amount of oil being small in the

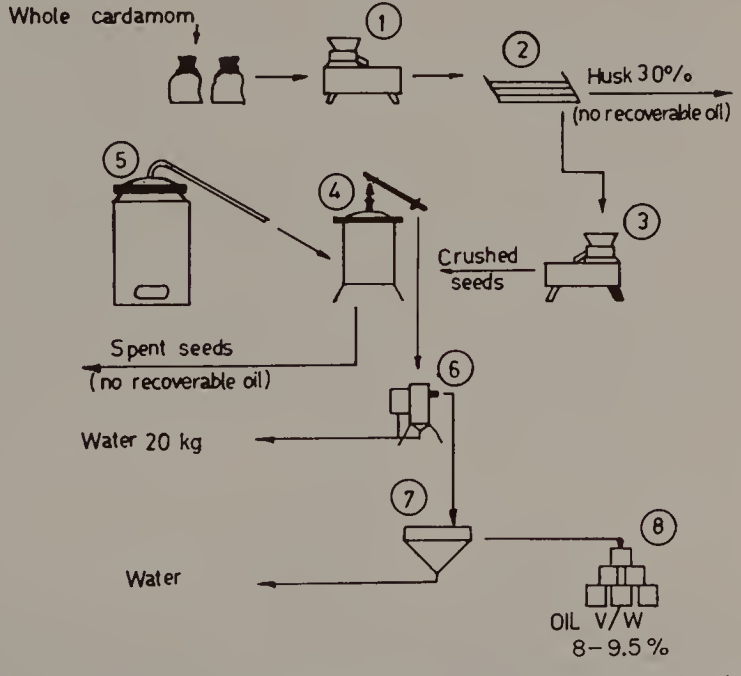


FIG. 16. A flow diagram of the distillation process for cardamom oil. (1) Disk-type mill. (2) Vibrating sieve. (3) Disc-type mill. (4) Distillation still. (5) Steam generator. (6) Continuous oil separator. (7) Conical vessel. (8) Aluminum cans containing cardamom oil. From Nambudri *et al.* (1968).

fresh spice, the retort of the still will have to be unwieldily large. With pepper, grinding itself becomes a problem. In cardamom, the wet material does not give up oil even after many hours of distillation. Because of these reasons, spice oils are generally obtained only by steam distillation of ground, dry material. In some spices, such as cinnamon, two fractions of the oil are obtained, one lighter than water, and the other heavier than water. The oil separator of the distillation equipment should be so designed as to collect both fractions and subsequently mix them. With cloves, a better quality oil is obtained by distilling whole cloves rather than ground cloves, but this increases the distillation time enormously. For several hours in the beginning, no oil at all is obtained. Generally, long distillation times are required to get the fully flavored oils, since the fractions of the oil that have rich odors are high-boiling and distill over very slowly (Lewis *et al.*, 1974).

Thus, the techniques of distillation vary from spice to spice, and the conditions have to be very carefully adjusted to get the maximum yields of best-quality oil at an economical cost, ensuring maximum recovery of all the odoriferous compounds of the oil.

Except in a few cases such as cardamom, clove, nutmeg, and sweet flag, the yields of oil from spices are generally low and quite variable (Table XLVIII). Hence, their costs of production run high, and, as such, the oils can be used for special purposes like fortification or blending or in perfumery. The essential oil, being volatile, escapes easily from processed foods during cooking. Therefore, larger quantities have to be used. However, their dispersal in vehicles such as propylene glycol or fixed oils reduces these losses. Spice oils are used extensively in the flavoring of processed meats, soups, sauces, pickles, baked goods, confectionary, soft drinks, and liqueurs, and in pharmaceuticals and perfumery.

Like other essential oils, the spice oils are also made up to terpenes, sesquiterpenes, and the corresponding oxygenated derivatives. The amount of hydrocarbons in some oils is small (5 to 10%); in others, they form 60 to 95% of the oil (Table XLIX). The oxygenated derivatives are recognized as the really important odoriferous compounds in the oils. However, without the terpenes, which are responsible for the characteristic top notes of the spice, the oils no longer smell like the parent spice.

VII. SPICE OLEORESINS

A closer approximation to the total spice flavor is given by its oleoresin. An oleoresin is an extremely concentrated, viscous, resinous extract obtained by solvent extraction and containing all the flavoring ingredients of spice soluble in the particular organic volatile solvent used. The oleoresins are prepared from the ground dry spice by extraction with a suitable volatile organic solvent or a

TABLE XLVIII VARIATIONS IN ESSENTIAL OIL CONTENT OF SPICES

Spice oil	Yield (%)	Source
Bay leaf	1.0	Tropical Products Institute, London (1968)
Basil	0.2–0.5	Dhingra *et al.* (1954)
Greater cardamom (Nepal)	2.50	S. S. Nigam and Purohit (1960a,b)
Cardamom	5.0–10.75	Lewis *et al.* (1966), Nambudri *et al.* (1968)
Coorg green	6.0–7.0	
Skleshpur bleached	6.0–7.0	Lewis *et al.* (1967)
Allepy green	8.0–10.75	Lewis *et al.* (1967)
Cassia		
Batavia "C"	0.96–1.04	Rosebrook *et al.* (1968b)
Batavia	3.07–3.29	Rosebrook *et al.* (1968b)
Korintze "A"	2.29–2.37	Rosebrook *et al.* (1968b)
Korintze	2.24–2.34	Rosebrook *et al.* (1968b)
Saigon	3.63–3.81	Rosebrook *et al.* (1968b)
Cinnamon/cassia	2.0–3.50	Lewis *et al.* (1974)
Celery	2.0–2.50	Lewis *et al.* (1974)
Cinnamon leaf oil	2.50–2.0	Pruthi *et al.* (1974a, 1975, 1978c)
Clove	16.00–18.00	Lewis *et al.* (1974)
Coriander	0.50–2.50	Lewis *et al.* (1974)
Cumin seed oil	2.30–5.00	Gupta and Patwardhan (1957)
Ginger	1.50–3.50	Lewis *et al.* (1974)
Mint oil	0.32–0.45	Gupta and Gupta (1959)
Nutmeg and mace	5.0–6.0	Lewis *et al.* (1974)
Pepper oil	1.5–3.5	Lewis *et al.* (1974)
Bold pepper (Indian)	2.1	Lewis *et al.* (1967)
White pepper (Indian)	2.4	Lewis *et al.* (1967)
Garbled pepper	2.0	Lewis *et al.* (1967)
Light pepper	3.25	Lewis *et al.* (1967)
Green pepper (*P. nigrum*)		
Panniyur var.	0.9–1.5[a] 2.0–8.4[b]	Pruthi *et al.* (1974b, 1975, 1976b)
Karimunda var.	1.2–2.5[a] 3.6–10.4[b]	Pruthi *et al.* (1974b, 1975, 1976b)
Local var.	1.4–1.9[a] 4.6–6.9[b]	Pruthi *et al.* (1974b, 1975, 1976b)
Peppermint oil		Gupta and Gupta (1959)
England	0.6–1.2	Gupta and Gupta (1959)
France	0.25	Gupta and Gupta (1959)
Germany	0.26	Gupta and Gupta (1959)
Hungary	0.8–1.65	Gupta and Gupta (1959)
India	0.12–0.32	Gupta and Gupta (1959)
Italy	0.20–0.30	Gupta and Gupta (1959)
United States	0.67–1.70	Gupta and Gupta (1959)
USSR	1.50–1.70	Gupta and Gupta (1959)
Spearmint oil	0.25–0.35	Virmani and Datta (1968)
Sweet flag oil	5.1	Gupta *et al.* (1956)
Turmeric	1.5–4.0	Lewis *et al.* (1974)

[a] Fresh-weight basis (%).

[b] Dry-weight basis (%).

TABLE XLIX APPROXIMATE CHEMICAL COMPOSITION OF SPICE ESSENTIAL OILS[a]

Spice	Monoterpenes (%)	Sesquiterpenes (%)	Oxygenated compounds (%)
Pepper	70	25	5
Ginger	5	65	30
Cardamom	8	—	92
Turmeric	10	25	65
Clove	2	8	90
Nutmeg	90	—	10
Cinnamon	10	4	86
Coriander	20	—	80
Cumin	50	—	50
Celery	60	15	25

[a] From Lewis *et al.* (1974).

mixture of solvents; unless great care is taken about the purity of such solvents, traces of the higher boiling fractions may remain in the oleoresin during stripping of the solvent and give it a decided off-aroma. On the other hand, excessive vacuum and heat treatment to remove last traces of solvent, now specifically required by federal regulations of different countries, invariably results in a degree of damage to the heat-labile flavor components or, in extreme cases, to an almost complete loss of the top notes of the essential oil present.

An oleoresin plant or oleoresin unit consists of an extractor for solvent extraction of spices, desolventization units for meal and miscella, and blending units. Jacketed vessles are used for desolventization. Removal of the last traces of solvent poses problems, owing to high viscosity and heat sensitivity. Scraped film evaporators are sometimes employed for this purpose. Handling of resins for blending is also a problem, owing to high viscosity and pungency. Solvent recovery and yield of resin dictate the economics of these units.

As the name implies, oleoresin consists of a blend of the essential oil and resinous matter of the spice and related compounds, nonvolatile fatty oil, and coloring matter soluble in the particular solvent used. The importance of choice of solvent can best be illustrated by considering the extraction of a typical spice—say, turmeric. If one extracts turmeric with petroleum ether, the product is a light, fluid oleoresin, which is highly aromatic and smells strongly of ground turmeric, but has little of its yellow coloring power. If, however, one extracts it with acetone, one gets a brilliant yellow extractive, which is a hard solid with only a very small amount of the characteristic odor. Both of these products are strictly oleoresins, but the difference in their physical condition, chemical composition, aroma, and coloring power is determined entirely by the nature of the solvent that is used in their preparation. Since the herbs and spices are variable, it

follows that the aromatic extractives obtained from them will also be variable, both in flavor characteristics and in flavor strength. The advantages and disadvantages of spice oleoresins, according to Heath (1972), are as follows:

A. Advantages of Oleoresins

1. They are hygienic, being free from bacteria, etc.
2. They can be standardized for flavoring strength.
3. They contain natural antioxidants.
4. They are free from enzymes.
5. They have a long shelf-life under ideal conditions.
6. They have less bulk in storage.
7. They have less weight in shipping.
8. There is no color disturbance or specks.
9. There is no danger of molding as in spices.

B. Disadvantages of Oleoresins

1. The flavor is good but as variable as the raw material.
2. They are very concentrated and hence difficult to handle and weigh accurately.
3. They range from liquids to viscous solids, which are difficult to incorporate into food mixes without hot spots.
4. Tannins are present unless they are specially treated and freed.
5. Flavor quality depends on the solvent used and also on the raw material.

C. Factors Affecting Yield and Quality of Oleoresins

The following factors significantly affect both the yield and the quality of oleoresin.

1. Selection of Proper Variety of Spice

Distinct differences in quality and yield of both essential oils and oleoresins have been noticed in varieties of important spices such as cardamom, chilies, ginger, pepper, and turmeric grown in India (Lewis, 1972). There are four important commercial varieties of ginger in the world market, which differ in appearance, texture, yield, and quality of oil and oleoresin or nonvolatile extract (Table L). The oleoresin content of pepper varies from 10 to 12%, chilies 12 to 16%, ginger 5 to 7%, and turmeric 4 to 7% (Lewis *et al.*, 1974). In twenty-seven varieties of ginger grown in India, the nonvolatile extractives obtained by the cold percolation technique varied from 1.05 to 3.95% when ethylene dichloride

TABLE L VOLATILE OIL AND OLEORESIN CONTENT OF INTERNATIONALLY IMPORTANT COMMERCIAL VARIETIES OF GINGER[a]

Variety	Appearance	Flavor characteristics	Volatile oil (% v/w)	Nonvolatile extract (EDC)[b] (%)
Cochin	Bold, light-brown, partly peeled	Lemon-like odor and flavor	2.2	4.25
Jamaican	Bold, very light buff color, clearly peeled	Delicate aroma and flavor	1.0	4.4
Sierra Leone	Plump, dark, partly peeled from sides	Pungent and slightly camphoraceous flavor	1.6	7.2
Nigerian	Bold, light color, partly peeled, fibrous	Very pungent, camphoraceous flavor	2.5	6.5
Japanese[c]	Dark, bold, unpeeled	—	0.5	4.6

[a] From Lewis *et al.* (1972).
[b] Ethylene dichloride.
[c] Commercially not very important.

was used as solvent, and from 1.9 to 8.34% when alcohol was the solvent (Lewis *et al.*, 1973). Likewise, Cowley (1972) has shown differences in yield and quality of oleoresin from vanilla beans grown in different regions of the world (Table LI). Suzuki *et al.* (1957) and Mathew *et al.* (1971a,b) have also shown wide variations in oleoresin content of several commerical varieties of chilies grown in different regions of the world (Table LII). For Jalpeno pepper varieties, see Weisenfelder *et al.* (1978).

2. *Conditioning or Drying of Spices*

The spice selected must be dried thoroughly to bring down the moisture to a critical level, which is different from the critical moisture content for the prevention of mold growth. This level also depends on the nature of the solvent to be used. For water-soluble solvents such as alcohol and acetone, the moisture level should be very low; otherwise it dilutes the solvent, and this, in turn, lowers the solubility of certain flavoring compounds. Besides, if spices are not dried, they require larger quantities of solvent and larger equipment, with the added difficulty of extraction mentioned above.

3. *Preparation of Material for Pulverizing*

In chilies, the calyx should be removed so that it does not imprat a greenish tinge to the oleoresin. In ginger and turmeric, the hands and fingers should be

TABLE LI VARIATIONS IN OLEORESIN AND VANILLIN CONTENTS AND OTHER PHYSIOCHEMICAL CHARACTERISTICS OF VANILLA BEANS FROM DIFFERENT GEOGRAPHICAL REGIONS OF THE WORLD[a]

Country of origin	Yield of oleoresin (%)		Initial vanillin content (%)		Vanillin content on storage (%)		Moisture content of bean (%)	Winton lead number	Nonvolatile ether extract—oleoresin (%)		Vanillin content on dry-weight basis (%)
	As-is basis	Dry-weight basis	I	II	I	II			I	II	
Reunion	35.2	55.8	1.31	1.34	1.34	1.32	37.0	0.674	79.5	79.8	2.10
Comores	38.7	50.7	1.84	1.85	1.85	1.87	23.6	0.575	84.1	83.7	2.42
Madagascar	29.9	*48.9*	1.39	1.40	1.36	1.40	38.8	0.645	79.8	79.9	2.28
Seychelles	37.9	59.2	1.47	1.49	1.47	1.46	36.0	0.596	81.2	80.5	2.32
Seychelles (dry)	37.6	53.8	1.50	1.48	1.50	1.50	30.0	0.623	83.2	82.8	2.14
Uganda	47.0	60.8	1.26	1.25	1.26	1.23	22.6	0.737	77.2	77.5	1.63
Tahiti	38.5	*64.8*	0.90	0.91	1.01	1.04	40.6	0.578	66.4	67.5	1.52

[a] From Cowley (1972).

TABLE LII VARIATIONS IN OLEORESINS, CAPSAICIN, PUNGENCY, AND COLOR OF CHILI VARIETIES FROM DIFFERENT GEOGRAPHICAL REGIONS OF THE WORLD[a]

Variety	Country of origin	Yield of oleoresin (%)	Capsaicin content (%)	Scoville heat value (calculated from that of oleoresin)	Total color as β-carotene (μg/g)
Whole chili (clipped)					
Sannam	India	16.5	0.33	49,500	1150
Mundu	India	16.0	0.23	34,000	1300
Usimulagu (Bird chili)	India	8.7	0.36	42,000	113
Mombassa	Africa	13.1	0.42	78,600	349
Bahamian	Bahamas	12.5	0.51	75,000	1220
Santaka	Japan	11.5	0.30	55,000	1392
Hontaka[b]	Japan	—	0.33	50,000	—
Mombassa[b]	Africa	—	0.80	1,20,000	—
Uganda[b]	Africa	—	0.85	1,27,000	—
Abyssinian[b]	Africa	—	0.075	11,000	—
Mombassa (whole)	Africa	13.1	3.2	48,000	—
Sannam (whole)	India	16.5	2.0	3,00,000	—
Sannam (pericarp)	India	10.3	3.5	5,00,000	—

Chili components	As % of whole chili	Total extractives (%)	Capsaicin content (%)	Total color content as β-carotene (μg/g)
Whole	100	14.8	0.23	1700
Pericarp	40	10.3	0.52	4000
Seeds	54	23.3	0.04	20
Stalks	6	5.2	—	—

[a] From Mathew *et al.* (1971a,b).
[b] From Suzuki *et al.* (1957).

coarsely cracked into small pieces before they are fed to the pulverizer. Properly dried spices should be cleaned to get rid of all extraneous matter. They are then pulverized to 30- to 40-mesh size. Although fine grinding of spices may give slightly higher yields, it has the disadvantage of caking and channeling, as in ginger and pepper. Chili powders of 0.5- and 0.35-mm mean mesh gave yields of 12% and 16% oleoresin, respectively (Nambudiri *et al.*, 1970; Mathew *et al.*, 1971a,b).

4. Choice of Solvent

The selection of solvent has received due attention from several workers. Sabel and Warren (1972) reported that, out of a number of solvents studied, acetone, ethanol, and isopropanol could be used very satisfactorily for the recov-

ery of oleoresin from ginger. There was little to chose among these three solvents. Mathew *et al.* (1971a) studied the comparative efficiency of hexane, ethanol, and ethylene dichloride and concluded that hexane was a poor solvent for extraction of capsaicin from capsicums or chilies. On the other hand, alcohol did not extract the color efficiently. Furthermore, alcohol, especially hot alcohol, gives a product that is semisolid instead of the usual free-flowing products. Thus, ethylene dichloride is a good solvent for the extraction of chili oleoresin, getting yields up to 13 to 14%.

Acetone and alcohol have been widely used for ginger extraction. Of course, acetone is costlier and more volatile than alcohol. However, the quality of oleoresin from acetone was found to be better than that from alcohol. In some varicties of ginger, the yield of oleoresin was too high (up to 20%) (Connel, 1970a,b; Lewis *et al.*, 1972). Because of the dilution of the solvent with the water present in the spice and hence the chances of extraction of more nonflavor components, it is preferable to use a water-immiscible volatile organic solvent such as ethylene dichloride for extraction. Lewis *et al.* (1972, 1974) have given the comparative values for four solvents (acetone, alcohol, hexane, and ethylene dichloride) with the cold percolation method of extraction.

5. Conditions of Extraction

Several conditions of extraction, such as the time taken for the solvent to wet the bed of material and the rate of flow of the extract, are commonly influenced by the size of the particles, the density of packing, the bed height, the area, the rate of percolation, etc. Optimum conditions for quick and efficient extraction have thus to be worked out for each spice, since time is a very important factor in commercial operation of the process. The method of extraction is yet another important factor to consider. Systematic work done at CFTRI, Mysore, shows that three—stage batch countercurrent extraction is satisfactory for all the major spices (pepper, ginger, chilies, turmeric, etc.). In addition, ethylene dichloride has been found to be the most suitable solvent because (1) the final product is clear and (2) open distillation can be used for desolventization (Nambudiri *et al.*, 1970; Mathew *et al.*, 1971a; Lewis *et al.*, 1972).

6. Solvent Stripping form Miscella

Great care should be exercised to minimize the hcat damage to the product during the disolventization of the miscella. Connel (1970b) has observed that excessive heating during removal of solvent from ginger extract can cause the formation of shogaol, zingerone, and aliphatic aldehydes. This can result in a decrease in pungency as well as the development of off-flavors. Thus, it is very necessary to control the temperature of the material carefully during distillation by use of vacuum. Constant stirring prevents overheating near the walls of the still. Volatile oil losses can be minimized by controlling the rate of distillation

and by using tall columns. High essential oil content in oleoresin is necessary to get a good-quality oleoresin of the desired pleasant aroma. Chili oleoresin is, of course, an exception, since it does not contain any essential oil (Mathew *et al.*, 1971a; Lewis *et al.*, 1974).

Unlike pepper, there is no necessity for "contact time" in the case of chilies for maximum extraction of solids. The diffusion of solubles into the solvent is fairly quick. Handling of both chili powder and extract requires special precautions, since they are highly pungent and irritate the skin and mucous membrane (Mathew *et al.*, 1971a).

The last traces of solvent are removed from oleoresin by vacuum distillation, followed by an azeotropic distillation technique.

D. Location of Oleoresin Plant

The location of the extraction plant, whether at the source or near the market, affects the marketing and economic aspects of oleoresin. Following are the major considerations to be taken into account at the time of location of a plant:

1. Availability of contemplated raw material
2. Reasonable cost and good quality of raw material
3. Scope for marketing of the product
4. Efficient plant design with minimum losses in recovery
5. Availability of technical expertise and management
6. Reasonable facilities for labor, transport, and utilities
7. Assurance of reasonable profits.
8. Government regulations regarding labor and transport
9. Availability of sufficient financing
10. Facility for economic disposal of spice waste, etc.

However, the ultimate success of the plant or project lies in the entrepreneur's ability to interpret the facts correctly, to translate his ideas into action, and to achieve the goals he has set for himself and the enterprise. Systems and check lists help in organizing routines, but in the final analysis, the key to success lies in the people who are behind the undertaking. This is true for all human endeavor and so also for spice extraction, whether at the source or at the market.

E. Product Evaluation

The quality evaluation of oleoresins manufactured from major spices such as pepper, chilies, ginger, and turmeric have been studied, reviewed, and discussed by Todd (1960), Pruthi *et al.* (1960a), Ikeda *et al.* (1962), Stahl (1965, 1972), Pruthi (1968a), Anonymous (1970b), Nambudiri *et al.* (1970), Shan-

karanarayana *et al.* (1970), Mathew *et al.* (1971b), Lewis *et al.* (1972), Sabel and Warren (1972), Wijesekara *et al.* (1972), Govindarajan (1973), Govindarajan and Raghuveer (1973), Govindarajan *et al.* (1973), Ananthakrishna and Govindarajan (1974), and Govindarajan and Ananthakrishna (1974).

More than thirty-seven published reports (up to 1970) on the use of different techniques for the estimation of capsaicin in capsicums have been reviewed in Chapter 3 and summarized in Tables XXIX and XXX. Subsequently, Mathew *et al.* (1971b) achieved the separation of capsaicin from other constituents in the capsicum oleoresin by TLC. The method is claimed to be simple and convenient for adoption as a routine method, but Govindarajan and Ananthakrishna (1974) have since developed a simpler, rapid, and accurate paper chromatographic method for the determination of capsaicin.

Govindarajan *et al.* (1973) also developed an interesting profile of ground black pepper through description of the odor of the components from column fractionation of the essential oil and total oleoresin of pepper as put in final form by a trained panel at round-table discussions. It has been successfully applied to different varieties of Indian black pepper (Fig. 17) as well as to trade types and is claimed to be useful in the selection of new, superior varieties of pepper, in conjunction with objective data on color, size, pungent principles, and yield taken together.

Govindarajan and Raghuveer (1973) developed a three-dimensional TLC technique that provides a composite picture of the group of components in the essential oil and oleoresin of ginger with different odor notes or characteristics. Ananthakrishna and Govindarajan (1974), after briefly reviewing the earlier methods of estimation of pungent principles in ginger, reported better separation of pungent principles (gingerol and shogaol) from the closely related nonpungent substances in ginger oleoresin by TLC under *unsaturated conditions* (Fig. 18A) than was obtained earlier by Connel and Sutherland (1969) under *saturated conditions* (Fig. 18B). Ananthakrishna and Govindarajan (1974) also obtained lower values for these two pungent principles than were obtained by Connel and Sutherland (1969). The higher values have been attributed to the possible interference of some nonpungent principles in ginger oleoresin. The method described is claimed to be quite suitable for routine evaluation of ginger oleoresin, etc., and awaits its utilization by those concerned.

F. Forms or Types of Oleoresins

In some food applications, essential oils and/or oleoresins may be used "as is." However, because of their limited solubility, and the minute quantities required to produce strengths equivalent to those of natural spices, spice manufacturers usually further process the extractives into forms that are more convenient to use from the standpoint of both solubility and strength.

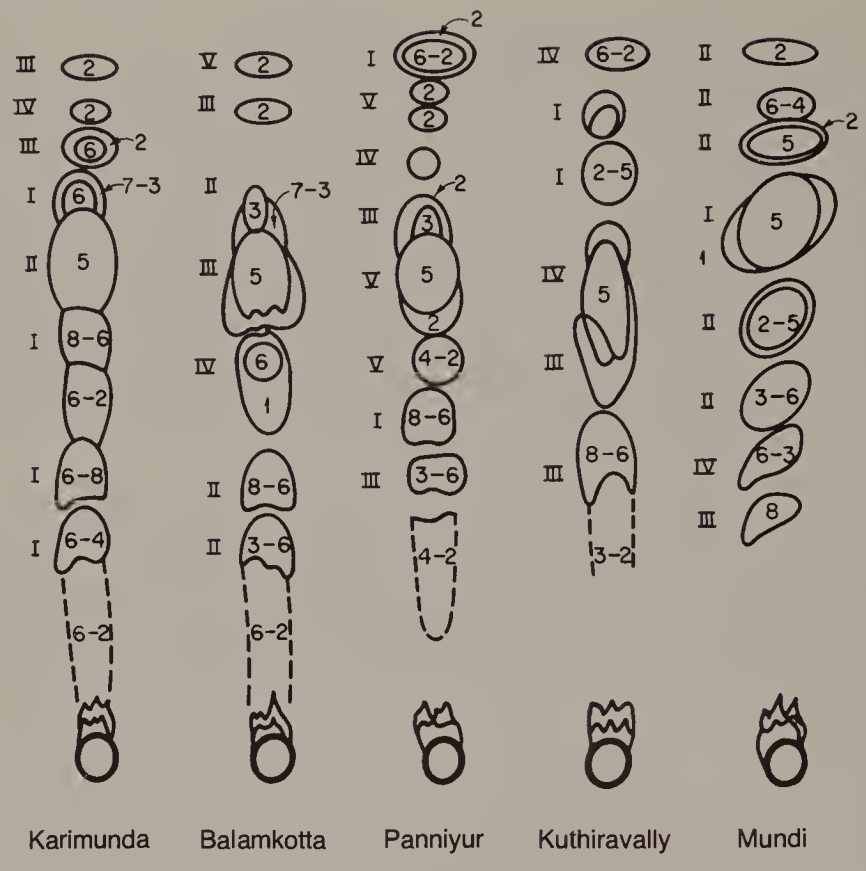


FIG. 17. Tracing of TLC analysis of oleoresins from five varieties of Indian black pepper. From Govindarajan *et al.* (1973).

1. Liquid Forms of Oleoresins

Liquid forms of oleoresins and essential oils are available in both oil-soluble and water-dispersible types as follows:

a. Oil-Soluble Types. In the oil-soluble types, the spice extractives are extended in a medium that will allow for quick and complete solubility in such products as shortenings, cake or cookie dough, icings, and any other food with high fat or oil content.

b. Water-Dispersible Forms. Oleoresins and essential oils are made water-dispersible by blending the extractives with a solubilizing agent such as Polysorbate 80. Solubilized spice preparations of this type may be added directly to water; they will spread readily throughout the vehicle, facilitating uniform dispersion of flavor and aroma in the finished food. Pickle packers, sausage makers, and soup and catsup processors often use this type of spice in their products.

2. Dry Forms of Oleoresins

Dry forms of oleoresins and essential oils consist of two types of products: (1) dry, soluble and (2) spray dried. Both forms are soluble or dispersible in the

mediums in which they are intended to be used. They are discussed later in this chapter.

3. Oleoresin Capsicum or Paprika

As early as 1949, Goldman described methods for the preparation of spice oleoresins—extracts of red and black pepper, paprika, celery, sage, cloves, and ginger—by extraction with Me_2CO or EtOH, filtration, and removal of the solvent by distillation. Soaking the cake before extraction improved the yield. A diagram of a commercial-size extraction apparatus based on a Soxhlet method is given in his report. Later, Berry (1935) reported examination of the extractives of capsicums. The oleoresins of capsicum vary in appearance, solubility, and degree of pungency according to the solvent used for extraction. Et_2O and alcohol extract much of the nonpungent matter from chilies.

Ferns (1961a,b) compared various methods of extraction of oleoresin from capsicum. A laboratory-scale countercurrent appartus was found to be the best. Acetone was selected as the best solvent for the extraction of oleoresins from capsicum. Its properties are discussed in relation to the extraction of essential oils.

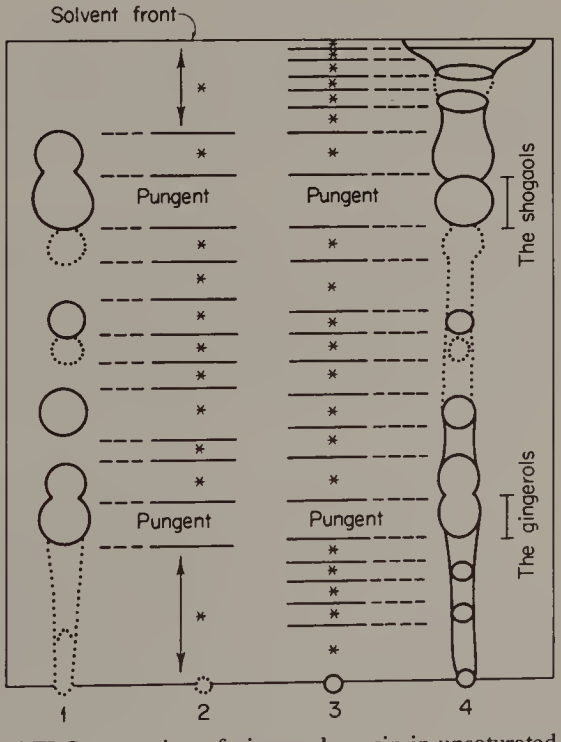


FIG. 18A. Tracing of TLC separation of ginger oleoresin in unsaturated chamber. (1) Preparation of total pungent substances according to Parry from ginger oleoresin—sprayed with Folin-Ciocalteu reagent. (2) Sample as 1—not sprayed, for control taste-testing. (3) Ginger oleoresin—not sprayed, for control taste-testing. (4) Sample as 3—sprayed with Folin-Ciocalten reagent. Asterisks indicate areas showing no pungent sensation by taste-testing. From Ananthakrishna and Govindarajan (1974).

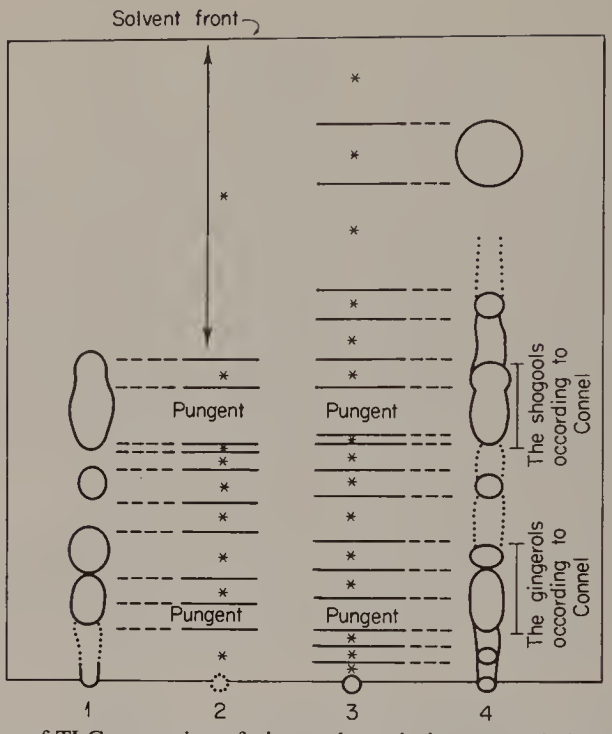


FIG. 18B. Tracing of TLC separation of ginger oleoresin in saturated chamber. (1) Preparation of total pungent substances according to Parry from ginger oleoresin—sprayed with Folin-Ciocalteu reagent. (2) Samples as 1—not sprayed, for control taste-testing. (3) Giner oleoresin—not sprayed, for control taste-testing. (4) Samples as 3—sprayed with Folin-Ciocalteu reagent. Asterisks indicate areas showing no pungent sensation by taste-testing. From Connel and Sutherland (1969).

Tandon *et al.* (1964) discussed the preparation of oleoresin of capsicum (red chilies) as well as its technological and chemical aspects.

Szabo (1969, 1970) discussed the manufacture of paprika oleoresin, and Mathew *et al.* (1971a) covered in detail the preparation and quality control of oleoresin capsicum. There are other reports on oleoresin from Hungarian paprika (Anonymous, 1972) and a Japanese patent on cayenne pepper extract (Sun Spice Co., Ltd., 1970).

Blazovich and Spanyar (1969) have described a method for the determination of capsaicin in oleoresin and some other preparations with high capsaicin content, while Orsi (1970) reported gas chromatographic detection of the acetone content of paprika oleoresin. Govindarajan and Ananthakrishna (1970) described the separation of capsaicin from capsicum and its oleoresin, and Mathew *et al.* (1971b) developed an improved TLC method for the estimation of capsaicin in capsicum oleoresin.

4. Oleoresin Ginger

According to Winterton and Richardson (1965), the yield of oleoresin extracted from Australian dried ginger varies with the time of harvest and the

solvent used. Extraction with 95% EtOH gave more than three times as much oleoresin as extraction with Me_2CO, but the Me_2CO extraction was more acceptable. Extraction with 1:1 EtOH–Me_2CO yielded approximately 60% acceptable extract. Extraction with 1:1 EtOH–Me_2CO yielded approximately 60% more oleoresin than with Me_2CO alone, and the extractive had satisfactory flavor. Connel (1970b) discussed in some detail the chemistry of the essential oil and oleoresin of ginger. Lewis *et al.* (1972) reported the results of their investigation on the standardization of the conditions of manufacture of ginger oleoresin. After a comparative study on the efficacy of four solvents—acetone, alcohol, hexane, and ethylene dichloride—they recommend ethylene dichloride.

A new approach to spice processing, including studies on laboratory-scale and pilot-plant scale preparation and examination of oleoresins from undried ginger, has been reported by Ashurst *et al.* (1972), who conclude that, despite the large volume of solvent required for effective extraction of oleoresin, the new product should be competitive for a given level of pungent substances with those at present available. However, it requires the cooperation of the flavor and other industries to establish this product commerically.

5. Oleoresin Pepper

Narayanan *et al.* (1964) reported a comparative study on different methods of extraction of oleoresin from pepper rejections as well as its storage stability. Natarajan *et al.* (1967b), while briefly reporting the preparation of oleoresin from black pepper, concluded that light pepper is the best raw material for its manufacture. Ziegler (1969) has patented the process for liquid pepper in Canada, West Germany, and the United States. Nambudiri *et al.* (1970) have described the factors affecting the manufacture of oleoresin of pepper. Shankaranarayana *et al.* (1970), Wijesekara and Jayawardena (1972), and Wijesekara *et al.* (1972) discussed the methods for the quality control of oleoresin of pepper. Govindarajan *et al.* (1973) developed a descriptive profile for pepper oleoresin, etc.

6. Thyme Liquid Extract

Nazarov (1959) reported the determination of the quality of liquid extract of thyme.

7. Oleoresin Turmeric

Earlier work on curcumin and nonvolatile extract has been reviewed by Srivas *et al.* (1963b). Eiserle (1966) and Cripps (1967) have briefly discussed the role of oleoresin turmeric in the pickling process. Krishnamurthy *et al.* (1975) have studied the conditions for the manufacture of oleoresin turmeric.

8. Other Aspects

Other aspects covered in the literature include the following: Oleoresins for the flavor chemists (Langenau, 1959); Modern trends in the application of spice (Clark, 1970); Evaluation of oleoresins (Anonymous, 1970c; Gilbertson, 1970); Oleoresins as flavor ingredients (Gilbertson, 1971); Chemical components of the benzene extract of cassia Saigon (Asakawa *et al.*, 1971); Oleoresin quality analysis: facts or fancy (Stahl, 1972); Theory and practice of oleoresin extraction (Sabel and Warren, 1972); Methods for the study of the aromatic components of vanilla extracts (Oliver, 1972); Vanilla and its uses, including oleoresin content as well as vanillin content and other characteristics of vanilla beans from different geographical regions of the world (Cowley, 1972); and Spices or oleoresin: a choice? (Staniforth, 1972). Cripps (1972) discusses the process, the market, and the future for oleoresins. Adamson (1971) discusses production and markets for oleoresins in the United Kingdom. Ahmad *et al.* (1976) discuss the effect of oleoresins on residual toxicity of pyrethrins.

VIII. SPICE ESSENCES AND EMULSIONS

A. Spice Essences

Because of the high concentration of the essential oils and the oleoresins and their limited solubility, most food manufacturers make use of dilutions in one form or another. This overcomes the grave errors of weighing out small quantities of powerful flavors; a small error can have a profund effect upon the final product. Such dilutions are achieved by the use of acceptable solvents such as glycerol, propylene glycol, and isopropyl alcohol, all of which are harmless in foodstuffs, although there are problems of acceptability on a legal basis in some countries (Heath, 1972). The liquid essences have enjoyed long popularity and are most suitable for use in canning processes. The popular essences used by the trade are allspice, basil, caraway, cardamom, cassia, cayenne pepper, celery seed, cinnamon, cloves, coriander seed, fennel, garlic, ginger, mace, marjoram, nutmeg, peppermint, spearmint, and vanilla (Stobart, 1970).

Bohnsack (1965) has discussed the methods of preparing pepper flavors and cinnamylideneacetyl piperidine. Kneilman (1970) has reported changes in regulations regarding essences, and Yelleracamino (1971) discusses rosemary essence in Spain and in Mediterranean countries. The literature on the scientific and technological aspects of spice essences is rather scanty.

The main disadvantages of essences are as follows: (1) Many manufacturers object to the presence of the solvent. (2) The essences cannot be used in dry soup mixes owing to difficulties of dispersion. (3) Also, some of the finest overtones

of flavor are sometimes lacking. (4) Commercial essences sometimes contain synthetic flavoring compounds, as in vanilla essence. One should verify the quality at the time of purchase from a manufacturer of repute.

According to Liverseage (1932), the tincture and essences of capsicum are used to increase the pungency in chewing and smoking tobacco, ginger, ginger ale, ginger soda, rum, etc.

B. Spice Emulsions

An alternative to the use of essences containing solvents is to present the blended oils and oleoresins in a water-dispersible form. This normally would be as an emulsion. Emulsions appear to have practical application in the manufacture of such products as soups and in canning, but it should be remembered that most of the essential oils are composed of highly reactive chemicals, and that hydrolysis, with the consequent change in flavor character, readily occurs in such emulsions during storage. Unfortunately, many of the seasoning emulsions available at present make use of low-grade essential oils only, mostly because the oleoresins are more difficult to emulsify and therefore result in a more costly process. Such emulsions are no substitute for the best seasonings that can be achieved (Heath, 1972).

Published literature on spice emulsions is scanty. Corran (1934), after discussing certain general aspects of spice emulsification in practice, describes his experiments on the preparation of emulsions, with particular reference to mayonnaise, using mustard as an emulsifying agent. Kilgore (1932) also demonstrated the emulsifying qualities of mustard by using different methods.

1. Types of Emulsions

An emulsion of oil in water, such as the mayonnaise type, consists of globules of oil floating about or dispersed in water. An emulsion can also take the form of globules of water dispersed in an oil. This latter type of emulsion is of the "greasy" type, which is generally not appreciated. The former is the "creamy" type. The best-known emulsion of this category is mayonnaise. The creamy type can be differentiated from the greasy type by its rapid miscibility with water; the greasy type of emulsion does not mix with water (Corran, 1934). As was pointed by Clayton (1918), the electrical conductivity of the greasy type of emulsion is decidedly less than that of a creamy one.

2. Emulsifying Agents

The most important factor in determining the emulsion type is the nature of the emulsifying agent or agents. An emulsifying agent present in the emulsion wraps itself around the globules of the dispensed phase and prevents their aggregation and separation. It has the effect of lowering the interfacial tension between the

two phases or of increasing the viscosity of the system, or both. Certain emulsifying agents, when added to an oil:water mixture, favor the oil-dispersed-in-water or creamy type, whereas others favor the greasy type. Generally, those emulsifying agents that are more readily wetted by water than by oil tend to form a creamy emulsion, and vice versa (Corran, 1934).

3. Mayonnaise

One of the most important applications of the emulsifying qualities of mustard is in mayonnaise. Mayonnaise consists essentially of a creamy emulsion of vegetable oil in vinegar. The emulsifying agents present are mustard and egg yolk. Other substances are added for flavoring purposes, but they do not materially affect stability. The specific role of these emulsifying agents has been discussed by Corran (1934), who has proved that mustard not only is useful for its condimental qualities in mayonnaise, but, in addition, exerts a valuable emulsifying effect, which helps in the promotion of stability in mayonnaise.

The stability of mayonnaise was measured by means of the Gardner mobilometer (Gray and Southwick, 1929). If a sample withstood one hundred plunges using a 1000-g weight, it was regarded as stable; if it broke, its limit of stability had been reached. The relative stability of batches of mayonnaise was measured by the number of days the mayonnaise would stand before it broke down within the first hundred plunges of the mobilometer. Krinitz (1971) reported infrared detection of false positives for gums in mayonnaise due to mustard flour.

IX. LIQUID SPICES

Another alternative liquid spice product is prepared from the essential oils and certain oleoresins by admixture of these with such acceptable solubilizing agents as Tween 80 and propylene glycol (Saiba Industries, Bombay, official communication, 1971). These products are water-soluble, depending on the concentration used, but, in addition to their lack of stability, they do tend to give to the product a slightly soapy note and may, under certain conditions, result in some frothing (Heath, 1972).

According to Gisske and Heidtmann (1958), the seasoning power of liquid spice mixtures corresponds to that of conventional spice mixtures. They improve the red color of sausages and give better color stability. However, liquid spice mixtures show counts of 10 to 450 microorganisms per cubic centimeter.

According to Saiba Industries, Bombay (private communication, 1971), liquid masalas (liquid spice mixtures or blends), because of their liquid state, purity, and concentration, inherit the following advantages over dry powdered spices.

A. Advantages

1. They are highly potent. For instance, 1 ounce of liquid red chilies, black pepper, or cumin is equivalent to 1 pound of the corresponding spice powder, and 1 ounce of liquid cardamom or liquid nutmeg is equivalent to 6 ounces of the corresponding spice.
2. They are more stable than whole or ground spices.
3. They are economical in use; much less in required than with ground spices.
4. They are favorable to the human system; no harm is done.
5. They are uniform in quality for better and easier blending.
6. They are more hygienic and clean.
7. They are compact, requiring less space.
8. They are easy to use.

B. Types of Liquid Spices

Liquid spices may consist of individual spices or a mixture of spices called liquid seasonings or liquid masalas in definite ratios to suit different types of curries and dishes. The type of spices to be used is dictated by the formulation, the method of manufacturer, and the anticipated shelf-life of the product in which it is proposed to be used.

C. Quality Evaluation of Liquid Spices

Liquid spices are tested for their flavor and pungency by the olfactory method, using the standard neutral soup technique. They contain up to 35% of the oleoresins concerned. The limit for the carriers (Tween 80 and propylene glycol) varies from 70 to 93%, depending on the spice (Saiba Industries, Bombay, private communication, 1971). There is need for standardization of liquid spices—at least of individual spices to begin with.

X. SPICE DECOCTIONS

In the sauce and pickle industries in particular, it is quite common to prepare spiced brine and vinegars by boiling the spices in water or vinegar and then use the resulting liquor. Such a liquor is called a decoction. Decoctions have the advantage of being sterile and soluble, but, as water is not a good solvent for the flavor constituents of the spices, only a very small percentage of the flavor is extracted. As one can imagine, the boiling of herbs and spices with water creates an ideal condition for volatile loss, and very little of the true aroma of the herbs

and spices is left in the finished product. This method of incorporating spice flavors is to be deplored strongly (Heath, 1972).

XI. DISPERSED SPICES OR DRY SOLUBLE SPICES

The total flavor components extracted from a spice are dispersed onto a soluble base such as salt, dextrose, milk whey, or flour, so that the strength of the flavor is equal to that of a freshly ground sample of a good-quality spice. This is, of course, the ideal, but many products do not live up to this standard. Such products may have been made by dispersing essential oils of a lower flavoring power than the ground spices.

Dispersed spices have the advantage of being able to replace the natural spices on a one-for-one basis in existing recipes (Heath, 1972). Haffof (1954) has described a method of manufacture, packaging, storage, and handling of instant spices or dry solubles. Baltimore Spice Co. (1957) has described special flavoring preparation by a vacuum absorption process. Gorsica (1959) has described stabilized seasoning composition by incorporation of antioxidants (BHA or BHT). Gisske (1954) reported sterile spice preparations (Dex pepper, Dex mace, Dex cardamom, etc.) for sausages, etc. Todd (1960) has studied in detail the recovery and estimation of solvents from dispersions of oleoresins on salt or dextrose, or from dry soluble seasonings, and also from several oleoresins.

The advantages and disadvantages of dispersed spices are listed below.

A. Advantages

1. They have a standardized flavoring effect.
2. They have a standardized flavor quality; the source of the raw material can be specified.
3. They are hygienically excellent, being free from bacterial filth and other impurities.
4. They are free from enzymes.
5. They are easily handled and can be weighed with accuracy.
6. They are readily dispersed in food mixes.
7. They are usually free from dust during handling.
8. They contain natural antioxidants.
9. They have a low water content.
10. They do not contribute unwanted specks or color to the end product.

B. Disadvantages

1. Allowances are necessary for the base used.
2. They lose volatiles on long storage, particularly if the ambient temperature is high.
3. They do not resist high-temperature processing under open conditions.

XII. ENCAPSULATED SPICES

In the production of spray-dried spices, the essential oils and/or oleoresins are dispersed in an edible gum solution, generally gum acacia or gelatin, spray-dried, and then blended with a dry base such as salt or dextrose. As water evaporates from the sprayed particles, the gum forms a protective film around each particle of extractive. The spice extractives are thus "entrapped" or "locked" in a capsule of edible, water-soluble gum, usually arabic (Neale and Klis, 1964).

The protective capsule prevents the spice extractive from evaporating and from being exposed to oxygen, which might have a deleterious effect on the flavor. Thus, these encapsulated spice extractives retain flavor longer than dry, soluble or ground spices. Each spray-dried particle has a diameter of about 150 microns and contains several minute globules of oil or oleoresin less than 15 microns in size. When the gum coating is dissolved and the extractives are released, the droplet size is sufficiently small to create a stable dispersion (Maleeny, 1961; Neale and Klis, 1964).

Spray-dried spices are free-flowing and dispersible in all water solutions. Thus, they have applications in such dry-mix food products as dehydrated soups, sauces, gravy, desserts, and beverage mixes.

The base, which is normally gum acacia or gelatin, is expensive. To cover this expensive and the cost of processing, it is necessary to make these products much stronger than the single strength usually aimed at in the simple dispersed spices. Many such encapsulated spices are claimed to have ten times the flavoring strength of the equivalent ground spice.

The disadvantages and advantages of encapsulated spices are listed below (Heath, 1972).

A. Advantages

1. Aromatics are fully protected from volatile loss and degradation.
2. They have a long shelf-life under all conditions.
3. They are readily incorporated into food mixes.

4. They are free from objectionable odors (this applies particularly to garlic and onion).
5. They are hygienically excellent, being free from bacteria, filth, and other impurities.
6. They are free from enzymes.
7. They have a low water content.
8. They do not contribute unwanted specks or color the end product.

B. Disadvantage

The concentration is usually tenfold, so its weighing is difficult.

C. Liquid Extraction Method

A liquid extraction method adaptable for the analysis of both solvent-soluble and water-soluble extracts has been used to determine the concentration of oleoresins of capsicums, black pepper, paprika, and turmeric encapsulated in gum acacia. Oleoresins of turmeric and black pepper proved somewhat troublesome because they contain volatile substances that are driven off during drying. The concentration of these materials can be determined indirectly by analysis of water-soluble components (Maleeny, 1961).

XIII. FAT-DISPERSED SPICES

In this type of product, the standardized spice extractives are dispersed in a fatty carrier, which may be either liquid (for example, a vegetable oil) or solid (for example, hydrogenated oil). Such products are designed for incorporation directly into any other fat component of a product (as, for instance, in pastry and in pies). Generally, they are formulated to be three to four times as strong as the equivalent ground spice. Those made in a liquid vegetable oil base are particularly suitable for spraying onto the surface of freshly baked biscuits, conveying both flavor and glaze to the product. They also enable manufacturers of mayonnaise to incorporate the spice into the oil phase of their product (Heath, 1972).

XIV. SPICE PASTES AND CONCENTRATES

A. Spice Pastes

Doring (1952) patented a process in Germany for the preparation of spice pastes by concentration of hot-water extracts to a stage at which the final paste

has a dry substance of 40 to 50% (including NaCl, 10 to 20%), or sugar can be admixed with it to produce a paste that will be stable on storage.

Black pepper berries are separated by air flotation into several fractions. The fraction having the lowest specific gravity, but containing the highest percentage of oleoresin, is subjected to solvent extraction for further processing into pepper paste.

Tandon and Lal (1960) and Tandon and Siddappa (1963) reported the results of their experiments on mustard flour used in prepared mustard—a paste product containing mustard flour, vinegar, spices, etc. Yamanishi *et al.* (1964) have discussed the development of off-flavor in mustard paste. Miyoshi (1969) conducted studies on flavor retention in processed mustard products.

Lee *et al.* (1970) conducted microbiological studies on the fermentation of red pepper paste with special reference to distribution of yeast during the fermentation period.

B. Spice Concentrates

A process for making tamarind concentrate has been developed at CFTRI, Mysore. The process essentially consists in extraction of all the solubles from the fruit with boiling water, clarification or straining of the extract, and concentration of the clear extract under vacuum to about 65% soluble solids. The concentrate becomes quite viscous, and, although fluid when hot, sets to a jam-like consistency on cooling. It can be easily dispersed in hot water (Lewis *et al.*, 1970). The setting is due to the presence of pectin in the fruit. Since setting is a desirable property (it avoids spilling and leakage of the material during transshipment and storage), it is best to make the product from freshly harvested, firm fruit, early in the season; otherwise pectolytic degradation takes place during storage of the fruit, and then the setting will be poorer. Fresh fruit gives a lighter colored product. Insect infestation is absent during the first few months of storage of the fruit, if it is of good quality and free from seeds. Insect infestation is serious when seeds are present. The presence of insect fragments is highly objectionable in the finished product. Hence it is necessary to use freshly harvested, hygienic tamarind pods. The yield is about 75 to 90% depending on the quality of pulp used.

XV. SPICE SALTS

A. Celery Salt

According to Canadian FDA regulations, celery salt shall be combination of ground celery seed or ground dehydrated celery with common salt.

Allen and McCaleb (1937) obtained a celery salt preparation by subjecting oil of celery and monosodium glutamate to minute subdivisions—that is, in a hammer mill—in order to bring them into intimate physical relationship and then mixing them with sodium chloride in appropriate proportions.

B. Garlic and Onion Salt

According to United States specifications, garlic salt is a properly blended, dry mixture of garlic powder, salt, and calcium stearate. Not less than 98% of the finished product should pass through a U. S. standard No. 30 sieve (0.023-inch openings). The finished product (on the "as-is" basis) should contain not less than 18% nor more than 20% garlic powder, and not more than 2% nor less than 1% calcium stearate; the remainder should constitute common salt. The moisture content should not exceed 2.5%.

Onion salt has precisely the same composition specification. The Canadian specification is slightly different from the U.S. specification in that it shall contain not more than 75% salt and not more than 2% of anticaking agent.

C. Peppersal

Black pepper is graded commercially by the air-flotation technique into several grades or fractions. The fractions having the lowest specific gravity (light pepper, pinheads, brokens, etc.), but containing the highest percentage of oleoresin, are used for the recovery of oleoresin. This oleoresin is then utilized in the preparation of peppersal.

Peppersal is a new type of spice flavoring or spice salt developed at CFTRI, Mysore. It is made by dispensing or absorbing the above oleoresin from pepper rejections in suitable quantities of refined, pulverized, free-flowing common salt (Ramachandra Rao *et al.*, 1959).

XVI. CANNED OR BOTTLED PIMIENTO

There is a demand for canned or bottled pimiento or capsicums *(Capsicum annuum)*. Pimientos (capsicums or chili peppers) are processed in brine in plain tin cans or glass jars. Factors influencing the processing of canned pimientos, rates of heat penetration, and suggested thermal processes have been reported from time to time and reviewed by Hight *et al.* (1954) and by Powers *et al.* (1961), but in the commercial pimiento canning industry, some spoilage was still occasionally reported. Hight *et al.* (1954) conducted studies (1) to determine the thermal death time characteristics of the microorganisms causing spoilage and (2) to establish processing times on the basis of these studies. The 212°F and "Z"

values were found to be 36.0 and 31.0, respectively, at pH 5.0. At pH 4.7, the corresponding values were only 7.0 and 4.2, respectively.

Powers *et al.* (1950) suggested the acidification and calcium firming of canned pimientos. Acidifcation increased the drained weight of the product significantly. The use of both an acidulant and calcium chloride resulted in a highly significant increase in drained weight and in firmness of the product. Sane *et al.* (1950) established the range of variation in pH and total titratable acidity in canned pimientos (pH 4.6 to 5.3, average pH 4.95, average acidity in red 0.28%, average acidity in green 0.6%). Unless the pods are fully colored red, the pH may be as high as 5.5. As the color becomes deeper red, the pH decreases to 4.7. Pimientos of good canning color have a pH between 4.7 and 5.3. Siddappa and Bhatia (1955) reported the satisfactory canning of green chilies.

Powers *et al.* (1961) studied the effect of acid level, calcium salts, monosodium glutamate (MSG), and sugar on canned pimientos. When 0.05% of MSG was added to acid-treated pimientos, the organoleptic acceptability was increased. Addition of sugar improved the palatability of the product. The above conclusions are based on exhaustive studies conducted by the authors during five packing seasons.

XVII. PROCESSING OF TENDER GREEN PEPPER

There has been a growing demand from several western countries for canned or bottled green pepper. In the absence of any published information, Pruthi *et al.* (1974b, 1976a,b, 1978b) conducted systematic studies (1) to standardize the conditions of processing of tender green pepper (canning, bottling, drying and dehydration), (2) to determine the optimum stage of harvesting of green pepper for canning, bottling, and drying, and (3) to formulate quality standards for (*a*) canned green pepper, (*b*) bottled green pepper in brine, and (*c*) green pepper pickle in brine, vinegar, and oil, with special reference to green color, texture, drained weight, etc.

A. Canned Tender Green Pepper

1. Process Evaluation

Studies on process evaluation of canned tender green pepper as berries and as spikes in 2% brine and in plain water have shown that the marginal atmostpheric thermal processes for pepper spikes packed in different sizes of cans were 15 minutes for 5-ounce baby-food cans (202 × 214), and 20 minutes for 1-pound jam cans (301 × 309) and A2½ cans (401 × 411). The marginal thermal processes for pressure processing (10 pounds of steam pressure) were 10, 15, 20, and 20 minutes for 202 × 214, 301 × 309, 401 × 300, and 401 × 411 sanitary

cans for pepper spikes, and 10 and 15 minutes for 202 × 204 and 301 × 309 cans for pepper berries, respectively. These results were further confirmed in the subsequent season (Pruthi *et al.*, 1974b, 1976a,b, 1978b).

Of the thirteen treatments tried for standardization of conditions for canning green pepper in 2% brine, plain water, or distilled water, with or without additives such as EDTA or citric acid, the addition of citric acid at a 0.15% to 0.20% level prevented the bluish-gray discoloration found in the covering liquid and also gave a good, clear covering liquid in both Panniyur and Karimunda varieties of green pepper irrespective of the location of plantation. There was neither gelation nor rupturing of berries nor cloudiness of brine in any of the lots. On the basis of these studies, the following method may be recommended for the canning of tender green pepper.

The pepper berries, after removal from the spikes, are thoroughly cleaned of foreign matter, pinheads, etc., washed thoroughly in running water, and then steeped in chlorinated water containing 20 ppm of chlorine for about one hour. They are then packed in plain 301 × 309 cans, covered with hot 2% brine containing 0.2% citric acid, exhausted to 80°C, sealed promptly, and processed in boiling water for 20 minutes, after which they are immediately cooled in running cold water (Pruthi *et al.*, 1974b, 1976a,b, 1978b).

2. *Determination of the Optimum Stage of Maturity for Canning and Bottling of Green Pepper*

Studies were conducted for the first time to determine the optimum stage of maturity for canning and bottling of green pepper of Panniyur (P) and Karimunda

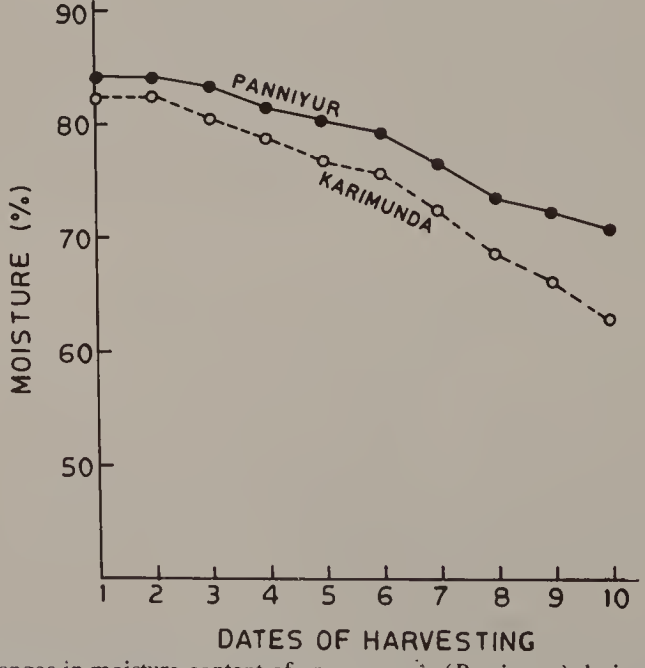


FIG. 19A. Changes in moisture content of green pepper (*P. nigrum*) during development, maturation, and ripening. From Pruthi *et al.* (1976b).

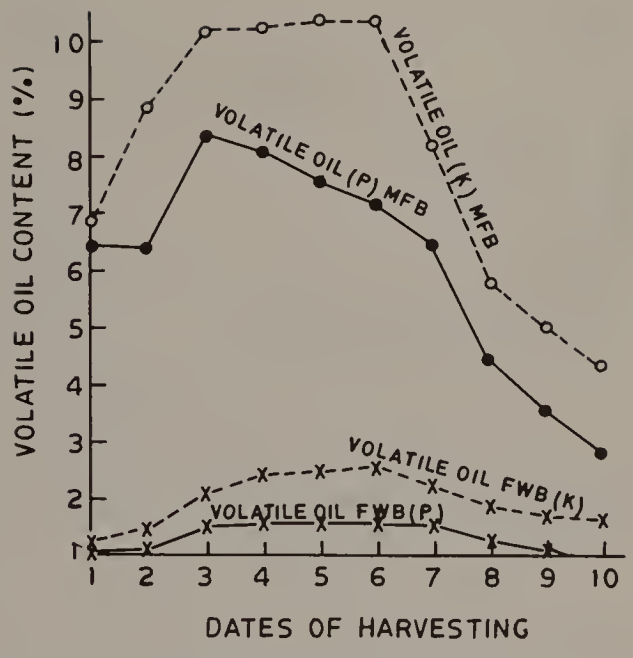


FIG. 19B. Changes in volatile oil content of green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. (MFB, moisture-free basis; FWB, fresh-weight basis.) From Pruthi *et al.* (1976b).

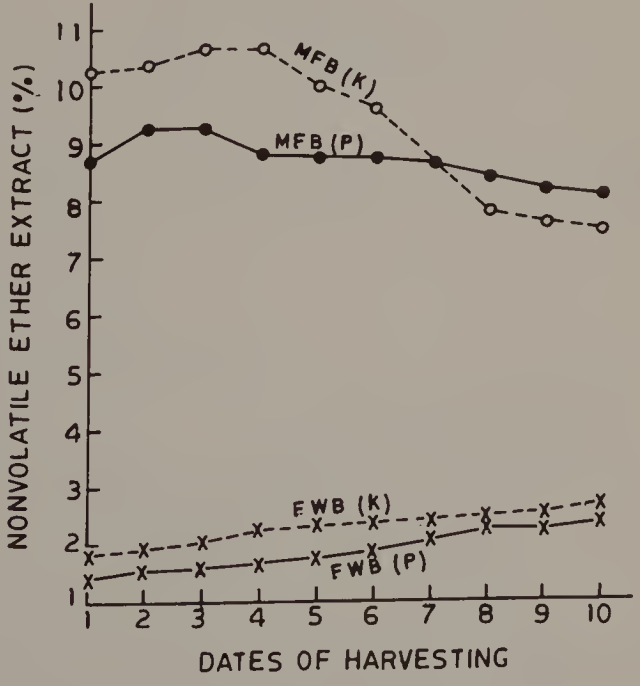


FIG. 19C. Changes in nonvolatile-ether extract in green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. MFB, moisture-free basic; FWB, fresh-weight basic. From Pruthi *et al.* (1976b).

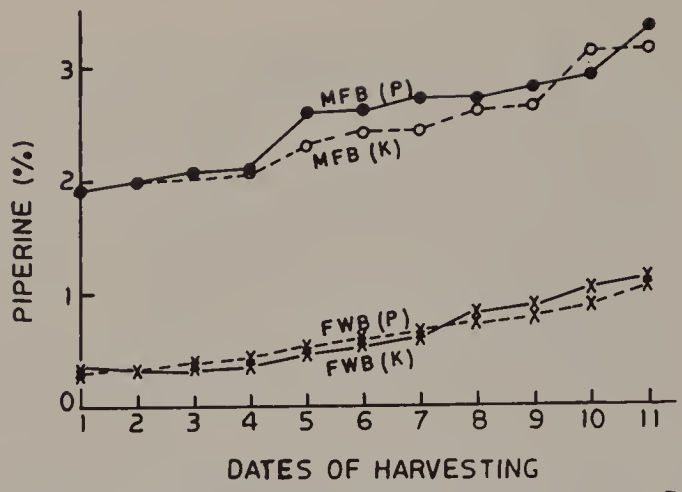


FIG. 19D. Changes in piperine content of green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. MFB, moisture-free basic; FWB, fresh-weight basic. From Pruthi *et al.* (1976b).

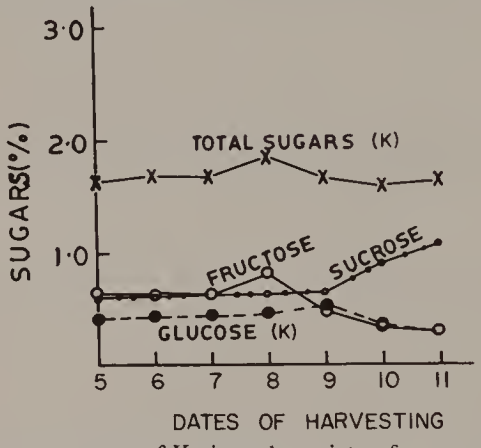


FIG. 19E. Changes in sugar content of Karimunda variety of green pepper (*P. nigrum*) during development, maturation and ripening. From Pruthi *et al.* (1976b).

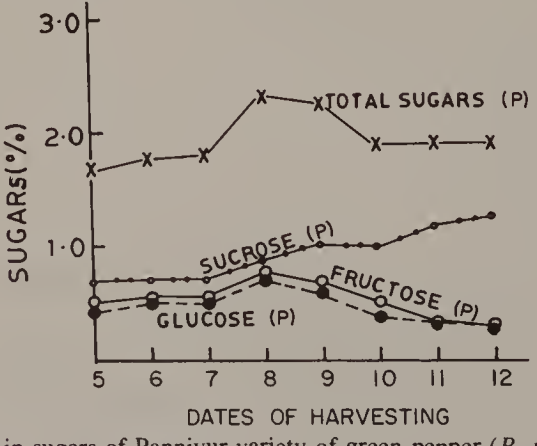


FIG. 19F. Changes in sugars of Panniyur variety of green pepper (*P. nigrum*) during development, maturation, and ripening. From Pruthi *et al.* (1976b).

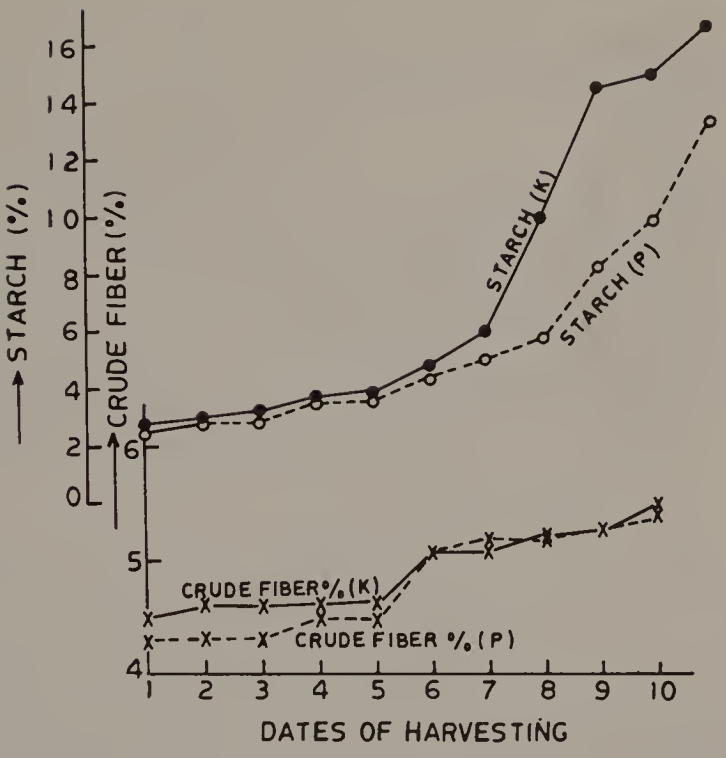


FIG. 19G. Changes in starch and crude fiber content of green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. From Pruthi *et al.* (1976b).

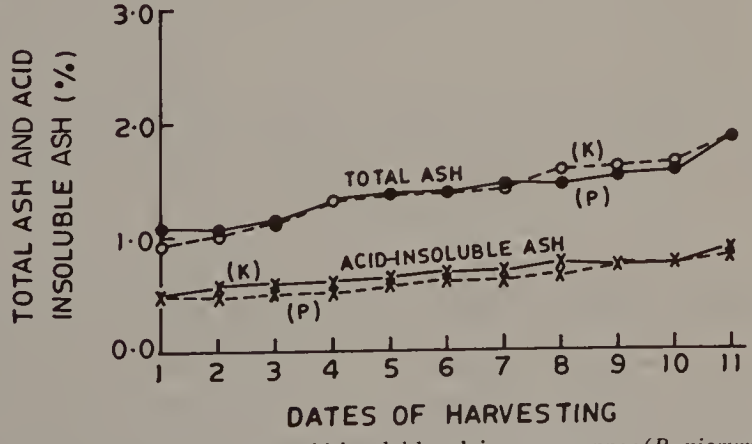


FIG. 19H. Changes in total ash and acid-insoluble ash in green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. From Pruthi *et al.* (1976b).

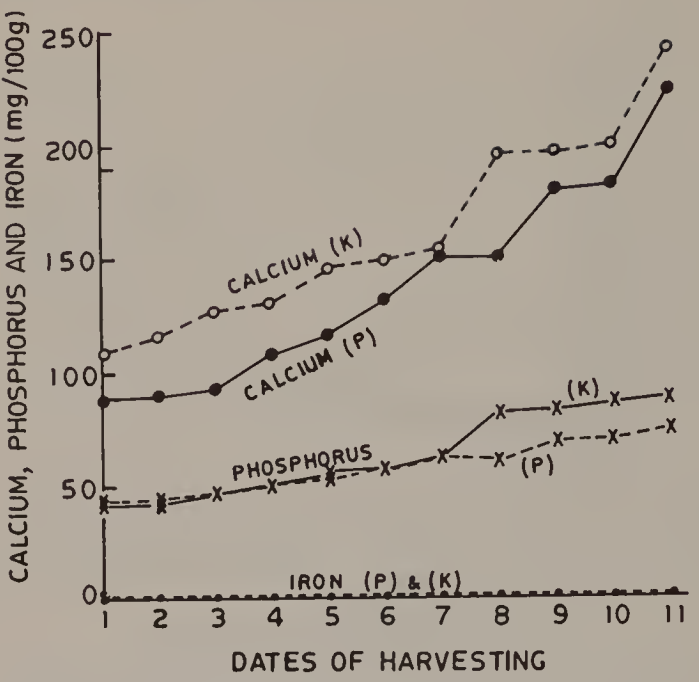


FIG. 191. Changes in calcium, phosphorus, and iron in green pepper (*P. nigrum*) var. Panniyur (P) and Karimunda (K) during development, maturation, and ripening. From Pruthi *et al.* (1976b).

(K) varieties (Pruthi *et al.*, 1974b, 1976b). Spikes of either variety were harvested at ten to twelve stages at regular intervals of 10 to 12 days each during October 1974 to February 1975. Studies on changes in their composition and quality attibutes have shown that both varieties were good for canning up to a certain stage, after which there were marked changes in some important chemical attributes (Figs. 19A–I). The Panniyur variety was good for canning and bottling up to the end of December, and Karimunda up to the middle of December. These were the critical stages with respect to their quality or chemical composition, for there was a sudden increase in starch and crude fiber contents, a decrease in moisture, volatile oil, and nonvolatile ether extract, and a gradual increase in piperine content, which made the berries too pungent, starchy, and hard (Figs. 19A–I). Further, dramatic changes were observed after the end of December in both the varieties of pepper, notably with respect to starch, crude fiber, minerals, volatile oil, nonvolatile ether extract, and sugars (Pruthi *et al.*, 1974b, 1976b).

B. Bottling of Green Pepper in Vinegar, Acetic Acid, or Brine

Studies undertaken to determine the optimum concentration of common salt, acetic acid, citric acid, etc., for the preservation of bottled green pepper (as spikes or berries) have shown that berries and spikes packed in 2% brine containing 2 to 4% acetic acid could be preserved. Airtight capping of bottles is essential

to prevent mold attack. Further studies showed that berries and whole spikes could be preserved in 16% brine containing 1 to 2% acetic acid. Berries or spikes could also be preserved without any difficulty in 2 to 4% acetic acid solution alone, without the use of common salt.

Of the twenty different treatments tried for bottling of green pepper of Panniyur and Karimunda varieties in 20% brine with the addition of different concentrations of citric acid, EDTA, SO_2, etc., addition of 100 ppm SO_2 + 0.2% citric acid in 20% brine is recommended as the best technique. However, the salt content in the final brine must be raised to 16% after 2 to 3 days of packing, as the salt concentration goes down to 12 to 13%, owing to osmosis, etc. (Pruthi *et al.*, 1974b, 1976b). The growing of the covering brine in bottled green pepper and its prevention have been discussed by Pruthi *et al.* (1978b).

1. Storage Studies on Bottled Pepper

Acetic acid was found to be a better preservative than citric acid from the microbiological angle, but color retention was better with citric acid during storage. Berries preserved in only 2% brine and 2% acetic acid had better color than blanched berries packed similarly. Light has a positive role in the darkening of green pepper berries during storage. This can be controlled by packing green pepper in brine and vinegar in PVC (polyvinyl chloride) containers instead of in glass containers as indicated in our studies (Pruthi *et al.*, 1974b, 1976b).

2. Other Green Pepper Products

Different types of mixed pickles (using green pepper as one of the components) prepared in brine, vinegar, and oil remained in good condition for nearly 6 months. Green pepper could advantageously be incorporated in place of black pepper in the preparation of rasam, soups, etc. Canned pepper rasam remained in good condition for 6 months. Based on the analysis of several market samples and laboratory samples of canned and bottled green pepper, quality standards have been proposed (Pruthi *et al.*, 1974b, 1976a,b).

C. Sun Drying and Dehydration of Tender Green Pepper at Different Stages of Maturity

The green pepper berries of Panniyur and Karimunda varieties harvested on ten different dates at an interval of 10 to 12 days during October to December 1974 were sun dried and also dehydrated separately in a cross-flow cabinet dryer after blanching at low temperature for a long period of time. In both varieties, the drying ratio varied from 7.0:1 at the initial stage to 2.0:1 at full maturity (Fig. 20A,B). The rehydration ratio also varied from 1 : 6.8 to 1 : 2.0 (Pruthi *et al.*, 1974b).

Studies conducted on the retention of green color in dried pepper have shown

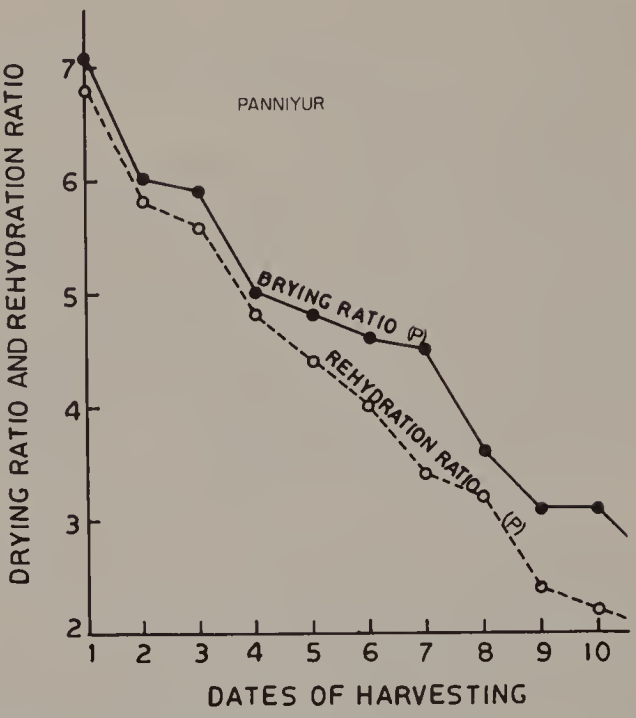


FIG. 20A. Changes in drying ratio and rehydration ratio in green pepper (*P. nigrum*) var. Panniyur, at different stages of maturity (D_1 to D_{10}).

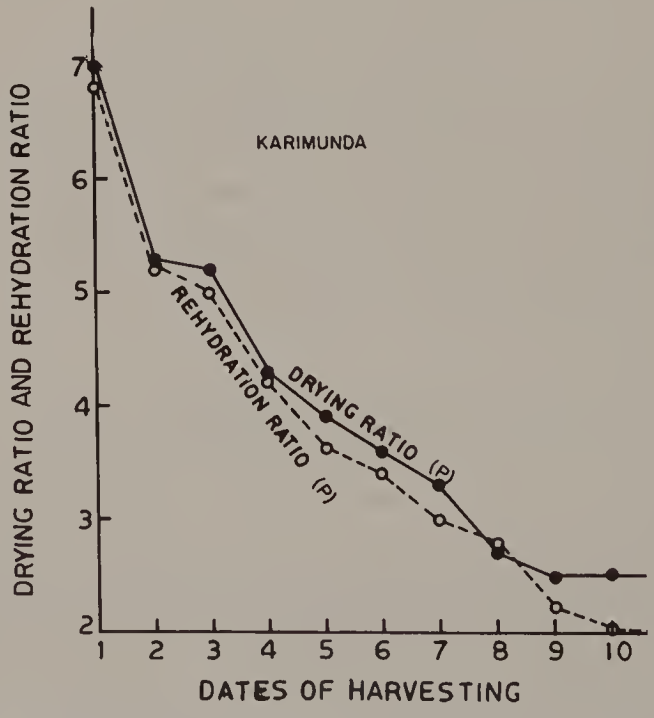


FIG. 20B. Changes in drying ratio and rehydration ratio in green pepper (*P. nigrum*) var. Karimunda at different stages of maturity (D_1 to D_{10}).

that berries steeped in acidified sulfited solution (1% potassium metabisulfite + 0.25% citric acid) gave good products after sun drying and dehydration. Many of the berries were green in color, and only a few were dark. The dehydrated product was slightly better than the corresponding sun-dried lot. Steeping in 8% or 12% hydrogen peroxide overnight bleached the color of the berries and gave a white product on sun drying. Steeping in 12% hydrogen peroxide overnight gave a whiter product than the one steeped in 8% hydrogen peroxide. The pepper berries after steeping for 5 hours in 12% hydrogen peroxide prior to dehydration gave a light-green product (Pruthi *et al.*, 1974b).

D. Physical and Chemical Changes during Maturation and Ripening of Green Pepper

The changes in physicochemical characteristics of green pepper during development and maturation of Panniyur and Karimunda varieties have been recorded for the first time by Pruthi *et al.* (1974b, 1976a,b). Chemical analysis of fully mature green pepper, yellow berries, and red berries of the Panniyur variety was also conducted [Table LII (A)]. There were significant differences in chemical constituents after maturation and during ripening (Pruthi *et al.*, 1974b, 1976b). The differences in the chemical composition of green pinheads, light

TABLE LIIA VARIATIONS IN CHEMICAL COMPOSITION OF GREEN, YELLOW, AND RED BERRIES OF PANNIYUR VARIETY PEPPER (JANUARY 1975)[a,b]

Chemical constituents	Pale greenish berries	Yellow berries	Red berries
Moisture (%)	68.01	59.20	58.98
Volatile oil (%)			
Fresh-weight basis	0.70	0.70	0.70
Moisture-free basis	2.10	1.70	1.70
Nonvolatile ether extract (%)			
Fresh-weight basis	2.59	2.96	2.88
Moisture-weight basis	8.10	7.20	7.00
Sugars			
Glucose (%)	0.11	0.83	0.78
Fructose (%)	0.11	0.63	0.70
Sucrose (%)	0.40	1.34	2.56
Total sugars (%)	0.62	2.82	4.04
Starch (%)	13.30	18.70	20.40
Crude fiber (%)	4.50	5.80	5.80
Acidity (%)	0.09	0.19	0.29
Ascorbic acid (mg/100 g)	3.20	3.60	3.70

[a] From Pruthi *et al.* (1976b).
[b] Unless specified to the contrary, the results are on a fresh-weight basis.

TABLE LIIB DIFFERENCES IN CHEMICAL COMPOSITION OF GREEN PEPPER, LIGHT BERRIES (GREEN), PINHEADS (GREEN), AND GREEN SPIKE STEM OF PEPPER (*P. NIGRUM*) OF A LOCAL INDIAN VARIETY[a]

Chemical constituent	Green pepper (mature)	Green light berries	Green pinheads	Green spike stem
Moisture (%)	68.46	82.42	78.12	74.60
Starch (%)	13.49	6.82	5.35	4.46
Crude fiber (%)	4.70	4.96	4.35	16.00
Nitrogen (%)	0.76	0.74	0.57	0.41
Protein ($N \times 6.28$) (%)	4.73	4.64	3.59	2.54
Alcohol extract (%)	5.24	0.87	1.03	0.80
Volatile oil (%)	1.20	1.30	1.35	Nil
Nonvolatile ether extract (%)	2.44	2.40	2.40	1.20
Total ash (%)	1.80	1.34	1.40	2.53
Ash insoluble in HCl (%)	0.64	0.65	0.54	0.75
Vitamin C (mg/100 g)	3.00	2.50	2.10	Nil
Calcium (mg/100 g)	288.00	388.00	393.00	245.00
Phosphorus (mg/100 g)	93.80	94.40	61.80	80.60
Iron (mg/100 g)	1.00	6.20	4.10	1.50

[a] From Pruthi *et al.* (1974b).

berries, mature green pepper berries, and spike-stem have also been brought out in Table LII (B) (Pruthi *et al.*, 1974b).

Studies were also conducted on the effect of storage of green pepper spikes with the green berries on, for 1 to 4 days, on the percentage of recovery of berries, wastage, time of separation of berries from spikes, moisture, volatile oil, and NVE (nonvolatile ether extract). Time taken for the separation of berries was reduced when the spikes were kept for 2 to 3 days. The recovery of volatile oil

TABLE LIIC EFFECT OF KEEPING GREEN PEPPER WITH SPIKES (VARIETY PANNIYUR) IN SHADE ON MOISTURE, VOLATILE OIL CONTENT, AND NONVOLATILE ETHER EXTRACT[a]

Storage period (days)	Moisture (%)	Volatile oil, fresh-weight basis (% v/w)	Volatile oil, moisture-free basis (% v/w)	Nonvolatile ether extract fresh-weight basis (% w/w)	Nonvolatile ether extract moisture-free basis (% w/w)
0	76.41	1.5	6.3	2.06	8.7
1 (blanched)	76.20	1.3	5.6	1.94	8.2
1 (control)	75.23	1.4	5.7	2.16	8.9
2	74.91	1.3	5.2	2.36	9.4
3	74.47	1.2	4.7	2.61	10.2
4	72.60	1.2	4.4	2.91	10.6

[a] From Pruthi *et al.* (1976b).

was higher when the green pepper was distilled at the fresh stage, and nonvolatile ether extract increased slightly during 2 to 4 days of storage [Table LII (C)]. However, there was some deleterious effect on the quality of volatile oil with respect to color and aroma after 3 to 4 days of keeping. Blanching reduced the recovery of both the volatile and the nonvolatile ether extracts (Pruthi *et al.*, 1976b).

Studies were made on the determination of the optimum time of steam distillation for maximum recovery of volatile oil from green pepper. Ninety percent of the volatile oil was removed in just 30 minutes of distillation. As a safety measure, distillation for an hour is recommended (Pruthi *et al.*, 1974b).

XVIII. OTHER SPICE PRODUCTS

Ansel (1955) developed patented products from black pepper. Pruthi *et al.* (1960a) reported the development of several promising products from ginger, in addition to essential oil and oleoresin, ginger essence, ginger ale, vitaminized powder, and vitaminized, effervescent ginger powder, ginger beverages (ginger cocktail and other soft drinks), ginger preserve and candy, alcoholic beverages, lime-ginger pickles, etc. Pruthi and Srivas (1962) reported an integrated process for the recovery of essential oil, oleoresin, and starch. Spent ginger contained 50 to 55% starch. Natarajan *et al.* (1970) reviewed and discussed product development of ginger including the use of ginger and ginger flavor in a number of products. Lewis *et al.* (1972) indicated the possibility of utilization of the spent, dried ginger meal (left after oleoresin recovery) in animal feed compositions. Srivas *et al.* (1962) determined the optimum time of harvesting of ginger for the production of dried ginger, ginger oil, and oleoresins.

Green peppercorns are basically low-acid fruit; consequently, they are acidified for canning so as to permit atmospheric processing in boiling water. Since there were no available data on acidity and pH of green peppercorn, Pruthi *et al.* (1976a) found the pH of nonacidified canned green pepper to be around 6.25 to 6.50 and acidity within 0.05% as citric acid. Powers and Shinholser (1979) determined the pH, total acidity, and buffering capacity of canned green peppercorns against increases and decreases in pH. The mean pH was 6.51 and acidity 0.048%, calculated as citric acid. The buffering capacity between the original pH of the peppercorns and pH 8.35 was 0.188 meq. To lower the pH to 4.4, 219–247 mg citric acid per 100 g can fill-in-weight of green peppercorns is required. Canned green peppercorns, acidified to pH 4.0, retained much of the pungency and pepperiness of fresh green peppercorns.

7

Utilization of Spice Wastes

The available published information on spice wastes is scanty, covering about one dozen spices, as discussed briefly below.

I. BASIL WASTE

Tharanathan and Anjaneyulu (1974) have studied some aspects of polysaccharides from the seed mucilage of *Ocimum basilicum* Linn. D-Glucose, D-galactose, L-arabinose, D-xylose, and L-rhamnose along with D-galacturonic acids are found to be constituents of the seed musilage of basil. The neutral sugars are in the ratio of 5:5:2:3:3:1.

The mucilage is partly O-acetylated (OAC, 5.08%) and contains lipids—free 25.4%, and bound 10.3%. Its graded hydrolysis results in the isolation of an acid-stable core polysaccharide (43%) composed of D-glucose and D-mannose in the ratio of 10:2. Fractionation by a different technique indicates the mucilage to be highly heterogeneous.

II. CAPSICUM WASTE

Tandon *et al.* (1964) found that spent chili meal contains about 28% protein, 36% carbohydrate, and 29% fiber. It could perhaps go into animal feed compositions.

According to Portales and Honrubia (1967), red pepper (chili) may be extracted with little degradation in an installation characterized by continuous rotary extractors that operate at room temperature and are fitted with indirect heating so that they can be used for drying the meal. Degumming is accomplished with H_3PO_4, citric acid, and sodium tripolyphosphate. The miscella are evaporated under vacuum, leaving an oil with 3 ppm of solvent.

Bough (1973) studied characterization of waste effluents from a commercial pimiento (capsicum) canning operation.

The first stage of processing, in which the roasted peel was removed by washing, accounted for the most concentrated effluent. Another concentrated effluent resulted from the citric acid dip just before the packing and closing area. Two effluents from the grading area were also obtained. The total wastes produced from the canning plant contained 3.2 pounds of suspended solids, 60.2 pounds of chemical oxygen demand, and 35.4 pounds of biological oxygen demand per ton of raw pimientos. Bough and Badenhop (1974) made a comparison of roasting and lye peeling of pimientos for the generation of wastes and quality of canned products. In general, lye peeling gave better results than flame peeling.

III. CASSIA AND CINNAMON WASTE

A. Cassia Leaf Oil

The Regional Research Laboratory (CSIR) in Jorhat, Assam, India, has developed a process for (1) steam distillation of leaves of *Cinnamomum tamala,* which yields about 2% essential oil rich in eugenol content (80 to 85%), and (2) isolation of eugenol from the oil and its conversion to isoeugenol required in the synthesis of vanillin (A.K.S. Baruah, official communication, 1974).

Eugenol can be isolated from cassia or cinnamon leaf oil either by fractional distillation or by extraction with alkali in which it is soluble. Acidification of the alkaline solution gives eugenol in substantially pure condition. It has some use in the perfumery industry, but its major use is in the synthesis of isoeugenol and vanillin.

Isoeugenol can be prepared from eugenol by heating it with alkali. It can also be prepared directly from cinnamon leaf oil by heating the oil under pressure with aqueous alkali and by acidifying the alkaline solution. It has considerable use in perfumery and in the production of vanillin.

Vanillin. Oxidation of isoeugenol with nitrobenzene and alkali yields vanillin. Until recently, vanillin was almost exclusively synthesized from eugenol, but a cheaper method based on the oxidation of lignin has been developed in the

United States, and it is unlikely that vanillin will henceforth be produced from isoeugenol.

B. Cinnamon Leaf Oil

Pruthi *et al.* (1974a,c, 1975, 1978c) have standardized the conditions of the recovery of essential oil from *Cinnamomum zeylanicum* by steam distillation. Mature green leaves give higher yield than fresh, pale-green leaves or fresh, tender, violet leaves. Storage of leaves for 2 to 3 days, followed by steeping in 6% brine for 2 to 3 days, facilitated the higher recovery of oil. The oil is quite aggressive or corrosive. It corrodes ferrous, copper, and to some extent aluminum equipment. Hence, stainless steel or an equivalent metal alloy is essential for the fabrication of the stills. Essential oil recovered in country stills made of iron or copper is dark-brown in color. However, this color can be removed or precipitated by the addition of suitable organic acids. Cinnamon leaf oil contains 80 to 90% eugenol and 1.0 to 2.5% cinnamaldehyde, and is soluble in 2 volumes of 70% alcohol. It is a pale yellow liquid with strong cinnamon aroma. Packaging and storage studies on cinnamon leaf oil are reported. Wijeskera *et al.* (1974), after briefly reviewing the earlier work done on cinnamon leaf oil, have also reported the GLC analysis of leaf, bark, and root oils.

Hohmann (1969) has used cinnamon leaves in curry powder.

C. Cinnamon Bark Oil

Cinnamon bark oil is extracted from inferior-quality bark, broken quills, inferior featherings, chips, etc., which are first reduced to a coarse powder, macerated in a saturated solution of common salt for 2 days, and then distilled. The yield varies according to the quality and age of the bark (Hattiangdi and Nimbalkar, 1958). Wijesekera *et al.* (1974) have reported the GLC analysis of the volatile oil of cinnamon stem bark.

D. Cinnamon Root and Fruit Oil

Cinnamon roots of old or cut trees and cinnamon fruits are also used for the recovery of essential oils that have medicinal and other uses (Hattiangdi and Nimbalkar, 1958). Wijesekera *et al.* (1974) have reported some significant differences in cinnamon leaf, bark, and root bark oils. α-ylangene, methyl and ethyl cinnamate in leaf oil, benzyl benzoate in bark oil, and 4-terpemine-1-ol in root bark are claimed to have been reported for the first time. The main constituents of leaf, bark, and root oils are eugenol, cinnamaldehyde, and camphor, respectively.

IV. GARLIC AND ONION WASTE

Onion peels, cores, tops, and fragments are usually considered waste products and are dumped. These solid materials may be hauled away as garbage or allowed to mix in the water effluent and run into the ground. After the water has evaporated or disappeared into the ground, the onion solids can be plowed under. The available facilities for land disposal or other means for getting rid of the water will depend on plant location and state or local regulations. If possible, the plant should be located where the intensely odorous air discharged from the dehydration plant will not constitute a nuisance.

Onions contain wax as one of the minor constituents, presumably having a role in protecting the soft tissue from enviromental injury and perhaps also in retaining the volatile flavor components. The waxy matter of the pigmented outer skin of onion found in its ether extract was identified as ceryl ceretate (Okajima, 1954). The presence of a waxy component was also observed in the ether extract of deskinned Red Globe onions (Bandyopadhyay *et al.*, 1970); it was found to be composed mainly of saturated C_{20} fatty alcohol and C_{16} fatty acid. However, minor amounts of eventual C_{26} fatty alcohol as well as C_{18}, C_{20}, and probably C_{26} fatty acids are also present. Gholap *et al.* (1974) have reported on the isolation and characterization of onion wax.

Pectic substances from onion (the white and red varieties) and garlic skins were isolated by Alexander and Subbelle (1973) by extraction with ammonium oxalate. White onion and garlic skins were found to contain 11 to 12% pectin, which can be recovered as a by-product in the dehydration industries. Characterization of these pectic substances in terms of jelly grade, molecular weight, degree of esterification, and methoxyl and uridine content was attempted. Pectic substances from white onions were superior to those from red onions in terms of jelly grade. Both types of onion pectin appeared to be of the rapid-set type, whereas garlic skin pectin was of the medium-set variety. Equivalent weight, methoxyl content, and degree of esterification by themselves did not give any clear indication of pectin grade. Intrinsic viscosity values indicated good correlation between jelly grade and molecular weight. The pectic substances from garlic skin differed from those from onion skin in certain respects, most remarkably in their viscosity behavior.

V. SPENT GINGER RESIDUE

Pruthi *et al.* (1960a) reported the utilization of ginger and also of spent ginger (after the recovery of essential oil from ginger scrapings from the skins and of oleoresin from dry ginger) in the manufacture of ginger ale and beverages. Starch

can also be recovered from spent ginger. Pruthi and Srivas (1962) reported an integrated process for the recovery of essential oil, oleoresin, and starch. Spent ginger was found to contain 50 to 55% starch, which compared favorably with other starches. They standardized the conditions for the recovery of starch. According to Lewis *et al.* (1972), the spent, dried ginger meal (after the recovery of oleoresin) contained about 48% starch and 12% protein. It could possibly be incorporated into animal feed compositions.

VI. NUTMEG REJECTIONS

A. Nutmeg Oil

Wormy nutmegs give a much higher yield of oil than do the sound ones (6 to 16%). In wormy nutmegs, most of the fixed oil (present in the endosperm), which tends to retain the volatile oil during distillation, will have been devoured by worms, while the strongly aromatic oil in the inner layer of perisperm remains intact. Commercial nutmeg oil is derived from broken and wormy nutmegs (Guenther, 1948–1952, see Vol. V; Council of Scientific and Industrial Research, Publication Directorate, 1949–1976, see Vol. VI).

B. Nutmeg Butter

Commercial nutmeg butter, a highly aromatic fat, is obtained from undersized, damaged, or worm-eaten kernels, which are unfit for sale as a spice. The material is ground and cooked or steamed before pressing. A yield of 24 to 30% is reported. The fat may be obtained by solvent extraction, but this process is not usually employed (Council of Scientific and Industrial Research, Publication Directorate, 1949–1976, see Vol. VI).

C. Nutmeg Starch

Nutmeg starch resembles legume starches in appearance. Individual grains, when examined under the microscope, show a well-developed, cracked hilum. The grains are irregular in shape and vary in size from 5 to 50 microns. Compound grains with as many as ten components are quite common (Thorpe, 1945–1956).

D. Utilization of Nutmeg Waste (Husk)

Fresh nutmeg husk constitutes about 80 to 82% of the whole nutmeg fruit (moisture 85 to 88%). It is a rich source of carbohydrates, organic acids,

polyphenols, and crude fiber (Pruthi *et al.*, 1978a). The fresh husk is a rich source of pectin (12 to 14%) of good quality (225 to 250 jelly grade). Factors affecting the recovery and quality of pectin have been studied by Pruthi and Satyavati (1979). A number of delicious products of commercial importance such as nutmeg jelly, chutney, jam, candy, preserve, and beverage have been developed by Pruthi *et al.* (1978a).

E. Nutmeg Leaf and Bark Oil

The essential oil distilled from nutmeg leaves (yield 0.41 to 0.62%) is reported to have weed-killing properties. It can also be used for the flavoring of food products, liquors, dental preparations, and confectionery. Steam distillation of dry leaves from East India gave 1.56% of colorless volatile oil. The bark of the tree yields only 0.14% of a volatile oil. A volatile oil is also obtained from the flowers (Council of Scientific and Industrial Research, Publication Directorate, 1949–1976, see Vol. VI).

VII. PEPPER WASTE

A. Pepper Rejections

Pruthi (1968a) has reviewed the utilization of pepper waste. During the grading of pepper, rejections amount to 5 to 6%. Pepper rejections comprise mainly light berries, pinheads, spikes and varagu (unfertilized buds). Among these trade wastes of pepper, pepper pinheads and light berries had the highest piperine content (4.68 to 5.96%), when compared with spikes (0.93%) and varagu (0.48) (Ramachandra Rao *et al.*, 1960). Narayanan *et al.* (1964) conducted studies on the recovery and quality of oleoresin from these pepper rejections and also on packaging and storage. The cold percolation (countercurrent) technique gave better yields of oleoresin than the hot extraction technique.

Peppersal has been developed at CFTRI, Mysore, by absorbing or dispensing the oleoresin from pepper rejections in free-flowing fine common salt.

B. Spent Residue

Preliminary studies on the utilization of the spent residue left after the extraction of oleoresin revealed that, for the economic recovery of good-quality starch, it is essential to get rid of the husk or the shell (Dewin, 1955). The pepper husk contains a very high content of fiber (23 to 26%) and is low in starch content (2.3 to 2.7%), in comparison with whole black or white pepper containing 13 to 48% starch.

Rat feeding trials have been in progress at CFTRI, Mysore, on the replacement of a rice diet at 5 to 10% levels with spent residue. Crude fiber stands in the way. A residue from which at least 50% crude fiber has been removed may yield better results.

C. Pepper Hulls, Shells, or Skins

Pepper shells left after the decortication of black pepper for the preparation of white pepper, or skins left after the preparation of white pepper by other techniques, amount to 29 to 30% of the whole black pepper. They consist moetly of stone cell fiber, and so far no economic use has been made of them, except for mixing them with other pepper for the recovery of oleoresins or oil.

VIII. PIMENTO WASTE

A. Pimento Leaf Oil

Large quantities of leaves accumulate during the process of harvesting of pimento berries annually. Essential oil can be extracted economically from the leaves, which can provide an added source of income to pimento growers (Rodriguez, 1969).

B. Pigment Source for Poultry Feed

Lavington *et al.* (1974) have conducted systematic studies on the utilization of pimento waste in providing a pigment source for poultry feed. Fresh pimento pulp waste was dehydrated in a pilot Arnold dehydrator to give meals rich in the red xanthophylls, primarily capsanthin. The xanthophyll content of the dehydrated meal ranged from 896 to 1114 mg/kg. Dejuicing of the pimento pulp prior to dehydration decreased charring during drying as well as the quantity of water to be evaporated. Carotenoid losses during dehydration ranged from 17 to 22% for xanthophyll and from 0 to 10% for carotene. Although the unprotected pimento carotenoids were rapidly lost during storage either alone or mixed with dehydrated lucerne, addition of the antioxidant ethoxyquin proved to be very effective in preventing losses of carotenoid during storage. Levels of 1% pimento meal should be adequate in poultry diets when fed in combination with lucerne meal.

IX. TAMARIND WASTE

Tamarind seeds constitute about 35% of the whole tamarind fruit. Savur (1955) perfected a process for the manufacture of polyose, which could be used as a substitute for pectin in the manufacture of jams, jellies, and marmalades.

The only disadvantage is probably that the polyose jelly is a little tougher than fruit pectin jelly. Softer jellies could be obtained by subjecting the polyose to a special patented technique (Pithawala and Sreenivasan, 1950). The various ways of utilizing the polyose from tamarind seeds have been discussed elsewhere (Rao, 1948, 1949, 1959).

The chief use of tamarind seed is in the manufacture of sizing powders. It is widely used in sizing jute and some cotton yarns. It is only half as costly as starch. Almost 30,000 tons of the powder are used annually in the jute industry alone in India (Rao, 1959). Tamarind kernel powder is also used as a creaming agent for rubber latex, as a soil stabilizer, and as a pectin substitute as reported above. Tamarind seed polyose can stand longer storage and higher storage temperatures than pectin. During a year's storage it hardly loses 2% of its gel strength (Savur, 1955). The chemical composition and the nutritional, pharmaceutical, and industrial uses of tamarind and tamarind seed have been reviewed in some detail (Rao, 1959; Lewis and Neelakantan, 1969).

According to the latest statistics compiled by the Processed Food Export Promotion Council, over 3.18 million kg of tamarind seed powder valued at 4.27 million rupees were exported from India to the United States, Italy, France, Canada, and twelve other countries during 1973–1974.

X. OTHER SPICE BY-PRODUCTS

Bhargava and Haskar (1962b) reviewed the earlier work done on essential oil from ajowan seeds and its by-products. Without the proper utilization of its by-products, it was not possible for natural thymol to compete with the synthetic product. The following by-products have been suggested: (1) Seed meal (left after the recovery of essential oil) containing 25.32% fat and 15.17% protein can be used either as a cattle feed or as manure. (2) The thymene fraction of the essential oil left after removal of phenols can be used as a low-grade soap perfume. (3) The aqueous distillate left after separation of essential oil and known as omum water is utilized in India for medical purposes. Marayana *et al.* (1967) have reported the recovery of fatty oil from the spent residue of ajowan.

8

Packaging of Spices and Spice Products

I. IMPORTANCE OF PACKAGING

Spices and spice products are hygroscopic in nature, and, being highly sensitive to moisture, their absorption of moisture may result in caking, discoloration, hydrolytic rancidity, mold growth, and insect attack. Furthermore, since spices contain volatile aromatic pirnciples, the loss of these principles and the absorption of foreign odors as a result of inefficient packaging may pose serious problems, especially in ground spices. In addition, heat and light accelerate deterioration, especially with oxygen-sensitive products. With reference to packaging, spices may be classified into four categories.

II. CLASSICATION OF SPICES WITH REFERENCE TO PACKAGING

A. Spices that Need Protection against Light

Spices containing natural coloring pigments, such as green cardamoms, capsicums, paprika, and chilies, need protection from light. The color of capsicums is mostly due to carotenoids, which are susceptible to oxidative deterioration in the presence of light. Spices containing chlorophyll, such as green cardamoms, also fade during storage if not protected against light and air. Similarly, turmeric, which contains curcumin pigment, also fades, although less rapidly than the spices containing carotenoids or chlorophyll. Saffron also is photosensitive.

B. Spices that Need Special Protection from Loss or Gain of Flavor

The loss of flavor depends on the amount and the degree of volatility of the essential oil of the spice. For example, onion and garlic powders contain highly volatile sulfur compounds and therefore need rigorous protection.

C. Spices that Need Protection against Ingress of Moisture and Oxygen

Almost all spices are generally marketed in dry condition, and most of them are hygroscopic in nature. Therefore, they need protection against the ingress of moisture. Besides, the essential oil components naturally present in most of the spices are subject to oxidation by atmospheric oxygen, particularly at high storage temperature, resulting in the development of off-flavors. The ingress of moisture also encourages mold attack and insect infestation in spices. However, most of the whole spices are protected by nature by a pericarp, and the natural antioxidants present therein, and therefore need less rigorous protection than do the ground spices.

D. Ground Spices

Ground spices, because of the greater surface area exposed, lose flavor faster as a result of the loss of volatile oil, and they absorb or lose moisture faster, depending on the atmospheric humidity surrounding them and the storage temperature. Thus they deteriorate much more rapidly than do whole spices. They are more susceptible to mold and insect attack. Therefore, ground spices need special attention in packaging to protect them against such rapid deterioration.

III. DETERMINATION OF PACKAGING REQUIREMENTS

For the systematic determination of packaging requirements and storage characteristics of foods in general, and of dehydrated or dried products like spices in particular, the measurement of equilibrium relative humidity (ERH) is of considerable importance. Hygroscopicity is one of the most important characteristics of dehydrated or dried foods; it is influenced mainly by the moisture content of the product itself and the humidity in the atmosphere over it. The relationship of hygroscopicity and hygroemissivity to the moisture content is usually expressed in the form of a humidity–moisture equilibrium curve or sorption isotherm, which shows the quilibrium relationship between the moisture content of the product and the relative humidity of the atmosphere immediately over it. Products having ERH less than 50% are usually considered hygroscopic, whereas those with ERH above 50% are hygroemissive.

Several methods have been reported for the determination of ERH, including the following:

1. Measurement of water vapor pressure by a manometric technique (Adam and Merz, 1919).
2. Measurement of ERH directly with an electric hygrometer Karel *et al.*, 1955; Landrock, 1957: Mossel and Van Kwik, 1955).
3. The mathematical formula method (Karel *et al.*, 1955; Henderson, 1952).

TABLE LIII INITIAL MOISTURE, EQUILIBRIUM RELATIVE HUMIDITY, CRITICAL MOISTURE, AND CRITICAL RELATIVE HUMIDITY AT WHICH MOLD ATTACKS WHOLE AND GROUND SPICES AT ROOM TEMPERATURE (25°–28°C)[a]

Spice	Initial moisture (%)	Equilibrium relative humidity (%)	Mold growth at moisture level (%)	Mold growth at relative humidity (%)
Cardamom, greater (brown) (amomum)	7.78	45	14.16–22.19	73 and above
Cardamom (green)	10.46	40	16.90–22.78	73 and above
Black pepper (whole)	8.19	28	17.74–23.34	73 and above
Black pepper (powder)	6.48	29	13.92–16.82	73 and above
White pepper (whole)	5.09	20	14.47–17.10	81 and above
White pepper (powder)	6.64	18	15.11–18.79	81 and above
Red chili (whole)	4.63	10	16.50–24.67	81 and above
Red chili (powder)	7.48	8	21.28–28.39	81 and above
Ginger (powder)	7.50	20	13.80–20.00	73 and above
Ginger powder (sweetened)	6.50	11	11.00 and above	73 and above
Curry powder	6.71	28	16.43–22.41	81 and above
Mace (whole)	4.33	43	9.74–13.34	81 and above
Fennel seed (whole)	8.16	50	24.33	91 and above
Aniseed (whole)	8.00	62	21.11	91 and above
Celery seed (whole)	8.96	53	25.09	91 and above
Fenugreek seed (whole)	7.73	40	23.59	91 and above
Coriander seed (whole)	6.68	43	13.57–19.19	81 and above
Coriander (powder)	7.18	22	21.81	91 and above
Cinnamon (whole)	3.24	4	16.67	91 and above
Clove (whole)	6.42	30	22.19	91 and above
Garlic (powder)	6.00	13	No mold	No mold
Garlic salt	2.46	5	No mold	No mold
Mustard flour (Coleman)	6.48	6	15.90–23.30	81 and above
Turmeric powder	8.0	22	12.43	91 and above
Onion powder	4.0	5	62.46	90 and above

[a] From Pruthi *et al.* (1962).

4. Wink's weight equilibrium method (Wink, 1946, 1947; Landrock and Proctor, 1951a).
5. The graphical interpolation method (Landrock and Proctor, 1951b), which requires exposure of the product to different relative humidities, as obtained by the Carr and Harris (1949) method, for only one hour.

The first two methods, although rapid and quite accurate, require expensive equipment. The formula method is also an empirical one, and its usefulness and applicability have not been well substantiated (Karal *et al.*, 1955). Methods 4 and 5 are most commonly used for the determination of packaging requirements, or packaging characteristics (Pruthi *et al.*, 1959b).

Pruthi *et al.* (1962) systematically studied the chemical composition and packaging requirements of a number of spices—aniseed, green cardamom, greater

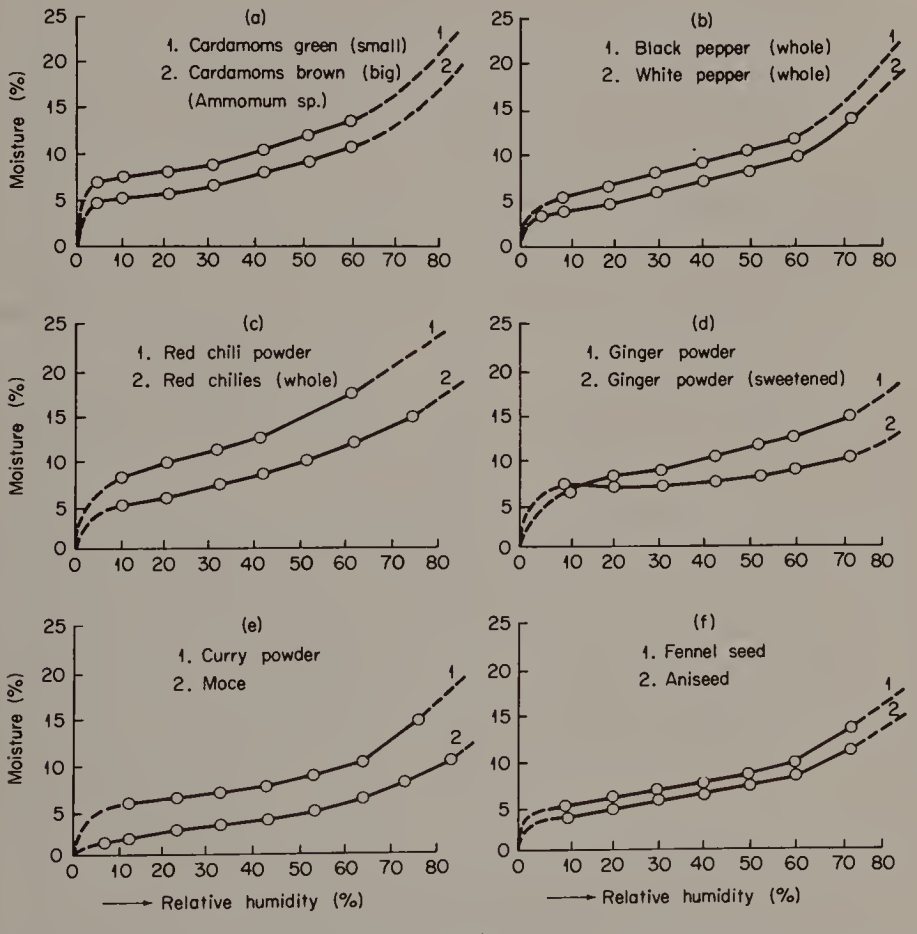


FIG. 21A. Humidity-moisture-equilibrium curves (sorption isotherms) for important spices and condiments at room temperature (25°–28°C). From Pruthi (1970d).

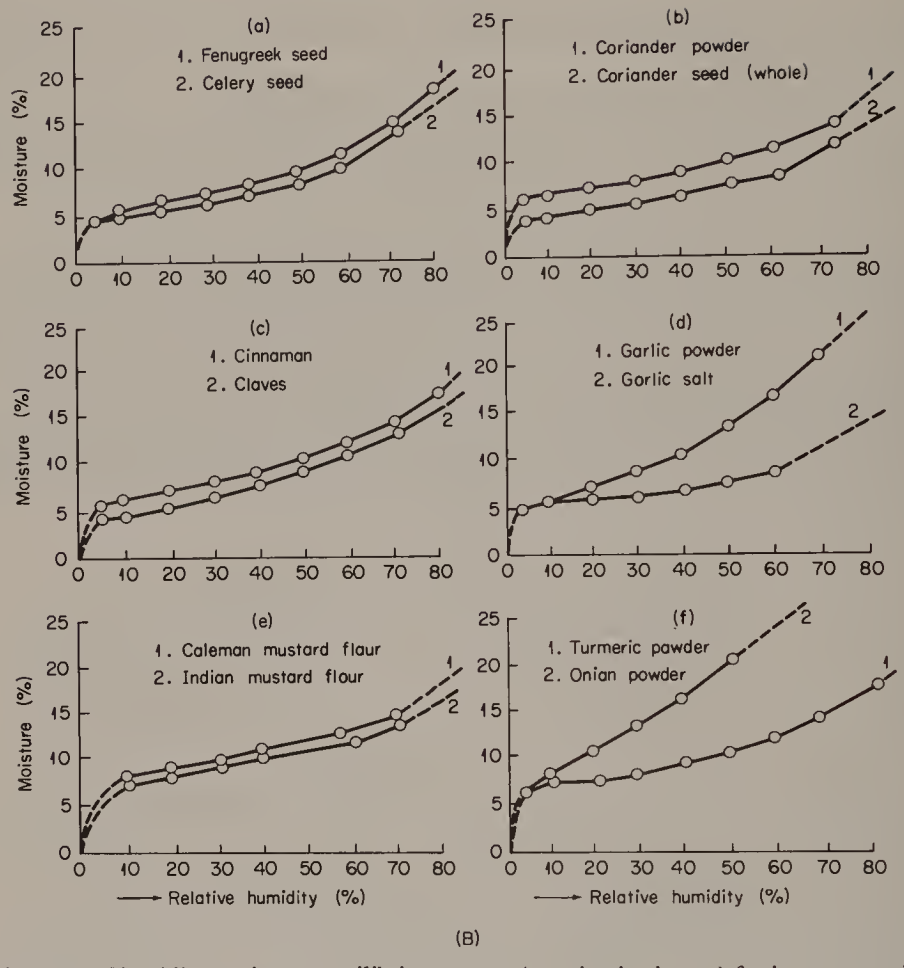


FIG. 21B. Humidity–moisture–equilibrium curves (sorption isotherms) for important spices and condiments at room temperature (25°–28°C). From Pruthi (1970d).

cardamom (brown), coriander seed, cinnamon, curry powder, cloves, garlic powder, garlic salt, ginger, mustard powder (imported and Indian), onion powder, onion salt, mace, pepper (black and white), red chilies, and turmeric (whole and powder). Data based on Wink's weight equilibrium method, and pertaining to sorption isotherms or equilibrium relative humidity (ERH), equilibrium moisture content (EMC), and the moisture content at which mold attack was visible, are presented in Tabel LIII. These studies revealed that, of the several spices and condiments studied, garlic powder, garlic salt, onion powder, mustard powder, cinnamon, and chili powder were comparatively much more hygroscopic than the other spices, and their ERH ranged from 5 to 10% only (Pruthi *et al.*, 1962). The sorption isotherms of these spices can be seen in Figs. 21A and B.

Detailed studies on the ERH of garlic tablets (Pruthi *et al.*, 1959e) and onion powder (Pruthi and Lal, 1960) have also been reported. Garlic powder was much more hygroscopic than the chocolate- or sugar-coated tablets prepared from it (Pruthi *et al.*, 1959e).

IV. SELECTION OF PACKAGING MATERIALS

Packaging of foods is usually utilitarian and protective. The primary purpose of a food packer is to preserve the flavor and keep the product in good condition until it reaches the consumer. The journey from prime producer to ultimate consumer is often long, sometimes halfway around the world. The waiting at the distribution way stations vary from days to months and sometimes is of unpredictable duration. Yet the product must reach the consumer in usable condition, with its fresh flavor and its attractive appearance unimpaired. Throughout the journey, the package must protect the product from thermal changes, humidity variations, hazards of rough handling, rodents, and insect infestation. An ideal package meeting all these requirements is extremely rare (Browne, 1951).

A. Objective Assessment of Packaging Materials

A large number of factors must be considered in detail when one is choosing a suitable packaging material for flavor foodstuffs. These factors may be grouped into two categories: basic factors and consumer acceptance factors.

Basic factors include (1) price to packager; (2) sanitary qualities (protection of product from contamination); (3) resistance to impact injury; (4) effectiveness of interior surface; (5) absence of handling problems; (6) space and other storage requirements in the filling plant and in distribution, including weight of package; and (7) special featurres relating to performance for packager.

Consumer acceptance factors include (1) size; (2) ease of opening (3) reseal features; (4) pouring qualities; (5) space saving on consumer's premises; (6) protection from light; (7) transparency; (8) tamperproof construction; (9) protection of contents from physical or chemical changes; (10) physical characteristics of outside surface, including appearance; (11) ease of disposal; and (12) special features relating to performance for consumer.

To summarize, the package should fulfill two important functions: It must sell its contents as well as protect them. The sale function includes attractive advertising potential, protection from mechanical damage, and reasonable cost. The protective function includes compatibility with the product, and protection against climatic conditions, microorganisms, insects, and filth, and flavor gain or loss. Finally, the package should fit in the production line to facilitate automation.

B. Types of Packaging Materials

The various materials suitable for packaging of foods include paper products, polyethylene flexible films, aluminum foils, glass, tin, hessian, and timber. The selection of packaging material intrinsically will depend on the nature of the

product and other considerations. Only the important ones are briefly discussed below.

1. *Paper and Cardboard Cartons.* Cartons are the least expensive unit packages for whole spices. They have good advertising potential and can be folded into any shape. Wax coating on the outside improves attractiveness as well as resistance to water (wet strength). Polyethylene coating inside gives extra protection as well as saleability. Paper and cardboard cartons are, however, unsuitable for ground spices, owing to their high permeability to flavor components and gases. This disadvantage can be overcome by an inner pouch of suitable polyethylene (Phillip, 1969).
2. *Aluminum foil.* Foil offers excellent potential for packaging ground spices. Aluminum is not transparent and is ideal for spices that need protection from light. Its high resistance to gas transmission is essential to protect the delicate flavor of many spices. It is subject to puncture, but this can be overcome by laminating the outside with paper. Heat sealability can be achieved by coating the inside with a heat-sealable film such as polyethylene (Phillip, 1969).
3. *Combination of Films.* Since a single film does not fulfill all the functional requirements, a combination of films can be used to obtain the desired effect. This can be done by lamination, coating, or co-extrusion. The properties of various films arranged in increasing order of their cost as prevalent in the United States are summarized in Table LIV.

TABLE LIV PROPERTIES OF PACKAGING FILMS[a]

Packaging materials[b]	Barrier properties[c]	Strength[d]	Functional properties[e]
Polyethylene	Poor	Poor	Good
Polypropylene	Poor	Good	Good
Aluminum foil	Superior	Superior	Good[f]
Polystyrene	Poor	Superior	Good[g]
Polyvinyl chloride	Poor	Poor	Good
Rubber hydrochloride	Good	Good	Good
Polyvinylidine chloride	Superior	Superior	Good
Nylon	Good	Superior	Good
Polyester	Superior	Superior	Good[g]

[a] From Phillip (1969).
[b] Packaging materials listed in order of their increasing costs.
[c] Barrier properties: resistance to water vapor, gases, and essential oil.
[d] Strength: rigidity and durability.
[e] Functional properties: clarity, machinability, heat sealability, and printability.
[f] Not heat-sealable and not transparent.
[g] Not heat-sealable.

4. *Tin and Wooden Containers.* Some of the costly spices possessing a delicate, thermolabile aroma, such as saffron, are packed in butter paper and further packed in suitable tin containers. Cured vanilla, cardamom seed, etc., are packed in wooden boxes suitably lined and made as airtight as possible.

V. PRESENT STATUS OF PACKAGING OF SPICES

The various types of containers used at present in different parts of India and other spice-growing countries for different spices, their sizes, capacity, tare weight, cost, etc., are described in a series of Marketing Reports published by the Directorate of Marketing and Inspection (1957, 1965a,b, 1966a,b, 1968a,b, 1970a,b, 1971); these reports have been reviewed by Pruthi (1970d).

VI. STUDIES ON PACKAGING OF SPICES

The available published information on different aspects of packaging of individual spices is briefly reviewed here.

A. Black Pepper

Among the several spices grown in India and exported to various countries, black pepper (whole) is the most important. Certain importing countries have shown keen interest in pepper (ground), preferably packaged in convenient consumer units. Therefore, systematic studies (Pruthi *et al.*, 1962; Narayanaswany *et al.*, 1962) were conducted on the packaging characteristics of whole and ground pepper. Garbled pepper (whole), conditioned to a moisture level of about 9%, was ground to about 60 mesh, and 20-g samples were packaged in pouches made of low- and high-density polyethylene, moisture-proof cellulose film (MST), polycell (a laminate of polyethylene and cellophane) and laminated, heat-sealable (polyethylene-coated) aluminum foil. Storage studies were carried out at 75°F and 85% R.H., as well as under accelerated conditiors—100°F and 90% R.H. The samples were periodically examined for their physical and organoleptic qualities. The results indicated that laminated aluminum foil gave a shelf-life of over 6 months even under accelerated climatic conditions. Polycell and MST cellulose film appeared to be suitable for the same period under normal conditions of storage, whereas polyethylene (both high and low density) did not give adequate protection against flavor losses under both conditions of storage.

White (1957) reported studies on the packaging, storage, and shipment of whole black pepper grown in the Orient. His tests showed conclusively that

whole black pepper, when properly cleaned and dried to less than 11% moisture, can be stored without growth of surface molds in double burlap bags with sealed polyethylene liners, 0.0003 inch or more thick. The exploratory laboratory tests on insect penetration were not conclusive, but when bags were handled carefully and the liners kept unpunctured, the risk appeared to be slight.

B. Cardamom

Cardamom is usually packaged in wooden chests lined with tinfoil or paper, weighing 1 cwt each, but even the artificially dried product packaged in these chests rapidly loses its attractive green color during transport and storage. Consequently, the product fetches a lower price than it would otherwise. As green cardamoms that keep their color during packaging and transport are valued in the export market, the effect of exposing green, dry cardamom to atmospheres of various humidities, and the effect of ultraviolet light on them, packaged in various types of materials and stored for 4 months, were systematically studied (Viraktamath *et al.*, 1964). The results revealed that cardamom with an initial moisture content below 10% could keep well during transport and storage if packaged in 300-gauge black polyethylene-lined packages stored in wooden chests.

In regard to the effect of ultraviolet light on the reversion of cardamom color, it was observed that samples kept in polyethylene bags other than the black showed gradual deterioration in the blue and yellow regions, and the deterioration was high, particularly in the blue region, after 4 months of storage. Samples kept in black bags, however, showed negligible reversion in color (only blue diminishing to 0.9 unit) even after 4 months of storage. It goes without saying, therefore, that when the samples kept in black film bags are packaged in wooden chests, they are sure to withstand color reversion even better.

C. Chilies, Paprika, and Bell Peppers

As a rough estimate, in India not less than 5% w/w of the production of chilies is lost owing to defective storage and spoilage by microorganisms, insects, mites, rodents, and birds, besides the loss of viability and other biochemical changes, resulting in a loss in nutritive quality. Grading, proper packaging, and storage are stressed. Wooden crate dunnage with layer of matting is ideal for bulk storage of chilies (Thomas, 1968).

Jute fabric in the shape of a borem or a bag, holding about 90 to 110 kg and 30 kg of chilies, respectively, is the usual packaging pattern for both export and internal trade.Owing to the low bulk density of this material, this package is very voluminous. Since the freight is charged on a volume basis, the transport charges are often quite excessive. Viraktamath (1964), therefore, conducted systematic

studies to develop a suitable package having reduced bulk for Madras Sanam variety of chilies (which is mainly exported from India) for bulk packaging. The product was equilibrated to a 10% moisture level, and the impact and compression characteristics of the chilies were studied. The optimum pressure found was 2.5 kg/cm^2, which was later adopted for further bulk packaging studies, since impact as well as compression studies indicated that the Sanam chilies tend to break if at 8% moisture and below. The humidity–moisture relationship showed that, at a moisture level of 15% and above, corresponding to a relative humidity of 73%, mold growth occurs. Bulk packages weighing 10 and 25 kg each were made by using detachable wooden frames of appropriate size. For bulk packages, moisture-barrier materials such as polyethylene (300-gauge) would provide protection against moisture, insects, and contamination from external sources. Chilies could be conditioned to about 10% moisture level by exposing the chilies to 70% R.H. for 1 day, and the desired compression of 2.5 kg/cm^2 could then be achieved by the use of any appropriate baling process (Central Food Technological Research Institute, Mysore, 1964).

According to an American report (David and Luh, 1967), red bell peppers (*Capsicum annuum* var. California Wonder) were cored, cut into 1.0- to 1.5-inch squares, frozen at -15°F, and dehydrated in a Stokes freeze-dryer under vacuum to 2.8% moisture content. They were packed in Mylar–Saran–polyethylene plastic laminate and aluminum-film combination (AFC) pouches under nitrogen and stored up to 12 months in the dark at 32°F, 68°F, and 86°F. In plastic laminate pouches, moisture content increased in 12 months from 2.8 to 9.7% at 32°F, and to 10.8% at 68°F. The AFC pouch was effective in protecting the peppers from moisture, oxygen, and light. Storage changes in the product were measured by ascorbic acid retention, formation of water-soluble pigments, and decrease in organoleptic scores. Cysteine, lysine, and γ-aminobutyric acids were less stable than other amino acids at 86°F. Loss of cysteine was accompanied by the formation of cysteic acid and taurine. The loss of carotenoids contributed to the loss of the product's attractive red color, which was accelerated at higher storage temperatures. The effects of storage temperature, moisture content, and packaging material on the stability of bell peppers were discussed by David and Luh (1967).

Ground chilies are known to lose both color and pungency (which is due to the presence of capsaicin) much faster than whole chilies, and they need more efficient packaging for protection against light and air. Packaging of ground chilies in AFC pouches or other efficient packaging materials needs further investigation (David and Luh, 1967). Singh and Ojah (1974) have reported the equilibrium moisture content of chilies.

D. Garlic Powder

On the basis of sorption isotherms (Pruthi *et al.*, 1959h, 1962), the packaging characteristics of garlic powder have been determined. It has been shown that (1)

garlic powder is a highly hygroscopic product, picking up moistire even at 20% R.H. (2) For a typical garlic powder (moisture 6%), the equilibrium relative humidity of 25°C is about 13%. (3) The critical point for garlic powder in regard to caking was found to be at the 10.6% moisture level, and the danger point at 9.5%. Garlic powder was found to be much more hygroscopic than chocolate-coated or sugar-coated garlic tablets. Garlic powder picked up moisture even at 20% R.H., whereas in either type of garlic tablets there was no significant increase in moisture content up to 50% R.H., after which the moisture uptake was fairly rapid. It was easier to store garlic powder in the form of chocolate- or sugar-coated tablets, than as garlic powder (Pruthi *et al.*, 1959e).

Singh *et al.* (1959b) investigated the effect of type of packaging and storage temperature on allyl sulfide, total sulfur, antibacterial activity, and volatile reducing substances in garlic powder. When the different containers stored at 0° and 2°C and room temperature were compared, tin cans appeared to be the best and polyethylene the worst in all the above respects. There was no significant differences among the other containers studied.

The same authors also reported on the packing and storage of whole garlic bulbs in different containers and observed no significant differences in bulbs stored in jute bags or baskets at all the three storage temperatures employed during this investigation: 0° to 2°C, room temperature (24° to 30°C), and 37°C.

Singh *et al.* (1959c) also studied the effect of type of packaging and storage temperature on the flavor and color retention of garlic powder. In general, flavor and color deterioration was maximum at 37°C and minimum at 0° to 2°C. When the different containers were compared at 0° to 2°C and at room temperature, tin cans appeared to be the best containers and polyethylene (250-gauge) the worst. At 37°C, however, there was less browning in garlic powder packed in polyethylene bags than in powder packed in other containers, but again the flavor loss was highest in the polyethylene bags. Furthermore, insect infestation was noticed only in polyethylene bags stored at room temperature. No insect infestation was noticed at the other storage temperatures.

Finally, packaging and storage of garlic powder of low moisture content (6% less) in hermetically sealed cans or in brown or white airtight bottles has been suggested. Polyethylene bags (250-gauge) and gelatin capsules are not suited for the packaging and storage of garlic powder, particularly under tropical or humid conditions (Singh *et al.*, 1959b,c).

Pruthi *et al.* (1959f) have reported on the effect of nitrogen packing and storage temperature on the quality of garlic powder packed in white and brown bottles and tin containers. The losses in allyl sulfide and antibacterial activity during 6 weeks of storage were slightly less in nitrogen-packed sets than in the controls. The losses in allyl sulfide in white bottles were slightly higher than those in brown bottles and tin cans. The losses in allyl sulfide were the lowest at 0° to 2°C and the highest at 37°C. Antibacterial activity, irrespective of the type

of container used, however, deteriorated in both nitrogen-packed and control samples at 37°C. At room temperature and at 0° to 2°C, the antibacterial activity of garlic powder ranged from 3 to 13 mm. With respect to organoleptic evaluation, in general, no significant difference was noticeable between nitrogen-packed and control samples after 6 weeks of storage.

E. Onion Powder

Onion powder was found by Pruthi and Lal (1960) to be highly hygroscopic, picking up moisture even at 10% relative humidity, and being even more hygroscopic than garlic powder. Furthermore, unlike garlic powder, mold attack was noticeable in onion powder stored at 90% R.H. On the basis of sorption isotherms, for a typical onion powder (about 4% moisture) the equilibrium relative humidity would be somewhat less than 5%. To keep the powder free-flowing, it is, therefore, essential that handling (detraying, milling, and packaging) occur in a room of low humidity (about 5%), which will avoid any material uptake of moisture during handling. With respect to caking, the critical point for onion powder would be at a 8.39% moisture level. For a typical free-flowing onion powder, the optimum moisture level would be about 4%.

Goerling (1962) conducted studies at different temperatuers with samples of dried onions and white leek with different moisture content in air and nitrogen packing and at low oxygen partial pressure. White leek and onions were found to be sensitive to storage temperature. Storage at low oxygen partial pressure preserved the color, taste, and keeping time for similar water content for one year, but for onions, the influence of nitrogen packing was slight.

F. Other Spices

Pruthi *et al.* (1962) determined the packaging requirements of twenty spices and condiments (including black pepper, cardamom, coriander, cinnamon, cloves, garlic salt, ginger, onion salt, mustard, mace, and turmeric) and discussed their ERH, their critical moisture content, and its effect on color, texture, and mold attack, etc.

Terricelli (1937) studied the loss of essential oil in the following spices kept in ordinary paper packages: anise, cardamom, coriander, fennel, cumin, sweet marjoram, mace, cloves, pepper, pimento, and cinnamon. Spices kept in small paper bags (containing 1 to 5g) in the dark for five years lost 47% (average) of essential oil; there was more loss in powder spices (up to 90%, average 62%), less in whole spices (maximum 56%, average 32%). The same spices, kept in the dark in glass flasks for six years in quantities of 5 to 100 g, lost 24% (average) essential oils. When the flask was hermetically corked and the spice filled the glass container, the loss was 0 to 5%, whether the spice was powdered or whole. If the flask was

only partly filled and not tighly sealed, the loss reached 62%. The spices keep well only when well-dried and well-conditioned.

Kaess (1950) evaluated quantitatively the penetrability of the odor of various odoriferous foods including spices, the odor of which is due mostly to volatile oils, esters, and adehydes. A special and fairly simple apparatus for the subjective analysis of odor is described. Various wrapping materials of German origin are tabulated according to their degree of permeability to odoriferous substances. Among the spices, cloves possess the highest penetration, whereas pepper, cinnamon, and caraway seeds are much less active in this respect. Coffee was nearly equal in penetrability to cloves. Types of bags suitable for the packaging of foods with strong odors are described in detail, and practical recommendations are made for reducing the amounts of volatile material given off by odoriferous food materials. The temperature of storage has an adverse effect on various types of wrapping materials (Kaess, 1950).

Kaess (1950) has also described an apparatus for the determination of odorous substances, which consists of a closed container with two compartments separated by the test sample of packaging material. Packaging material is tested with cloves as the odorous material. The test stands for 2 to 10 days at 20°C. The top compartment is then opened, and a rating is made organoleptically of the intensity of the odor with numbers of 0 to 4. As alternatives, one might use other spices or foods to determine the efficiency of the packaging material for the specific sample, or one might use holding temperature, which represents the normal holding conditions. Results from nineteen commercial packaging films and use of cloves, pepper, nutmeg, cinnamon, paprika, marjoram, and onion as the odorous material are tabulated. However, the test is rather time consuming, taking 2 to 10 days.

Packaging materials available in India have been screened for their susceptibility or resistivity to insect penetration and their resistance to transmission of vapors and odors. The relative merits of these materials for use in packaging spiced products have been discussed (Sreenathan *et al.*, 1962).

The difference in volatile oil content of nine spices and drugs stored in paper bags and in sealed tin boxes was investigated for two years by Balcar and Kozlowski (1962). The rate of continuous decrease of oil content in drug made from the dead parts of plants, such as chamomile *(Anthemis), Mentha* leaves, *Salvia officinalis* leaves, *Angelica archangelica* root, and *Acorus calamus* root, was considerably higher when they were stored in paper bags. In the umbelliferous fruits, a significant increase of oil content during the second year of storage followed a decrease during the first year. This was probably due to the secondary ripening of the fruits and the varying atmospheric conditions. Enzymatic decomposition of glycosides is possibly the cause of the permanent increase of oil content in some cases. The necessity of standardization of storage conditions of spices and drugs has been stressed.

Bulgarian anise fruits, stored in the dark at 5° to 25°C in paper bags containing 1 to 2 kg, were studied by Georgiev (1965). The fruit can be stored between crops under normal conditions without loss of quality. After four to five years, losses were as high as 50%. Half-ripe fruits retained the oil better than full matured ones. The content of anethole after four to five years was still 80%, making its production possible.

An attempt has been made by Srivastava *et al.* (1962) to review the work done on the storage and transporation of several important Indian spices with a view to suggesting improvements in existing practices. Some data on the role of pre-treatmtnets and the future line of work have been discussed.

Data are presented by Muthu and Majumder (1962) on the "serial fumigation" of different commodities, including packed spices, dosages, and the reuse value of the residual fumigant concentrations. The closed system in which the entire operation is carried out and the economy in the use of the fumigant are points to be considered for adopting the tehcnique for packaged spice products.

An infrared lamp, a hot-air oven, and a spin pasteurizer were tried by Muthu *et al.* (1962) for disinfesting Indian spiced multipurpose food and curry powder articiallly infested with *Stegobium peniceum*. The results showed that, for heat disinfestation of products containing spices, they should be subjected to heat only when packed in hermetically sealed containers. The same was found in spin pasteurization, which proved to be the best technique for disinfestation.

VII. PACKAGING OF CURRY POWDERS

A survey of the literature on the chemistry, microbiology, and technology of curry powders by Pruthi (1964) revealed that only four published reports (Misra and Pruthi, 1962, 1963a; Pruthi *et al.*, 1962; Pruthi and Misra, 1963b) were available on the packaging requirements of curry powders and other spice mixtures such as *samber* powder, *rasam* powder, *chutney* powders, and *garam* masala (Pruthi *et al.*, 1962; Pruthi and Misra, 1963b). Their main conclusions were as follows: (1) From the nature of the ERH curves obtained, the spice mixtures were arranged in the following decreasing order of hygroscopicity: pickle masala, chutney powder, curry powder, samber powder, garam masala, and rasam powder. The difference in hygroscopicity of different spice mixtures is due to differences in salt content and different compositions of the products under test. (2) The addition of 5% common salt to the curry powder slightly increased the hygroscopicity of the product, but had the advantage of delaying mold attack up to 22 days, compared with 5 days in unsalted curry powder. Thus, addition of common salt seems to act as a mild preservative against microbial spoilage in curry powders. (3) No marked change in the color of curry powder was noted up to 15% moisture content, but at moisture levels above 10%, there was a slight

tendency to cake. The materials remained quite free-flowing up to 8 to 9% moisture content. Therefore, it is better to keep the moisture content of curry powders and other spice materials down to well below 9%. (4) During open storage of curry powder at 50 to 80% R.H. at 37°C, there were considerable increase in moisture pickup and heavy losses in volatile oil content. At 70% R.H., mold attack was also observed, therefore, packaging of curry powders and spice mixtures in moistureproof containers is essential.

Misra and Pruthi (1963a,b) conducted systematic studies on the effect of type of container, vacuumization, gas packaging, and storage temperature on the overall quality and shelf-life of curry powders and other spice mixtures during 5 months of storage at different temperature. Their main findings were as follows: (1) Irrespective of the type of container and the storage temperature, there was a decrease in volatile oil and volatile reducing substance, the losses being the highest in polyethylene and the lowest in cans. (2) Vacuumization and gas packaging did not materially help in the retention of volatile oil. No significant change in moisture content was observed during the entire storage period in all the containers at all the temperatures, except in polyethylene bags stored at 37°C and 55°C, which lost most of the flavor and about 20% of the initial moisture content. The flavor retention was the minimum in polyethylene bags, because of the escape of volatile oil. (3) Color retention is polyethylene bags was, however, the best, probably because of the lowering of moisture content, which retarded the maillard reactions. The effect of moisture and storage temperature on the color of the product has been illustrated by studying the absorption spectra. There was, in general, maximum deterioration in both color and flavor of samples stored at 55°C and 37°C. (4) No correlation seemed to exist between the volatile reducing substances and organoleptic evaluation of samples stored at 55°C, although at room temperature and in fresh samples they seemed to be somewhat related. (5) In general, samples packed in hermetically sealed cans could be kept at room temperature or even at 37°C for 5 months. Glass bottles were the *next* best, while 250-gauge polyethylene bags were unsuitable for the packing of curry powders.

VIII. PACKAGING OF SPICE OILS, OLEORESINS, DRY SOLUBLES, AND DRY SEASONINGS

The available published information on packaging of spices oils, oleoresins, and dry seasonings is rather limited.

A. Spice Oils

An investigation by Kohler (1957) has been made of the resistance of polymethacrylate (Plexiglas), polyacrylonitrile (Plexidur), Ph OH-HCHO syn-

thetic resin, filled (Trolon), polystyrene without filler (Trolitul), filled polystyrene (Styron), and cellulose acetate butyrate (Cellidor) to vapor and alcohol solutions of essential oils (anise, caraway, and clove oils). The numerical values obtained of the changes in weight (grams per square meter per day) and the changes in the surface characteristics serve as a measure of the resistance of plastics to alcohol and these essetnial oils.

Recently, Pruthi *et al.* (1977) have shown that cinnamon leaf oil is corrosive and should not be stored in metallic containers. Packaging in dark-brown bottles gave better stability to the oil than did the white bottles. Packaging of cinnamon leaf oil in suitable PVC containers also appears to be a practical feasibility. However, more detailed study on commercial scale is needed in this respect.

B. Oleoresins

Packaging and storage studies conducted by Narayanan *et al.* (1964) reveal the stability of pepper oleoresin packed in 125- and 30-ml aluminum bottles at different temperature and humidity.

C. Seasonings

A new form of dry seasonings, known as Spisoseals, when encapsulated in a protective colloidal shell can be stored much longer than ordinary spices, without significant loss of aroma, flavor, or essential oil. The oil and/or oleoresin are dissolved in an edible gum solution, spray-dried, and then blended with a dry base such as dextrose or salt. The losses in flavor (volatile oil) in encapsulated Spisoseals were of the order of 2.4 to 3.4%, as compared with 28 to 62.4% in stored natural spices. Comparative data on flavor loss in encapsulated Spisoseals prepared from allspice, basil, celery, caraway, dill, ginger, black pepper, and nutmeg are presented along with those for control samples (Food Processing Staff, 1960).

Propylene (a polypropylene film) and Carplene (a nylon film) have been found to be superior to polyethylene in their impermeability to gases, odor, and flavor; in addition they can be heated, sterilized, and autoclaved (Food Processing Staff, 1960).

9

Storage and Transportation or Shipment of Spices

Scientific storage or warehousing is an important stage in the series of operations required for successful and orderly marketing of any crop. This applies particularly to spices, which are quite sensitive to moisture, storage temperature, light, and air or oxygen. In the absence of proper packaging and storage conditions, spices not only lose their characteristic aroma, flavor, and taste for which they are so highly valued, they also pick up moisture, become moldy and unattractive, lose volatile oil, and later are attacked by insects. Then spoilage sets in.

Since different spices behave differently during storage, they must be handled and treated accordingly. Spice packs are stored to different heights depending on the nature of the spice, the storage period, etc. As spices are likely to absorb moisture from floor space, dunnage is necessary. Ideal dunnage consists of wooden crates with one layer of mats spread over them or polyethylene sheets sandwiched between two layers of mats. Stacking is done by storing the bags in lengthwise and breadthwise layers. Usually only well-dried spices are stored. Care is also taken to store them in separate rooms with proper stacking, in a well-ventilated and damp-proof pucca godown or warehouse. In addition to all these arrangements, preservation methods such as dusting, spraying, and fumigation are undertaken.

The readily available published information on the storage of different spices is briefly reviewed.

I. STORAGE OF WHOLE SPICES

Except for a few spices such as onion, garlic, chilies, and black pepper, the published information on storage of whole spices is rather scanty. Information on commercial and bulk storage of spices is almost absent, as will be shown below. This subject needs further study.

A. Angelica

During two years' storage of angelica, mint, sage, and sweet flag in paper bags, there was a greater decrease in volatile oil content of mint leaves than in the corresponding sets packed in tin boxes (sealed). In umbelliferous spices, on the other hand, a significant increase in volatile oil content during the second year followed a decrease during the first year. This was probably due to the secondary ripening of the fruits, the varying atmospheric conditions, etc. (Balcar and Kozlowski, 1962).

B. Bay Leaves

According to the findings of Pruidze (1968), dry bay (laurel) leaves lost 30% volatile oil and 40 to 60% chlorophyll content during one year's storage. Because of the loss of chlorophyll, the leaves turned grey. A suggested method of assessing leaf quality is to determine the "coefficient of brightness" in relation to color. Of course, during storage, the hygroscopicity of the leaves was reduced, and leaf weight declined.

C. Caraway

Kofler (1936) reported an increase in essential oil content of caraway and fennel seeds during storage.

D. Cardamom

Work done at the Central Food Technological Research Institute, Mysore (1963), revealed that a moisture level of 12% and above is deleterious because of mold attack and color reversion, which is greater at higher moisture level as the storage period increases. Color reversion in green cardamom is mostly due to the combined effect of high moisture, high storage temperature, and light. Packaging of green cardamom in black 300-gauge polyethylene or in laminated aluminum foil helps in retention of green color at the end of 4 month's storage (Viraktamath *et al.*, 1964). However, bulk packaging for export of green cardamom needs to be developed for maximum retention of natural green color.

The natural attractive green color can also be stabilized to a great extent by steeping the green cardamom in 2% sodium carbonate for 10 minutes prior to air drying (Natarajan *et al.*, 1968a).

The two main constraints in the packaging of green cardamom are as follows: (1) The use of tin containers is uneconomical. (2) Bags of hessian, multiwall paper, or laminates are not suitable, as they are most likely to be punctured by the use of hooks in handling at various points of loading, unloading, transshipment, etc.

It is felt that a five-ply corrugated fiberboard box with a low-density polyethylene gusseted bag liner duly heat sealed would be a suitable pack for units of 25 kg of cardamom. To prove its efficacy, transport worthiness and storage tests need to be carried out before commercail exploitation of the idea.

E. Cassia and Cinnamon

Of the eight samples of cinnamon stored in closed glass containers for one year, two samples showed no change in essential oil content, while the remaining six samples showed a decrease of 2 to 8%. Samples stored in open paper sacks exhibited losses as high as 11 to 33% (Horvath, 1932). DuBois (1949) reports that freezing had little effect on the seasoning strength of cinnamon, nutmeg, or sage. Similar tests with applesauce flavored with cinnamon or nutmeg (stored at 21°C for 6 hours before serving) also showed that samples stored for 12 months compared favorably with freshly prepared samples.

F. Capsicums, Red Chilies, or Paprika

1. Storage Changes

In international collaborative studies, Pruthi (1969a) reported the loss of 3.68 to 16.48% of capsaicin in three grades of Hungarian paprika powder after 3 months' storage and a loss as high as 65.40 to 70.94% after 13 months' storage at room temperature. Kardose (1960) illustrated and discussed data relating to storage of red pepper under various conditions prevalent in the industry. Csiba and Kilb (1959) described the large-scale damage caused by *Rhizopus nigricans* Ehr. in paprika stored in strongs. This mold grows on the inner surface of the paprika, as a result of which the pericarp can be pulled back from the disintegrated mesocarp like a glove. The species discovered shows high enzymatic activity (pectinase, proteolytic enzymes), which causes the tissues to fall apart.

Chilies should be stored separately in suitable godowns or warehouses.

2. Factors Affecting Color Retention

Pruthi (1969a) reported that losses in color or capsanthin content of Hungarian paprika powder stored for 13 months at room temperature (25° to 40°C) were as

high as 58.9 to 64.2%. The color deterioration in chili or chili powder has been attributed to oxidation of carotenoid pigments, which is greatly influenced by moisture content, storage temperature, atmosphere, and light. Other factors, such as harvesting conditions, variety, and drying temperature, also influence color retention in whole and powdered chilies. The effect of important factors on the retention of color in whole chilies and in chili powder during storage will be briefly reviewed below.

a. Effect of Moisture Content. Chen and Gujmanis (1968) observed that chili powder samples stored with moisture contents of 9 to 10% retained better color than did samples stored with lower moisture contents of below 7.0% in oxygen atmosphere. Natarajan *et al.* (1969) studied the storage behavior of whole chilies stored in sealed cans for 6 months. They observed that the stored samples with moisture contents of 11.0 to 12.9% gave higher color values expressed as β-carotene than did samples stored with moisture contents below 9.0%. They also pointed out that the whole chilies stored with moisture contents of 11.0 to 12.0% turned black, whereas samples with moisture contents below 7.0% turned pale. The spectral curves of the color extract of blackened samples showed a shift in the absorption maximum, and there was an increase in carbonyl compounds, indicating the possibility of a nonenzymatic type of browning reaction (Krishnamurthy and Natarajan, 1973). These studies indicate that moisture has a profound effect on the deterioration of color in chilies. There is a need to differentiate the color in blackened samples in order to make any valid conclusion about the color deterioration in whole chilies.

b. Effect of Storage Temperature. In general, storage at higher temperature increases the rate of color destruction. The blackening of whole chilies is accelerated by the higher storage temperatures (Krishnamurthy and Natarajan, 1973). The loss of color is markedly slower at low-temperature storage (Lease and Lease, 1956a). However, when the refrigerated samples were brought to higher temperatures, the samples suffered from accelerated color loss.

Autoclaving of chilies before storage has no beneficial effect on color retention. This indicates that color changes are not due to enzyme reactions (Van Blaricum and Martin, 1951; Chen and Gujmanis, 1968).

c. Effect of Light. Sunlight exhibits a pronounced effect in bleaching the color and brings about maximum discoloration of the red pigments in chilies (De La Mar and Francis, 1969).

Studies on chili powder stored in glass bottles and in tins at 25°C for 10 months showed no significant difference in color retention between samples in the two containers. All the samples in bottles showed surface bleaching, and the color of the samples in tins faded uniformly with time (Lease and Lease, 1956a; Chen and Gujmanis, 1968).

d. Effect of Fat Content. Van Blaricum and Martin (1951) have tried to correlate the fat content of chilies with color retention. Seven samples that

retained most of the red color had a fat content of 10.0 to 13.8%; those samples that lost their color had a fat content of 28.0 to 30.8%. This hypothesis is further supported by the fact that, when the samples were ground without seeds, the color was much more stable. However, Lease and Lease (1956a) and Chen and Gujmanis (1968) could not confirm this observation. But the presence of high amounts of unsaturated fat in chilies and the fact that the pigments are present as esters of fatty acids cannot be overlooked in oxidative deterioration of color.

e. Effect of Antioxidants. Antioxidants for the preservation of the red color of chili powder have been successfully tried, but no significant effect on the retention of color in whole chilies has been observed. Antioxidants such as santoquin (6-ethoxyl-2-dihydro-2,2,4-(=trimethylquinoline), diphenyl-*p*-phenylenediamine (DPPD), L-tocopherol, butylated hydroxyanisol, propyl gallate, and ascorbic acid were effective for the retention of red color in chili powder during storage (Lease and Lease, 1956b). The treatment of chili powder with ethoxyquin (santoquin) suggested by Chen and Gujmanis (1968) afforded both substantial protection against color deterioration and an improvement in surface color during storage. The treatment was most effective in low-moisture samples. Preliminary work on the treatment given to whole chilies with butylated hydroxyanisole, propyl gallate, or ascorbic acid did not show a very significant effect on the preservation of color during storage (Natarajan *et al.*, 1969).

In view of the complex nature of the various chili pigments and the several factors affecting them, it is not surprising that controversies about the coupled oxidation of fat and pigments, the protective action of moisture in oxygen atmosphere, and the success or failure of antioxidant treatment on the retention of color do exist. All these points need further investigation. The phenomenon of blackening of whole chilies during storage also needs further research so that suitable control methods can be developed.

G. Clary Sage

Sviderskaja and Govsina (1968) have reported the dynamics of losses of essential oil in harvested clary sage flowers (*Salvia sclaria*) during storage in the open and under cover between harvesting and processing. On a dry-weight basis, essential oil losses from flowers stored in the open during the first few days amounted to 40%. In contrast, under cover, during the first 7 hours, the essential oil actually increased.

H. Coriander

Tanasenko and Sobo'eva (1961) studied the effect of the conditions of the storage of coriander on the linalool (coriandrol) content of coriander oils. Tanasenko (1962) observed that splitting of the fruits of coriander during picking

and threshing leads to a significant reduction in their essential oil content. Whole fruits lost 50% of the oil, while halves lost about 70% oil on storing for 6 months. The number of halves in a harvest was reduced to one-half by improving the picking method. During a comparative evaluation of different varieties of coriander with regard to their essential oil content, it is considered necessary first to separate the halves before making determinations of their essential oil content.

According to Rakova (1961), coriander seeds lose 0.18 to 1.5% of their essential oil upon prolonged storage, even at a storage temperature of 14°C. Only seeds stored in glass bottles showed the same essential oil content as was found in fresh seeds. Coriander seeds packed in hermetically sealed large containers lost about 30% of the oil. Among the wrapping materials examined, cellophane was the best, with an oil loss of 33% after one year's storage.

Viswanath and Ayyar (1934) recommended FFA (free fatty acids) values of the ether extract of coriander seeds as a valuable index of quality and age or storage life of the produce. In fresh coriander seeds, FFA ranged only from 4.3 to 8.1 mg of KOH per gram of oil. Samples showing 19.5 mg were definitely characterized as coming from old stock. Thus, the FFA of the ether extract of coriander could help in determining the degree of freshness of coriander seed.

Coriander generally darkens during storage. Baker (1921) studied the effect of bleaching of both fresh and dry coriander seeds with 4% SO_2. The results gave a satisfactory yellow color. Twice the amount of SO_2 did not further improve the color, and half the amount proved insufficient. In the wet steeping technique, coriander subjected to 2% SO_2 for 1 minute gave the most satisfactory results. However, compared with unbleached coriander yielding 0.195% essential oil, the bleached coriander yielded none.

I. Garlic

1. Physiochemical Changes during Storage

Singh *et al.* (1959a) studied the effect of type of packaging and storage temperatures on losses in weight, moisture, allyl sulfide, and antibacterial activity during storage of garlic, which increased progressively during storage at 37°C and at room temperature, but at 0° to 2°C there was no loss in allyl sulfide and very little loss in antibacterial activity. After storage for 20 weeks at 0° to 2°C, 24° to 30°C, and 37°C, the losses in allyl sulfide amounted to 0.0%, 31.6 to 34.4%, and 73.1 to 82.2%, respectively. Sprouting in the bulbs was noticed after 6 weeks' storage at 0° to 2°C only, whereas the bulbs at room temperature and at 37°C gradually dried up during storage.

The physiological losses in the weight of garlic bulbs were of the order of about 8 to 10% at 0° to 2°C, 21 to 22% at room temperature, and 58 to 60% at 37°C. No significant difference was noticeable in garlic bulbs stored in jute bags or baskets at the three storage temperatures employed (Singh *et al.*, 1959a).

Komissarov and Andreeva (1965) studied the changes in the carbohydrates of four varieties of garlic during 6 months' storage under cold (0° to 4°C) and warm (18° to 22°C) conditions. Under warm conditions, the inulin content decreased to 25% of the initial value, the disaccharides decreased to 10% of the initial value, and the monosaccharides decreased essentially to zero. Under cold storage, the decreases in the three types of carbohydrates were much less than they were under warm storage. Further, the changes in carbohydrates were directly related to changes in organoleptic quality.

2. Effect of Preharvest Foliar Spray with Maleic Hydrazide

Garlic and onion sprayed with maleic hydrazide (30 ml of solution at 750, 1000, and 1500 ppm per 40 to 50 onion plants or 90 to 100 garlic plants) 6 weeks before harvest and cured for 1 week at room temperature were stored at 32° to 35°F (0° to 2°C) and 80 to 90% R.H. There was not much depressant action on the respiration rate. Garlic did not root at all during the 6 months' storage, and sprouted only during the sixth month. Onions showed 100% rooting within 2 months; however, sprouting was slower than in the nontreated control (Date, 1960).

3. Smoke Curing of Garlic Bulbs

Although the practice of smoke curing of foods for improving their keeping quality as well as for imparting desirable organoleptic properties can be traced back to prehistoric times, it is only during the past two to three decades that attempts have been made to control the various steps in the process of smoke curing in order to obtain better products. The resultant desirable qualities in smoked foods are due to (1) partial dehydration, (2) incorporation of natural antioxidants, (3) impregnation of antiseptic constituents, (4) beneficial effect of heat on the destruction of microorganisms, and (5) improvements in organoleptic properties.

For the production of smoke, shavings and sawdust of hardwoods, particularly oak, are generally preferred to those of soft woods. The preserving properties of smoke have been attributed to aldehydes, phenols, and aliphatic acids. After briefly reviewing the subject of smoke curing, Srivastava and Mathur (1956) reported that Jamnagar garlic bulbs smoke cured for 7 hours were acceptable organoleptically and were better than the controls. On a 10% wastage basis, the storage life of garlic bulbs smoke cured for 7 hours was 38 weeks, as compared with 10 weeks of controls. Some evidence was also obtained to show that smoke curing inhibits sprouting of garlic bulbs to some extent.

4. Effect of Gamma Irradiation on Storage of Garlic

Metlitskii *et al.* (1964) observed that both garlic and onion are appreciably different from potato in their radiosensitivity. In irradiated onions, there is a

limited growth of the leaves of the previously formed cells. At the same time, in an onion irradiated with low doses, the flower stalk never forms, although the leaf develops normally. This has great significance in relation to the practical aspects of storing onions. It is quite possible that the dependence of the radiobiological effect on the time of irradiation of the garlic and onion will permit low doses to be used to retard germination, if the irradiation period is sufficiently precisely chosen. Messiaen and Pereau-Leroy (1969) treated garlic bulbs by spraying the plants with 30% maleic hydrazide at 180 and 360 ml/hl or with 40% 2,4-D at 30 and 60 ml/hl 1 week before harvest or by irradiation at 5000 and 1000 rads from a ^{60}Co source 20 days after harvest. The effect of 2,4-D on shoot and root growth was negligible, and that of maleic hydrazide was greatly inferior to that of irradiation, which suppressed growth.

J. Ginger

Srivastava *et al.* (1962) have reviewed the work done on storage and transportation of major spices including dry giner, chilies, pepper, cardamom, and turmeric. In another paper, Subramanyam *et al.* (1962) discussed the problems of storage, transit, and marketing of fresh ginger. Because of their perishable nature, ginger rhizomes rot, sprout, root out, and shrivel. To prevent such spoilage, and to extend its storage life under various conditions, numerous treatments were tried at three storage temperatures. A protective skin coating containing a fungicide and a plant growth inhibitor coupled with packaging in polyethylene bags gave the best results at all storage temperatures. With this treatment, ginger rhizomes could be kept in an acceptable condition for 4 days at 115° ± 1°F and 40% R.H., for 70 days at room temperature (70° to 85°F, 60 to 90% R.H.), and for 120 days at 35° to 38°F, 90% R.H. The respiration rate and rhizome rot of ginger were considerably lower. Moisture retention was better at the end of the storage period with this treatment than with any other treatment at all storage temperatures.

Brown (1973) reported on storage of fresh ginger in acidified metabisulfite solutions. After 3 months' storage, ginger suffered rapid and relatively more severe tip-tissue deterioration in sulfited solutions acidified with H_2SO_4 than in solutions acidified with citric acid. The extent of deterioration was related to acid concentration but was not directly affected by SO_2 content. Fermentation and turbidity were observed in several vats, but these conditions were effectively controlled at a SO_2 content of no less than 0.1% and a pH value no greater than 4.0. Information on the storage of dry ginger is rather scanty.

K. Leeks

Aas (1966) reported that leeks were more sensitive than onions to low-temperature injury and therefore more susceptible to disease during storage.

According to Suhonen (1970), the long-term storage of leeks for 6 months resulted in very heavy losses. At least half of this deterioration was due to evaporation of moisture, and this could be reduced by gas storage. Continuous storage in 10% CO_2 reduced the total loss to 42 to 48%. Storage at the freezing point (of leeks) also reduced evaporation, but the lower temperature caused damage by frosting. Predrying of outer leaves before storage did not reduce moisture loss. Keeping quality was improved by planting the leeks in moist peat or sand, since they rooted and absorbed moisture, which compensated for evaporation. The use of heavy nitrogen fertilizing lowered keeping quality, as did frost damage in the field.

L. Mustard

Twelve months' storage of whole mustard seeds (*Sinapsis arvensis*) reduced their volatile oil content by 54%, and that of ground seeds by 84% (Nikolaev, 1962).

M. Nutmeg

According to Sanford and Heinz (1971), prolonged storage of nutmeg resulted in changes in the volatile composition as determined by chromatographic and spectral analysis, etc.

N. Onion

There is voluminous published information on different aspects of onion storage, including curing, the effect of preharvest foliar spray with plant growth regulators on inhibition of sprouting during storage, physicochemical and biochemical changes during storage, cold storage, and controlled atmosphere storage, and physiological disorders.

1. Effect of Preharvest Foliar Spray with Plant Growth Regulators (Maleic Hydrazide) on Inhibition of Sprouting during Storage

Patil (1960) reported that preharvest foliage sprays of water or an aqueous maleic hydrazide (MH-40) reduced onion shrinkage in storage. Villalobos *et al.* (1966) observed that foliar sprays of 1200, 1800, and 2400 ppm MH applied 15 and 7 days before harvesting inhibited sprouting in stored Valencia onions. After 313 days' storage, all the treated onions, irrespective of concentration or application time, had sprouted less than the controls. Increasing the concentration of MH had no effect on sprout inhibition when sprays were applied 7 days before harvesting, but sprouting decreased linearly over the rising range of concentrations when stronger sprays were applied 15 days before harvesting. Earlier,

Mathur *et al.* (1958) observed that the noteworthy effect of a preharvest spray of MH was its pronounced inhibitory effect at a higher temperature range than at a lower one with regard to reduction in rooting, sprouting, and physiological losses in weight. It also reduced the wastage due to fungal diseases at both the high and low temperatures, although the effect was greater in the lower range.

Dhesi *et al.* (1966) studied the effect of MH and of α- and β-NAA (napthalene acetic acid) on storage losses in onions caused by sprouting, rooting, and rotting. The three growth substances were applied as preharvest foliar sprays, but only MH proved effective. The best concentration tested as 2000 ppm, followed by 1000 ppm. The timings of the preharvest spray did not make any significant difference.

Kepkowa (1966) in four-year trials applied MH at 2500 to 5000 ppm in 1000 to 2000 lb/ha on different dates in August or September to onions, which were then stored until June in four types of storage facility. The MH almost completely inhibited shoot and root growth in storage, practically doubled the amount of marketable onions, and also reduced disease and weight loss. Best results were obtained with treatment applied when 40 to 60% of the tops had bent over but were still green. Differences between the MH concentrations and the amount of the solutions were slight and inconsistent. The highest rate (5000 ppm in 1000 lb/ha), however, seemed to be the best for preventing growth both in storage and after planting out the stored bulbs. Large bulbs showed a greater response to MH than did small bulbs.

Javela and Rodriguez (1966) stated that spraying with MH at 1500, 2500, or 3500 ppm, 1, 2, or 3 weeks before harvesting, reduced the sprouting of onions stored at 4°C and 85% R.H. or under ordinary atmospheric conditions. The effect was directly related to the concentration. The weight of shoots produced was the smallest when the treatment was applied 2 or 3 weeks before harvesting. The low incidence of sprouting in untreated controls during the first 75 days of storage was attributed to good postharvest drying. The percentage of injured bulbs in the controls was much higher in ordinary than in cold storage; in the treated bulbs it was the lowest with the highest concentration of MH applied 2 weeks before harvesting. The international MH residue limit of 15 ppm was exceeded only in the case of the highest concentration applied 1 week before harvesting and with ordinary storage. Randhawa and Nandpuri (1966) noted the effects of foliar sprays of MH at five concentrations ranging from 100 to 2000 ppm, of β-NOA (napthoxy acetic acid) at 500, 1000, and 2000 ppm, and of the same concentrations of NAA applied to onions 3, 2, or 1 week before harvest. The most sprouting occurred in unsprayed control plots, but the inhibiting effects of β-NOA and NAA were very slight. The higher concentrations of MH were more effective than the lower, but the time of application had little effect.

According to Villalobos and Sagues (1967), the best results were obtained by spraying 15 days before harvesting with 6.5 liter/ha of the commercial product MH-30 in 500 liters of water. After 313 days' storage, 72% of the untreated

controls had sprouted, but only 4% of the treated bulbs sprouted. Blanco and Leiderman (1968) sprayed onion plants (var. *Pera bojuda*) with MH at 1000, 1500, and 3000 ppm, 18 or 10 days before harvesting. The bulbs were stored for 310 days. In the untreated controls, 60% of the bulbs sprouted, and 40% of the bulbs were in marketable condition and completely prevented from sprouting. The earlier treatment allowed scarcely any sprouting, but in this case only 20% of the bulbs were fit for marketing. Ertan and Duvekot (1969) observed no sprouting after 6 months in cold storage at 0° to 2°C and 85 to 92% R.H. Storage in wire mesh pens in the field also resulted in less sprouting than did traditional primitive methods; since it is simple and economical, it is recommended to local growers for storage for 4 to 5 months.

Kato (1970a,b) compared the effects of eight preharvest chemical treatments on stored onions. Maleic and isonicotinic acid hydrozides were equally effective, but thiouracil, hydrozine, and semicarbazine caused decay and functional disorders. MH inhibited transminase activity, which lowered protein synthesis and prevented leaf primordial growth. Increased concentrations of thiouracil, MH, isonicotinic acid, hydrazide, and hydrazine, in order of effectiveness, inhibited respiration. The molecular component of MH active in inhibition was determined. The occurrence of functional disorders and decay was attributed to an analog of nucleic acid and hydrazine or a hydrazide base. Kato (1970a) also conducted systematic studies on the mechanism of retardation of sprouting and the occurrence of functional disorders in onion bulbs sprayed with maleic hydrazide.

2. Cold Storage and Controlled-Atmosphere Storage of Onions

Large-sized onions lose less weight during storage than small ones. Because of high humidity, lots cold stored at 32° to 35°F as well as those stored at 46° to 50°F begin to root and germinate soon after commencement of storage. There is practically no rooting or germination at room temperature (59° to 87°F). It is generally recognized that dry storage is suitable for onions (Mathur *et al.*, 1952). Chawan and Pflug (1968) obtained the best results when onions were stored at 40°F either in 10% CO_2 and 3% O_2 or in 5% CO_2 and 5% O_2. The controlled-atmosphere onions appeared to be superior to onions stored in air at 40°F.

3. Physiochemical Changes during Curing and Storage

Moursi and Tewfic (1962) observed that the total content of fleshy scale leaves was very high during the early period of storage, declined from the sixth to the eighth week of storage, and later increased again. The decrease was coincident with differentiation and growth in the bulb. The percentage of protein nitrogen decreased, whereas the percentage of soluble nitrogen increased at this state of storage. Nitrate nitrogen increased slightly after the fourth week of storage and then decreased. Both amide and amino nitrogen decreased during the later stages

of storage. Woyke and Manczak (1965) stored bulbs at six temperatures (14° to 20°C, 8° to 12°C, 4° to 7°C, −3°C, −4°C, and −4°C), plus heating at 42° to 44°C for 2 days before planting out. The largest number of seed stalks from one bulb, on an average of all the varieties studied, was obtained with bulbs stored at 8° to 12°C, followed by bulbs stored at 4° to 7°C, and the lowest with bulbs stored at −3°C and above 17°C. The number of seed stalks and the seed yield from one bulb were not closely correlated. The highest average seed yield (11.92 quintals/ha), produced by bulbs stored at 8° to 12°C, 0°C, and 4° to 7°C, was significantly higher than that of seeds from bulbs stored at 17°C or from bulbs heated for 2 days before planting out.

Franklin *et al.* (1966) conducted storage experiments for four years on yellow sweet Spanish onions cured either in the field or in the store. The onions were stored (1) in bulk bins (8 × 8 × 10 feet and 12 × 12 × 10 feet), (2) in boxes (4 × 4 × 4 feet), or (3) in sacks and crates stacked on pallets. They were given the following combinations of treatments: (1) hand topping and store curing; (2) hand topping and field curing; (3) machine topping and partial field curing; (4) green, untopped onions cured in storage. The onions could be harvested early enough for forced ventilation curing to avoid the adverse effect of rain and cold on storage quality. Curing could be accomplished with forced ventilation using unheated air if harvesting was completed in September. The depth of bulk storage was found to be limited to 8 feet.

Kato (1966a) observed that onion bulbs had a rest period of about 30 days followed by a dormancy period of 60 days. Just before sprouting, leaf and root primordia started to elongate, especially when the bulbs were stored at 17°C. The rest period could be shortened considerably by removing the outer thickened leaves. The time of sprout emergence was influenced by light intensity and application of nitrogen fertilizer during growth. There was close correlation between bulb maturity and time of sprouting. Reducing the oxygen supply during storage retarded sprout emergence and led to physiological disorders. Treatment with NAA, glutathione, thiamine, or pyridoxine retarded sprout emergence, whereas GA (gibbrelic acid) induced sprout emergence in dorment bulbs, but not in resting bulbs. Kato (1966b,c) also studied changes in the metabolic activity of carbohydrates, nitrogen compounds, and growth substances during storage in relation to dormancy.

The National Agricultural Advisory Service (NAAS) of the Ministry of Agriculture, London (1970), have issued a useful bulletin giving advice on soil preparation, manuring, pre- and postsowing cultivation, seed sowing, harvesting, drying, curing, storing, grading, and marketing, followed by recommendations on the use of onion sets, on growing onions for salad and for pickling, and on chemical weed control.

For good storage, both high temperatures and high humidity must be avoided. The sensing elements in the stack should be placed at one-third of the height of

the stack from the top of the stack. For onions stored no later than the end of March, the stack must be kept as cold as possible, but not frozen. When the temperature of the ambient air is below the stack temperature, but above 0°C (zero), it is blown through the stack. The volume of air required is 100 ft^3/min per ton. The fan is controlled by a differential thermostat. Sensing elements are placed in the stack and outside the building. The air used should be at least 3°C colder than stack air. A frost thermostat is also incorporated to prevent the fan's blowing at temperatures below 0°C under British conditions or in colder countries. When onions are removed from ventilated storage to refrigerated storage, they should be passed over grading tables and the unsound bulbs removed. They can then be placed in bulk bins or netted and the nets placed in the bulk container. Containers should be close stacked and the store filled to force the air through the onions and prevent it from rushing down large spaces between. Several other useful and practical suggestions are given regarding drying after harvesting, curing, and storage, including refrigerated storage under special circumstances (National Agricultural Advisory Service, 1970).

Shalaby (1966) studied internal "doubling"—the development of auxiliary buds enclosed by scale leaves in onion bulbs—in relation to soil type, season, temperature, planting method (transplanting or direct seedling), planting distance, and size of the mature bulb. Results showed that both the genotype and the environment contributed to the occurrence of internal doubling. A correlation between internal doubling and fusarium-induced storage rot was indicated.

Kato (1966c) stated that a color reaction to tetrazolium by dormant bulbs took place in the terminal part of the stem, whereas sprouting bulbs developed color throughout. Fresh juice from a dormant bulb inhibited the growth of onion seedlings and germination of seeds (more so in dry than in moistened seeds). The inhibition was removed by washing the seeds in running water, although subsequent germination remained low. Heat treatment did not affect the inhibiting action or the pH of the juice. The inhibiting action decreased during storage at room temperature.

Bottcher (1967) published a review discussing the effects of fertilizers, irrigation, and harvest date on the storage of onions, types of onion storage, ventilation, and the possibility of limiting attacks by *Bortrytis allii*. Siregar *et al.* (1967) determined the storage conditions on standard plate counts of raw onions. Kopec (1967) observed considerable variability in the activity of reducing and oxidizing enzymes in stored fruits and vegetables (onion, parsley, and pepper) in relation to species, variety, and storage conditions. Enzyme activity declined during storage as a result of overripening, chemical retardants, and polyethylene packing. In contrast, during sprouting or during microbial infection, a substantial rise in enzyme activity was noted. Wayse (1967) observed that *Kharif* onions responded significantly to applications of 40 pounds of nitrogen, 60 pounds of nitrogen, or 40 pounds of nitrogen plus 60 pounds of potassium per acre with yield increases

of 30%, 10%, and 69%, respectively. The keeping quality of Kharif onions (harvested by the end of December) was much poorer than that of the Rabi crop (harvested by the end of February). Application of nitrogen significantly reduced weight loss during storage, but significantly increased sprouting in Kharif onions and decreased it in Rabi onions. Phosphorus and potassium had no effect on the keeping quality of bulbs.

Schipper (1968b) discussed factors affecting postharvest changes in quality of vegetables (including onions). Data on respiration of onions at various temperatures are given, and brief recommendations are made on methods of harvesting and handling to reduce losses in weight, appearance, and quality during storage. Schipper (1968a) observed that the loss in weight was not directly related to temperature but was closely related to the moisture deficit of the surrounding air. Although high humidity effectively reduced weight loss, it also favored development of fungus, especially at higher temperatures, and at 5°C and 10°C many of the onions formed new roots. At 20°C or below, loss in weight was about 0.25 to 0.35% per week. At 25°C, the corresponding loss was about 0.6 to 1%, depending on humidity.

Bottcher (1969) suggested that, since the weather has a great influence, fully mechanized harvesting should begin as early as possible. The necessity for removal of impurities, the treatment of the harvested onions with hot air, and the speedy removal of the filled sacks from the field are also discussed. Singh and Kumar (1969) stored onions grown at three levels of nitrogen (44, 78, or 112 kg/ha) and two levels of phosphorus (25 or 50 kg/ha) at room temperature. Rooting, sprouting, and weight loss were significantly greater in onions grown at higher nitrogen levels, whereas keeping quality was improved by the higher phosphorus level. At the start of storage, the moisture, protein, and vitamin C contents and the respiration rate were the highest and the carbohydrate content was the lowest in bulbs grown at the highest fertilizer levels. Changes in these values during 90 days of storage are tabulated, and their probable uses are discussed. Bottcher (1970) observed that cool-air ventilation only partially reduced rotting of stored onions. Ventilation with warm air (5° to 6°C) improved the external quality and more effectively reduced rotting, but hot air (30°C) was the most efficient. Thompson *et al.* (1977) reported on onion storage in the tropics.

Apeland (1971) characterized onions with poor scale quality as having thin, light-colored, dry scales, which crack easily and eventually fall off, resulting in a product that does not comply with the requirements set for Grade I in Norway. The results indicate the dry scales have a pronounced effect on respiration, and thus good, dry scales on the onions are one of the factors influencing its storage life. Kepka and Sypien (1971) observed that (1) onions were at their optimum maturity when more than 50% of the plants had their tops down, more than one-third of the older leaves were yellow, and the outer scales on the bulbs turned yellow and began to dry. (2) Onions harvested mechanically kept a little less well

than those harvested by hand, but the differences were insignificant. (3) Four methods of curing onions in the field were compared, but no difference in their keeping quality was observed. (4) The effect of increasing nitrogen (from 0.35 kg/ha, with 180 kg of phosphorus and 180 kg of potassium per hectare) on the keeping quality was studied. However, there appeared to be no regularity in the effect of nitrogen fertilizer, and any differences between treatments were small, changeable, and insignificant.

Vik (1972) studied the keeping quality and skin color of stored onions in 1968. The small onions produced by high rates of sowing kept better than did larger ones in the store. No difference in keeping quality was found between onions raised under a plastic tunnel and those grown in the open air. However, the spring crop grown under plastic tunnels had darker skins.

Freeman and Whenham (1974a,b) investigated flavor changes in freeze-dried samples of onions stored at ambient temperature for 40 to 224 days by sensory and biochemical methods. A probable explanation for the observed increases in flavor strength with increasing periods of storage is discussed.

In conclusion, many factors influence the storage life of onions. Under all storage conditions, onion bulbs continually lose water and dry matter, but the more serious losses arise from storage rots, sprouting, and rooting. Among the factors that are critical for successful onion storage are (1) choice of cultivar, (2) methods of culture, harvesting, and curing, (3) control of temperature and humidity in storage, (4) design of storage structures, and (5) use of sprout-inhibiting chemicals such as MH.

4. Effect of Gamma Irradiation on Storage of Onion Bulbs

Patil (1966) observed that irradiation of Ninomiya Globe onion bulbs with total dosages of 5000 to 60000 rads, applied in small or large daily doses, prolonged their storage life at 24°C and reduced shrinkage, as compared with untreated controls. External and internal sprouting were inhibited by treatment, but storage decay was increased. The growing points of treated bulbs were injured or killed and the growth of inner buds was prevented by irradiation. No precise relationship was established between the radiation dosage and the percentage of externally sound bulbs, shrinkage, external sprouting, or decay. Rounanizadesh (1969) almost confirmed these observations. Onions harvested on May 15th were given doses of 4000 or 6000 rads/hour on June 1st. They were then stored for 5 months at 22° to 25°C and 55 to 70% R.H. Sprouting occurred during storage in all nonirradiated onions, whereas only 10% of those given 4000 rads and none of those given 6000 rads sprouted.

Tada and Shiroishi (1969) observed no significant changes in the sugar contents of onion bulbs of cultivar Sappora yellow after irradiation with 7000 and 15,000 rads from a ^{60}Co source or after storage for 1 month at room temperature. Nandpuri *et al.* (1969) concluded that there was complete sprout inhibition with

doses of 9000, 12,000, 15,000, and 18,000 rads, but rooting increased with doses above 3000 rads and weight loss was reduced with doses above 9000 rads. The carbohydrate content rose slightly with the dose up to 6000 rads. The crude protein decreased, whereas ascorbic acid content rose with dose. Kawakishi *et al.* (1971) studied the effects of γ irradiation on the activity of cysteine sulfoxide lyase in onions and on the development of di-*n*-propyl disulfide in onion macerates. The lyase activity was decreased by irradiation with relatively low doses (7 to 15 krads) used for sprout inhibition but was restored to the level of the nonirradiated control during storage for 3 months at room temperature. The formation of disulfide was also reduced by irradiation with the same doses and to the same extent as that observed in the lyase activity. These relations between the lyase activity and the formation of disulfide were not so conspicuous at higher irradiation doses, and it is thought that other factors such as radiolysis of the sulfoxide may be involved.

Nair *et al.* (1972) observed that sprout inhibition of onions by γ irradiation is influenced by the physiological state of the bulbs and storage temperature after irradiation. Sprouting is accelerated at low temperature (10° to 20°C) or when the diurnal temperatures fluctuate widely (30°C maximum to 20°C minimum). Irradiation at 6 to 9 krads inhibits sprouting during storage under the above conditions only if radiation treatment is given when the bulbs are in the deepest state of rest. Doses above 12 krads cause a transient stimulation of sprouting regardless of the physiological state of the bulbs at the time of irradiation. Related biochemical mechanisms have been discussed.

Bandyopadhyay *et al.* (1972) suggest that γ irradiation in the dose range of 6 to 9 krads can be effectively employed for prevention of sprouting in stored onions, provided the bulbs are irradiated within a short time after harvest. Assessment of wholesomeness of irradiated onions through conventional feeding experiments poses certain difficulties, since onions, even where unirradiated, are not acceptable to laboratory animals except in very small quantities. However, information in this regard can be obtained by analysis of changes, if any, in chemical constituents of onions as a result of irradiation. In the present work, freshly harvested Red Globe onions were irradiated at doses of 6, 10, 20, and 50 krads and stored at ambient temperatures (25° to 30°C) for 3 months along with unirradiated control samples. A comparative study of volatile as well as some nonvolatile constituents of control and irradiated samples was carried out with the help of gas–liquid chromatography, thin-layer chromatography, and infrared spectroscopy. The results suggest that there is no effect of γ irradiation up to a dose of 50 krads on freshly harvested Red Globe onions with respect to the chemical parameters studied.

Vajdi and Peperira (1973) observed that γ irradiation destroyed bacterial flora in spices; ethylene oxide treatment affected the oil content of the spices and the color of paprika. The highest increase in microbiological flora was observed in

garlic sausage prepared from untreated spices, followed by ethylene oxide-treated and γ-irradiated spices during storage. The panel preferred sausages made with the γ-irradiated spices to the ethylene oxide-treated samples.

Salem (1974) investigated γ irradiation (8000 rads) of fresh onion bulbs used in the dehydration industry in Egypt. Irradiation reduced sprouting of the fresh bulbs, but did not affect the rotting of the bulbs. No effects were observed on sugar content or color (optical density at 330 nm) of the fresh bulbs and dried onion flakes. The irradiated onion had a noticeably lower ascorbic acid content, pungency (measured as volatile reducing substances), and free amino nitrogen content in comparison with values in nonirradiated bulbs. Darkening of onion flakes during storage for 8 weeks at 40°C did not differ when the bulbs were treated with γ rays before dehydration.

O. Black Pepper

White (1957) conducted large-scale storage and shipping tests on whole black pepper, with and without mold inhibitors, prophylactic treatment in lots of 2½ to 5 tons from each of the important producing countries such as India, Indonesia, Sarawak, and Singapur. On the basis of these studies, the following recommendations are made for storing and shipping of whole black pepper: (1) Pepper should be sun-dried to less than 11% moisture and garbled. All molds and insects should be removed from the pepper berries by cleaning, washing, drying, and garbling prior to storing. (2) The toluene distillation method should be used to determine moisture content in pepper. (3) After drying, the pepper should be placed in double burlap bags with polyethylene liners, preferably 3 mils (0.003 inch) or more in thickness. The liners should be slightly larger than the bags, to permit the strain to be taken by the burlap bags. A liner of 28 × 55 inches is suggested for Malabar and Lampong pepper, and 31 × 55 inches for Sarawak pepper. (4) The liners should be double-tied with strong cord (tied once, then the loose end folded over and tied again) and should be independent of the outer bag. (5) The burlap bags should be sewn shut in the regular manner, *without puncturing the liners*. (6) In storage or in transit, the filled bags should be handled with proper precautions to prevent puncturing the liners.

P. Shallots

Aycock and Jenkins (1960) reported that treating diseased shallot bulbs with Dowicide B before planting resulted in increased yields. Soil treatment with PCNB was also beneficial if conditions favored development of *Sclerotium ralfsii*. Drying the bulbs in the field after harvest resulted in less loss in storage than did bringing them to storage immediately (even with 6 days' artificial drying in heated chambers). The use of Dowicide B on dried bulbs also reduced the

amount of *Aspergillus niger* and improved the appearance of the bulbs. Sinnadurai and Amuti (1971) stated that shallots are grown in tropical areas where the length of day and the temperature range do not permit the growing of onions. Shallots were dormant for 70 days from the time of harvest to sprouting in storage at room temperature (75° to 95°F), during which period about 40% of the bulbs were lost owing to drying out and disease. Bulbs stored at lower temperature (40° to 60°F) sprouted more rapidly. Cool storage enables the crop from the previous harvest to be used for planting within 30 days of harvest.

II. IMPORTANT REQUIREMENTS OF WAREHOUSING AND HANDLING OF SPICES

For satisfactory arrangements for storage and handling of spices, particularly at ports, the following points need attention: (1) Warehouse facilities close to industrial units for the storage of raw materials as well as finished products. (2) Availability of quick and efficient transportation to the ports. (3) Dependable clearing, warehousing, and shipping arrangements at the port at economic rates. (4) Avoidance of unnecessary and careless handling. (5) Streamlining of the present time-consuming formalities so that the cargo does not have to face shutouts. (6) Provision of more and better space in the transit shed in the docks. (7) Avoidance of delays in carting so that the consignment does not miss the delivery schedule. (8) Establishment of an agency that specializes in this kind of package service and can ensure scheduled shipments. (9) Ideal pattern of shipping. (10) Despite a steep rise in rates of wharfage and demurrage in some countries, railway sheds and wagons are sometimes being used as warehouses. This again calls for a concerted effort to provide warehousing facilities for the inbound rail and road traffic. (11) A coordinated scheme to meet the needs of both the domestic goods traffic and seaborne trade is urgently required. Thomas (1965) has discussed in some detail the warehousing of spices in India.

III. STORAGE CONDITIONS ON CARGO SHIPS

Storage conditions on cargo ships should be examined. The temperature and humidity in the ships' holds must be watched closely, while en route, twice daily. Whenever the readings show that the temperature of the cargo is approaching the dew point, exhaust fans should be started to prevent excessive moisture condensation. A circulation fan should be kept in continuous operation. Spices should be stored on wooden platforms, off the floor and away from the sides of the ship. Any condensation of moisture on the sides of the ship should drop into a gutter and immediately be carried away before the bags become wet. A moist pepper

"hold" (lower cavity in deck for storage) has a tendency to heat, and thus it is essential that pepper be dried properly to a moisture content of 10 to 11% prior to being stored in the hold. It is apparent from the examination of numerous cargo ships that conditions conducive to insect and mold development will arise while the ships are en route to the importing countries, if the proper precautions are not taken in drying and garbling, even if every effort is made by the ship's personnel to regulate temperature and humidity.

IV. STORAGE OF GROUND SPICES

A. Coriander Powder

According to Torricelli (1937) of Switzerland, coriander powder lost 62% of its essential oil in five years when kept in small paper bags in darkness. Some samples lost as much as 90%, and samples kept in glass flasks in the dark lost about 24%. Hermetically corked flasks lost 0 to 5%, and flasks that were partly filled and not tightly sealed lost about 62%. Coriander will keep only when well dried and well conditioned (Rao *et al.*, 1925; Torricelli, 1937).

B. Garlic Powder

Results of storage studies on garlic powder packed in different containers—white and brown bottles, hermetically sealed tin cans, polyethylene bags, white and pink gelatin capsules—or in the form of plain and coated tablets and stored at three different temperatures—37°C, room temperature (24° to 30°C), and 0° to 2°C—for 10 months have been reported by Singh *et al.* (1959b).

In general, losses in allyl sulfide, antibacterial activity, and volatile reducing substances were higher at higher storage temperatures. At 0° to 2°C and at room temperature, cans appeared to be the best and polyethylene the worst in all three quality aspects, there being no significant difference among the other containers.

Loss of allyl sulfide and antibacterial activity progressively increased during storage at 37°C and at room temperature, the losses being highest at 37°C. However, there was no material loss of allyl sulfide and very little loss of antibacterial activity and moisture at 0° to 2°C. Other intersting aspects, such as the effect of nitrogen packaging and storage temperature (Pruthi *et al.*, 1959f), flavor and color (Singh *et al.*, 1959c), and nonenzymatic browning in garlic powder (Pruthi *et al.*, 1960b), have been discussed in Chapter 8.

Pruthi *et al.* (1959i) reported the thermal stability of allinase and enzymatic regeneration of flavor in odorless garlic powder. The stability of allicin and alliin in aqueous extracts of garlic and in garlic powder during storage at different temperatures was studied by Sreenivasamurthy *et al.* (1961), whose results show that allicin in the aqueous extract of garlic loses its antibacterial activity during storage, but alliin in aqueous extract as well as in garlic powder remains stable

during storage over long periods. Freeman and Whenham (1974b) investigated the flavor changes in onions stored at ambient temperature for 40 to 224 days by employing sensory and biochemical methods on freeze-dried samples. A probable explanation for the observed increases in flavor strength with increasing periods of storage has already been discussed.

C. Onion Powder

Factors known to affect caking of spice powders include moisture, storage temperature, relative humidity, particle size, composition, electrical charge, and method of packing or nature of container. Peleg and Mannheim (1969) studied caking in onion powder during storage, since caking reduces its commercial value. The addition of anticaking agents at several moisture levels was investigated. Low storage temperature, moisture content of less than 4% in the powder, and some of the eighteen anticaking agents tested (calcium stearate, aluminum silicate, etc.) prevented caking of onion powders for prolonged periods. The earlier work on the subject has also been reviewed.

Dehydrated kibbled onions of the Egyptian variety, containing 4 to 5% moisture and packed separately in cans or in polyethylene or Saran pouches, were stored at 15°C, 25°C, and 35°C at 44% R.H. for 39 weeks by Peleg *et al.* (1970). The temperature of storage had the most pronounced effect on product quality. The cans were found to be superior to the flexible pouches; however, Saran pouches could be used under certain conditions. Moisture content increased significantly in polyethylene pouches at all temperatures, but in Saran pouches only at 25°C. The absorbance of onion extracts as a measure of browning increased significantly with storage temperature and to a smaller extent with different packaging materials. Products in polyethylene deteriorated most, but the difference between Saran- and can-packed products was small. A similar trend was found in tristimulus color determinations. All products stored at 15°C and those in cans at 25°C were found to be of good organoleptic quality. The pyruvic acid development potential could not be used as a sensitive indicator for determination of product quality. Similar observations had been made earlier by Pruthi and Lal (1960) during short storage of onion powder exposed to different relative humidities at ambient temperatures.

However, intensive investigation is needed to obtain precise information on the exact nature of the caking phenomenon—its causes, mechanism, rate, and methods of inhibition.

D. Other Spice Powders

Griebel and Hess (1940) tabulated data on original volatile oil content (%) and percentage of losses thereof in ground spices in storage for 6 and 10 months in yellow paper, parchment, cellophane bags, tinned boxes, and glass bottles with metal caps. For cinnamon, the values were 1.00%, 0.82 to 0.95%, and 70 to 75%;

for pimento, 5.10%, 3.08 to 5.0%, and 2.18 to 3.76%; for cardamom, 1.95%, 0.80 to 1.80%, and 0.80 to 1.7%; and for white pepper, 0.95%, 0.10 to 0.79%, and 0.15 to 0.75%. Bottles were only slightly superior to the parchment and cellophane bags as storage containers.

Available published information on the storage of chili and turmetric powders is rather scanty (Lease and Lease, 1956a,b; Chen and Gujmanis, 1968). Pruthi (1969a) reported changes in capsaicin content and color (capsanthin content) of ground Hungarian paprika and Indian chili powder. There is need for systematic studies on packaging and storage of several such spice powders.

V. STORAGE OF CURRY POWDERS AND SEASONINGS

Misra and Pruthi (1963a) conducted systematic storage studies on the effect of the type of container, vacuumization, gas packaging, and storage temperature on the quality and shelf-life of curry powders and other spice mixtures, the results of which have previously been discussed in Chapter 8. Misra and Pruthi (1963b) also standardized a method of color and flavor evaluation of fresh and stored curry powders and spice mixtures. Pruthi (1964) reviewed all important aspects of curry powders, including manufacture, packaging, and storage.

Earlier, DuBois and Tressler (1943) studied the effect of seasonings on the maintenance of quality in storage of frozen pork and beef. Mathur *et al.* (1974) reported the development and storage characteristics of spice mixtures for use in dehydrated mutton curries.

VI. STORAGE OF OTHER SPICE PRODUCTS

A. Spice Essential Oils

The stability of citral in coriander oil decreases in the presence of atmospheric oxygen and moisture, at higher temperatures, and with increasing content of methyl-heptenone. The addition of an antioxidant mixture composed of 4-methyl-2,6-di-*tert*-butylphenol and 3-*tert*-butyl-4-hydroxyanisile and storage in containers made of aluminum or glass increase the stability of commercial citral. After 6 months' storage under these conditions, the product meets the technical specifications (Dmitrieva *et al.*, 1967). Sanford and Heinz (1971) studied the effects of storage on the volatile composition of nutmeg.

Spearmint oil may be stored in tin cans, galvanized drums, or aluminum kegs. Botta and Morassutti Botta (1956) studied the effect of storage conditions on the physicochemical properties of the oil of spearmint (*Mentha spicata*, Scotch var.). The oil is corrosive on lead and lead-containing tin, less on iron and copper, and not at all on zinc, aluminum, and tin. Iron and copper give the oil a pink and a green color turning to brown, respectively. Lead and lead-containing

tin cause some opalescence with a whitish precipitate. Carvone, esters, acid number, and specific gravity increase with age.

Pruthi *et al.* (1978c) conducted studies on physicochemical changes in cinnamon leaf oil (packed in brown and white bottles) during storage at room temperature (27° to 32°C) and concluded that it remained quite stable even after 15 months' storage without material changes in specific gravity, refractive index, optical rotation, eugenol content, solubility in 70% alcohol, flavor etc. However, there was some decrease in cinnamon aldehyde content, from 2.5% to 1.9%, and its color (optical density) increased from 1.021 to 1.180 in brown bottles and to 1.200 in white bottles, indicating that brown bottles are better for packaging and storage of the oil. In subsequent studies, PVC containers were also found to be suitable. Owing to its high content (85 to 90%) of phenol (eugenol), cinnamon leaf oil was found to be corrosive and attacked metallic containers of aluminum and even stainless steel gradually. On contact with metallic ions (Cu, Fe, Al, etc.), the color of cinnamon leaf oil first changed to borwn and then to a darker shade during prolonged storage. Iron and copper were found to react quickly, whereas stainless steel and aluminum reacted at a slow rate, indicating that metallic containers are unfit for packaging and storage of cinnamon leaf oil and, instead, brown bottles and PVC containers are recommended by Pruthi *et al.* (1978c).

B. Storage of Oleoresins

Hot oleoresin soon after its manufacture is run into cans or drums or further processed if necessary. Oleoresins prepared from seed spices (such as celery) require no further additions, since the fixed oil present in the seed results in sufficient fluidity of the oleoresin. However, oleoresins prepared from leafy spices (such as sage) and from bark spices (such as cassia) show a thick tar-like or hard resin-like consistency upon cooling. In order to convert such oleoresins into a usable form, it is customary to make additions of the corresponding essential oils, blend, seal, and store. In some cases, propylene glycol (or other food-grade solvent) is added (Langenau, 1959).

Narayanan *et al.* (1964) reported the results of storage studies on black pepper oleoresin and concluded that it was quite stable when kept packed in 30- and 125-ml aluminum bottles at different temperatures and humidities.

C. Storage of Encapsulated Spices

Spices encapsulated in a protective coating have greater stability.

Comparative flavor losses (volatiles) from Spisoseals of allspice, basil, celery, black pepper, and nutmeg and corresponding dry spice solubles and crude spices are presnted and discussed. Loss of flavor (volatiles) in Spisoseals of the above spices at the end of the 60-day storage period amounted to less than 4% in all cases and less than 3% in the majority of sample tests. Average loss was 2.7%.

This small loss is the result of mechanical breakage of some of the particles, with subsequent exposure and loss of volatiles. There were much higher losses of the volatile constituents of tested dry solubles, which averaged almost 50%, and corresponding ground crude spices lost over 40%. Likewise, flavor stability as indicated by organoleptic evaluation was better in encapsulated basil, celery, caraway, dill, and ginger than in the corresponding dry solubles or crude spices (Food Processing Staff, 1960).

D. Storage of Other Spice Products

During storage of tamarind concentrate, corrosion of tin containers, despite the high acidity of the concentrate (pH 2.3), is not so rapid, because of the solid nature of the product. However, even acid-lacquered cans show corrosion after long periods of storage (12 to 15 months). It is, therefore, desirable that the product should be dispensed only in glass containers with plastic lids. Large tins could be used for export, since the industrial users abroad take it out of these tin containers after arrival (Lewis *et al.*, 1970).

Thomas (1958) reported on the storage stability of dehydrated cream of onion soup fortified with vitamins (thiamine, ascorbic acid, and vitamin A palmitate) after storage at 70°F (21°C) and 100°F (38°C) for 6, 12, and 24 months. Flavor deteriorated, but there was no significant difference in palatability rating for fortified and unfortified soups throughout storage. Better vitamin retention was obtained in canned, dehydrated soup at 70°F (21°C). No significant vitamin loss took place during its reconstitution.

Balasubrahmanyam *et al.* (1979) conducted packaging and storage studies on ground turmeric (*Curcuma longa* L.) in flexible consumer packages and concluded that a moisture level of 12.1% was critical with respect to free flow characteristics of the product, aluminum foil laminate offered maximum protection against loss of volatile oil and ingress of moisture during storage at room temperature, a double pouch of 300 MSAT cellophane or glassine inside and 250 gauge low-density polyethylene outside offered adequate protection to the product for well over 4½ months under different conditions of storage, when the initial moisture content of turmeric powder was 9%, and polyethylene pouches alone were inadequate in giving the desired protection against loss of volatile oil, as nearly 60% of the oil was lost within 135 days of storage. Besides, printing on the polyethylene pouches became disfigured and smudged. Also the pouches became rather sticky. Similar studies have been reported on ground black pepper (Balasubrahmanyam *et al.*, 1978) and chili powder (Mahadeviah *et al.*, 1976). Pruthi (1980a,b) reviewed the quality control, packaging, and storage of ginger and turmeric (whole and ground) and has made several suggestions.

10

Sanitation—Microbiological Aspects

I. NATURE AND EXTENT OF MICROBIAL CONTAMINATION

Most of the spices used in food products are known to be contaminated to varying degrees with mold spores, yeasts, bacteria, and sometimes insects. They are also known to cause spoilage occasionally in canned foods to which they are added. Jesson *et al.* (1934) reported that spore-forming organisms present in a spice mixture containing coriander and white pepper were responsible for swells in canned chopped hams. Crossley (1938) found coloring materials and spices to be a prolific source of bacteria, including putrefactive anaerobes. The bacteriological studies conducted by James (1938) showed that ground mustard contained more microflora than the whole seeds, and that these microflora consisted of molds, yeasts, and certain bacteria causing "flat-sour" spoilage.

Fabian *et al.* (1939) observed that mixed spices used in pickles were responsible for their spilage, since all other factors were controlled. Detailed bacteriological tests carried out by the same authors on thirty-two whole mixed spices, whole spices, ground spices, and curry powder revealed that the total bacterial load varied from 4.0×10^4 per gram (in red pepper) to 6.7×10^7 per gram in whole mixed spices.

Yesair and Williams (1942) reported that sage and other spice ingredients used in pork sausage contained bacteria that caused rapid deterioration of the product. They conducted systematic microbiological examination of a number of spices and one sample of curry powder, to determine total bacteria, acid-tolerant bacteria, molds, total spore formers, spore gas formers, etc.

Castell (1944) found aerobic and anaerobic thermophiles in relatively large numbers in spices, unprocessed cereal products, food stabilizers, and powdered milk products.

Microbiological examination of seven samples of Indian curry powders reported by Lal and Sadasivan (1944) showed mixed bacterial flora; the bacterial counts taken after 2 months' storage at room temperature indicated a marked decrease in the count over that in the original fresh samples (from 5.4×10^7, it came down to 4.0×10^5). Proctor *et al.* (1950) and Pohja (1957) reported the microbial content of solid and liquid spices, and Coretti (1955a) determined the bacterial content of spices and described routine methods for bacteriological examination of spices.

Misra (1962) and Rao *et al.* (1962) conducted detailed microbiological examination of different brands of Indian curry powders and other spice mixtures (Table LV). The total bacterial load ranged from 4.5×10^5 to 9.0×10^7 per gram. The classification of different types of organisms present revealed that putrefactive mesophiles (anaerobic spore formers), yeast, and molds were absent in all the samples. The mesophilic aerobic spores ranged from 0 to 4.2×10^5 per gram. Flat-sour organisms were absent in some samples of curry powder, samber powder, and garam masala; in others, they ranged from 10.0 to 1.7×10^4 per gram, whereas all the samples except five had thermophilic aerobic spores (Misra, 1962; Rao *et al.*, 1962; Pruthi, 1964).

The accompanying tabulation indicates the loads of *Clostridium perfringens* in different types of pepper, which possibly served to contaminate foods that would otherwise be free from it (Silliker, 1963):

Type of spice	*Cl. perfringens*/g
Whole pepper (ground)	6
Cayenne pepper	2
White pepper	4
Black pepper	4
Chile pepper	12
Paprika	4

Black pepper contaminated with aerobic spores enormously increased the spore count of sausage (Silliker, 1963).

Strong *et al.* (1963) found that 5% of sixty spices examined were positive for *Cl. perfringens*.

Warmbrod and Fry (1966) found pepper to have a microbial load ranging from 3×10^3 to 23×10^6 per gram, and a mold count from 3×10^3 to greater than 3×10^5, but coliforms were fewer than 100 per gram. The prepared sausage seasonings had greater total counts and coliforms than did the individual ingredients.

According to Kinner *et al.* (1968), onion, celery, and white pepper powders

TABLE LV MICROBIAL EXAMINATION OF INDIAN CURRY POWDERS AND OTHER SPICE MIXTURES[a]

Curry powders and other spice mixtures	Microbial load per gram						
				Anaerobic spore-formers			
	Total bacterial count	Mesophilic aerobic spores	Flat-sour spores	Total count	Putrefactive mesophiles	Thermo-philes	Yeast and mold
Curry Powders							
Commercial	3.0×10^7	1.0×10^3	3.20×10^3	1.1×10^4	Absent	Absent	Nil
samples	8.0×10^7	8.0×10^7	1.70×10^4	8.1×10^5	Absent	Present	Nil
	6.0×10^7	1.4×10^4	2.00×10^2	2.9×10^4	Absent	Absent	Nil
	4.5×10^5	2.0×10^3	1.00×10^1	1.6×10^3	Absent	Present	Nil
	1.8×10^6	1.5×10	1.50×10^2	6.5×10^3	Absent	Present	Nil
	1.0×10^6	Nil	Nil	5.0×10^5	Absent	Present	Nil
	6.0×10^6	8.5×10^4	Nil	2.0×10^5	Absent	Present	Nil
	5.4×10^7	Nil	Nil	5.0×10^4	Absent	Present	Nil
	1.0×10^6	1×10^4	4.25×10^3	7.5×10^4	Absent	Present	Nil
	2.8×10^6	1×10^4	Heavy	3.7×10^5	Absent	Present	Nil
Laboratory samples							
Without salt	3.9×10^6	8.5×10^4	2.25×10^2	5.1×10^4	Absent	Present	Nil
With salt	1.3×10^6	2.7×10^4	4.25×10^3	7.5×10^4	Absent	Present	Nil
Imported sample (curry powder)	1.1×10^7	1.4×10^6	Heavy	2.4×10^6	Absent	Present	Nil
Samber Powders	3.0×10^7	8.7×10^4	9.60×10^3	2.2×10^5	Absent	Present	Nil
	9.0×10^7	2.3×10^4	2.40×10^3	1.7×10^4	Absent	Present	Nil
	1.5×10^6	Nil	Nil	Nil	Absent	Absent	Nil
Rasam Powder	9.0×10^7	7.0×10^3	8.60×10^3	1.4×10^4	Absent	Present	Nil
Garam Masala	9.0×10^7	5.0×10^3	2.20×10^3	4.0×10^4	Absent	Absent	Nil
	Nil	Nil	Nil	Nil	Absent	Absent	Nil

[a] From Misra (1962), Rao *et al.* (1962), Pruthi and Misra (1963c), and Pruthi (1964).

were contaminated with aerobic spore formers, the maximum being in celery. Pepper contained fewer than 10 coliforms per gram.

Hall (1969) reported the enhanced recovery of *Salmonella montevideo* from onion powder by the addition of potassium sulfite to lactose broth.

Bacillus subtilis and *B. mesentericus* were present in 94% of the spices examined by Goleiz (1969), coliform contamination being the highest in pepper, paprika, cardamom, and caraway. Vaughn (1970) found that dehydrated garlic and onion contained a limited number of thermophilic spores without any sulfide stinkers. In many countries, the majority of spices are imported, and in many cases by the time they reach the consumer they have deteriorated to some extent in quality, owing to loss of volatiles, insect infestation, or microbial action (Clark, 1970). The International Organization for Standaridization (1972) has drawn up microbiological specifications for dehydrated onion and garlic.

According to Krishnaswamy *et al.* (1971, 1973), the total microbial load in eight spices and three spice mixtures ranged from 20×10^3 per gram (refined salt) to 55×10^6 per gram (turmeric). Coliforms were present in black pepper, coriander, mustard, fenugreek, cumin, fennel, and curry powder; coriander had the maximum load (2400 per gram), and fenugreek the minimum (130 per gram). Yeast and mold infestation was present in all spices, black pepper being heavily infested (9800 per gram). Aerobic mesophiles were the highest in turmeric (20×10^5 per gram) and the lowest in fennel (6×10^2 per gram). The incidence of mesophilic putrefactives ranged from 26 per gram (fenugreek) to 920 per gram (coriander and fennel), but refined salt was free from it. Thermophilic flat sours were present in limited numbers in cumin and refined salt only. A noncoagulate type of *Staphylococci* was present in minimum numbers in some of the spices. *Clostridium perfringens* was present in black pepper, turmeric, coriander, mustard, and fenugreek (30 to 170 per gram). Sulfide stinkers and *Salmonella* were completely absent.

Swarup and Mathur (1972) have reported on the seed microflora of some umbelliferous spices.

Julseth and Deibel (1974) investigated the microbial profile of some selected spices and concluded that no organisms posing a public health problem were found in the spices examined. In those rare instances where such a contamination had taken place, a multiplication of these organisms (*Micrococci, Enterococci, Escherichia coli,* and *Salmonella*) appears to be precluded, and the longevity of vegetative cells in the dry state appears to be limited. When *Salmonella* cells were inoculated into pre-enrichment cultures containing allspice, cassia, onion, and oregano, there was a definite inhibition of growth.

Likewise, Krishnaswamy *et al.* (1974c) made preliminary observations on the survival of *Salmonella* in curry powder, samber powder, coriander, and chili powders. Microbiological examination of these products inoculated with a type culture of *Salmonella typhimurium* and stored at 37°C and at room temperature

revealed a decrease in *Salmonella*. However, in the coriander powder, *Salmonella* survived longer. In the initial stages of storage, destruction of *Salmonella* was slightly accelerated at 37°C. Powers *et al.* (1975) have reported on the microbiology of some processed spices.

II. CHANGES IN BACTERIAL LOAD DURING MILLING OPERATIONS

Pruthi and Misra (1963c) and Pruthi (1964) observed that, during milling of spices, the temperature of the material rose to 80° to 95°C, and a considerable reduction in the total bacterial load was noted in all the spices examined. Thus, the bacterial load came down from 3.0×10^8 to 5.0×10^3 in cumin, from 8×10^{11} to 1.0×10^5 in coriander, from 5.0×10^{10} to 1.0×10^5 in chilies, and from 3.5×10^9 to 1.2×10^6 in black pepper.

III. CHANGES IN MICROBIAL LOAD DURING PROCESSING OR COOKING

A. Effect of Heat Treatment on the Microbial Load of Curry Powder

Exposure of curry powder to 37°C did not cause an appreciable reduction in the bacterial load, but a gradual fall in count was noticed after 3 days' storage at 55°C; after 12 days' storage at 55°C, the sample of curry powder gave negative results for different groups of organisms and showed considerable reduction in total bacterial count (Misra, 1962; Rao *et al.*, 1962; Pruthi, 1964). This mild heat treatment was, however, not enough to completely destroy all the organisms.

B. Effect of Acidification (pH), Salting, and Cooking on Bacterial Load

The results given in Table LVI clearly indicate that the mere addition of acidified hot water (pH 5.0 to 5.5) to the curry powder was enough to destroy all the organisms (Misra, 1962; Rao *et al.*, 1962; Pruthi, 1964). This observation lends strong support to the conventional practice followed in India and some other countries of acidifying the curries either with tamarind extract, pomegranate seeds (Anardana), or tomatoes, which help considerably in the elimination of most of the spoilage organisms present in spices or curry powders.

In the salted and unsalted curry powders, there did not appear to be much difference in microbial load after 60 minutes of heating (Table LVI). It has further been observed that some microbial contribution is made by the common salt used in the curry powder (Misra, 1962; Pruthi, 1964).

TABLE LVI EFFECT OF BOILING OF CURRY POWDER WITH AND WITHOUT SALT AND ACID ON THE LOAD OF MICROORGANISMS[a]

	Microbial load per gram								
	Curry powder alone			Curry powder + 5% salt			Curry powder + 0.5% citric acid[b]		
	Time of boiling (min)			Time of boiling (min)			Time of boiling (min)		
Groups of microorganisms	Control	30	60	Control	30	60	Control	30	60
Total bacterial count	3.9×10^6	5.0×10^2	4.0×10^2	8.0×10^2	2.4×10^3	3.0×10^2	Nil	Nil	Nil
Mesophilic aerobic spores	8.5×10^4	1.0×10^4	1.1×10^3	4.4×10^4	1.5×10^4	1.6×10^3	Nil	Nil	Nil
Flat-sour spores	2.3×10^3	1.9×10^2	Nil	Heavy	3.3×10^2	1.1×10^1	Nil	Nil	Nil
Anaerobic spore-formers									
Total count	5.1×10^4	8.0×10^7	7.0×10^2	5.1×10^4	2.6×10^3	2.0×10^2	Nil	Nil	Nil
Putrefactive mesophiles	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil
Thermophiles	Present	Present	Nil	Present	Present	Present	Nil	Nil	Nil
Yeast and mold	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil

[a] From Misra (1962), Rao *et al.* (1962), and Pruthi (1964).

[b] The pH was adjusted between 5 and 5.5.

IV. INACTIVATION OF MICROFLORA IN SPICES

To eliminate spoilage due to natural contamination of spices with spoilage microorganisms, it is desirable to treat the spices either at the source of production or prior to their use in perishable foods such as meat and sausages.

Several processes have been described in the literature for the elimination of spoilage organisms in spices. They can be classified broadly as follows: (1) Cold sterilization by irradiation. (2) Vacuum fumigation, with or without heat treatment. (3) Thermal inactivation.

A. Cold Sterilization by Irradiation

Cold sterilization of spices and other foods by irradiation is a comparatively recent development. The first study in this regard appears to have been conducted by Proctor *et al.* (1950), who observed that cathode rays very markedly reduced or completely eliminated the bacterial content of a number of spices and condimental herbs at a dosage of 1.33×10^6 rep; more than 99.9% of the organisms were killed by exposure to the cathode rays from a Van de Graff instrument.

The various factors to be studied in any consideration of ionizing radiations as sterilizing agents for food include the following: the best type of radiation; appropriate packaging, with especial regard to size; dose required for sterilization; effect of treatment on flavor, odor, texture, enzymes, nutrients, and storage life of foods; and choice and costs of equipment and building. Work on these aspects has been reviewed by Proctor and Goldblith (1951).

Robinson *et al.* (1957) obtained similar results with soft x rays. They further reported that pepper lost some of its flavor with a dose of 1.5×10^6 rep. In a series of eight papers by Hall (1955–1958), the effects of ionizing radiations on spices were presented. This useful research work, done under contract with the U. S. Army Quartermaster Corps, is not generally available. The sterilizing action of the ionizing radiation on spices as well as a change in the sensory characteristics of black pepper and cinnamon was reported.

Lerke and Farber (1966) studied the effect of a dose of 2 Mrep of irradiation from a resonant transformer electron beam generator on the content of bacteria, yeasts, and molds of twelve spices. The microbial population of the products (bacteria, molds, and yeasts) were either completely eliminated or reduced to insignificant levels, but in the twelve spices studied there was a considerable change in the odor complexes that was noticeable sensorily, as well as in their content of volatile reducing substances (Table LVII). Black pepper lost most of the aromatic component. Cinnamon lost the pleasing aromatic component, but retained a more redicinal type of odor. Cloves had a weaker characteristic of odor.

The Hungarian workers Torok and Farkas (1961) studied systematically the

TABLE LVII EFFECT OF BETA IRRADIATION ON THE MICROBIAL CONTENT OF SPICE CONDIMENTAL HERBS[a]

	Microorganisms per gram × 10^{-3}[b]								
	Before β-irradiation				After β-irradiation				
	Bacteria		Yeasts and molds		Bacteria		Yeasts and molds		
Spices	Range	Average	Range	Average	Range	Average	Range	Average	VRS[c] reduction (meq/g)
Allspice	207.56–584.80	365.57	2.44–5.20	3.76	0	0	0	0	115.9
Black pepper	18,999–49,998	29,000	0.516–1.13	0.858	0	0	0	0	58.8
Cayenne pepper	1.15–3.47	2.32	0.048–0.132	0.084	0	0	0–0.003	0–001	15.5
Chili pepper	3.9–9.00	6.87	0.–0.003	0.001	0–0.006	0.003	0	0	35.9
Cinnamon	0.096–1.54	0.74	24	24	0	0	0	0	132.8
Clove	0.50–4.00	1.92	0	0	0	0	0	0	147.0
Ginger	0.72–2.76	1.62	0–1	1	0	0	0	0	—
Nutmeg	0.42–4.67	2.55	0.30–4.68	2.13	0	0	0	0	86.3
Oregano leaf	0.16–16.40	8.28	5.84–49.64	24.54	0	0	0–0.024	0.012	53.9
Paprika	71.95–155.78	101.90	0.048–0.216	0.098	0	0	0	0	24.7
Sage	1.40–5.68	2.31	0.80–1.36	1.24	0	0	0	0	65.5
Sweet basil	6.48–62.72	27.52	0.720–1.28	1.07	0	0	0–0.072	0.034	69.2

[a] From Lerke and Farber (1966).

[b] For ease of presentation, the microbial counts represent the actual count divided by 1000.

[c] Volatile reducing substances: 10-minute aeration period for all samples. A 0.025 *N* $KMnO_4$ solution was used. Results are expressed as milliequivalents of $KMnO_4$ reduction in VRS.

effect of ionizing radiation on the bacterial count of three ground paprika samples of unusually high bacterial population. The bulk of the bacteria found in the Hungarian paprika samples consisted of non-spore-forming, radiation-sensitive bacteria. Therefore, an irradiation level of 300 to 400 krads was sufficient to reduce bacterial count by 99.0 to 99.9%. The residual bacterial population after irradiation consisted mostly of radiation-resistant spore-forming microorganisms. Complete sterilization was possible only with the aid of an irradiation level of 1600 to 2000 krads. The effect of irradiation in reducing bacterial count was not influenced by equilibrium relative humidity (ERH) in the range of 10 to 90%. The quality of the Hungarian paprika was not affected by the radiation dosage necessary for pasteurization as established by the aid of a rotating-disk type of Munsell colorimeter, by the determination of the capsaicin content, and by the investigation of the absorption spectra of the benzene extracts of the respective pigments.

According to Buzinov (1962), crops of basil and anise and their essential oil contents increased when the seeds were irradiated with small doses of β rays.

Schonberg (1952) used an ultraviolet lamp or sterisol for the sterilization of spices that were spread in a thin layer on a conveyor belt 1 to 2 meters wide, 1.5 meters from the source of radiation. The industrial application of this technique as well as its economic feasibility needs further study.

B. Vacuum Fumigation

Smith (1940) reported that sausage prepared with pasteurized spices (heated with ethylene oxide under vacuum) kept 33% longer than sausage prepared from unheated spices and containing many bacteria and molds. He suggested that the use of practically "germ-free" spices in the food industries would materially improve the storage quality of many perishable foods.

Woodward (1949) found that the microorganisms of natural spices were reduced by treatment with 1000 to 20,000 ppm of ClO_2 in air or inert gas followed by heating the treated spices from 100°F (35°C) to about 225°F (107.2°C). Thus, pepper containing 3,250,000 bacteria per gram had only 750 bacteria per gram when treated for 30 minutes at 225°F.

Hall (1951) described a process in which the spices or other vegetable materials were first heated under vacuum and then impregnated with ethylene oxide in sufficiently high concentration to destroy bacteria, yeasts, and mold spores. Practically any ground spice can be treated by this process without loss of more than 1% of volatile oils and without the occurrence of residual ethylene oxide in the products. Sterilization by the ethylene oxide process is very effective in destroying spores, especially when the material treated is in a normally dry state. Dry, treated spices can be kept for a long time without danger of the spore count's increasing. Foods (for example, meats) containing sterilized spices would also have better keeping quality than those containing untreated spices.

According to a publication of the American Meat Institute (1953), ethylene oxide and carbon dioxide are used in gas purification treatment to reduce the microbial contamination, and methyl bromide is used for the control of insect infestation.

Another cold sterilization process, using ethylene oxide gas, kills bacteria in spices and other dry food ingredients. The process is said to be superior to fumigation and the prolonged heating methods of infestation control (Anonymous, 1954).

Coretti (1955b) reported that the exposure of spices to ultraviolet light is not effective for the inactivation of microflora. It is, however, possible to prepare nearly "bacteria-free" extracts from spices and to reduce the bacterial content of spices by heating, by treatment with ethylene oxide, and possibly also by means of β or γ radiation.

Coretti (1957) also found that ethylene oxide gas was very effective as a sterilizing agent for spices. However, strict control should be kept on the sterilization process, or the color and flavor can be adversely affected. Spices sterilized in this manner are considered quite nontoxic, as the gas is completely removed after sterilization. He suggested bacterial limits of 3000 organisms per gram for black pepper and 1000 organisms per gram for other spices.

Rauscher *et al.* (1957) investigated vacuum sterilization of spices, using pure ethylene oxide and Etox (90% ethylene oxide and 10% CO_2). They concluded that ethylene oxide could be used for disinfecting many foodstuffs including spices and dried egg powder. The best results were obtained by a vacuum of 95% decreasing gradually during sterilization to 20%, a temperature between 25° and 35°C, and, in most cases, 500 g of ethylene oxide per cubic meter is sufficient. Moisture fluctuations in the Hungarian climate did not influence the sterilizing effect. Tables are given showing the effect of ethylene oxide on various spices of bacteria at different concentrations and temperatures, and varying periods of exposure.

The U. S. Food and Drug Administration (1957) prescribed a maximum tolerance limit of 50 ppm for residues of ethylene oxide in or on whole spices. Misra (1962) and Pruthi *et al.* (1962) attempted the disinfestation of spiced products by fumigation with ethylene oxide and methyl bromide using different techniques such as in-package vacuum fumigation, bulk fumigation, and serial fumigation of artificial infected curry powder. Pruthi (1964) reviewed the subject of sterilization of spices and curry powder.

C. Thermal Disinfestation

Yesair and Williams (1942) suggested a drastic wet heat treatment of spices, such as their sterilization in steam at 15 psi (121°C) for 15 minutes and 5 psi steam pressure, which reduced the bacterial flora from 8.35×10^6 to 1 per gram

and from 7.5×10^5 to 100 per gram, respectively, but there was deterioration in the flavor of the spices due to high loss of the volatiles as well as other heat damage.

Flick (1949) presented data on temperature and time required for sterilization of glass-packed spice extracts in a high-pressure autoclave.

According to Harrington (1951), the milled spices, in the cartons or bags in which they are packed for distribution, are sterilized by heating under vacuum. Essential oils are extracted from some of the ground, sterilized spices (pepper, cloves, cinnamon, and vanilla) with a solvent, this operation being carried out in a closed system under vacuum. Schonberg (1955) claimed that, by suitable heat treatment of pepper and other spices, products free from bacteria and spores could be obtained. Eolkin and Bouthilet (1967) patented a process according to which dry particulate food was sterilized by a process consisting in vaporizing a pyrocarbonic acid diester by heating the mixture throughout the dry food. Thus, diethyl pyrocarbonate was heated to 30° to 50° while a flow of air was maintained through it to carry the vaporized diethyl pyrocarbonate through a conduit to penetrate the black pepper. After 15 minutes, the pepper was sterilized.

Misra (1962) and Muthu *et al.* (1962) attempted the disinfestation of spiced products by direct heating under an infrared lamp, stationary heating, and, for canned products, heating with agitation in a spin pasteurizer developed by Pruthi (1957) and Pruthi *et al.* (1959g).

Recent studies have shown antibacterial activity of onion components (N. F. Lewis *et al.*, 1977). Sharma *et al.* (1979) tested the various extractives of onion for their inhibitory activity toward the growth of the aflatoxin-producing fungi, *Aspergillus flavus* and *Aspergillus parasiticus*. Ether extract and lachrymatory factor (LF) of onion, which has been earlier identified as thiopropanol-s-oxide, were found to have potent antifungal activity. Steam-distilled onion oil, which is devoid of LF, was not as potent as ether extract and LF. Its major component, dipropyldisulfide, was ineffective as a fungal inhibitor. Ethyl acetate extract containing phenolics was also ineffective. Exposure of onions to γ irradiation at a sprout-inhibiting dose (6 krad) did not alter the inhibitory potency of the onion extractives which, however, appeared to be heat labile.

11

Sanitation—Insect Infestation and Its Control

Spices, like other agricultural commodities, are known to be contaminated with spoilage organisms such as bacteria, molds, yeasts, and insects. Some of these may be pathogenic bacteria and toxigenic molds. For instance, the presence of 1.5 to 2.5 μg of aflatoxins B_1, B_2, G_1, and G_2 per kilogram of pepper extracts has been detected by Scott and Kennedy (1973), who further analyzed twenty-four ground black and white pepper samples and over seventy capsicum pepper samples, which revealed the presence of low concentrations of aflatoxins in several samples. Mayr and Suhr (1972) have recommended fumigation of spices with Etox (ethylene oxide and CO_2, 9:1) and Krishnaswamy *et al.* (1974b) with ethylene oxide + methyl formate (1:1) for reducing or controlling the microbial count. In Chapter 10 the nature and extent of microbial contamination and changes in microbial load during milling, cooking, or processing operations were discussed, as well as the inactivation of microflora or sterilization of spices by (1) cold sterilization or irradiation, (2) vacuum fumigation with or without heat treatment, and (3) thermal treatment. This chapter will review the readily available literature on the nature and extent of insect infestation and its control.

I. NATURE AND EXTENT OF INSECT INFESTATION

The selective infestation of spices and spiced products by the spice beetle *Stegobium paniceum* is very commonly reported (Muthu and Majumder, 1974). There are reports of economic losses in exported spices, and the exporters must pay fumigation charges for shipped infested materials at the ports of landing. The

accompanying tabulation lists the species of insects that generally attack the eleven important spices, according to the observations of these authors.

Spices	Insect species
1. Black pepper	*Lasioderma serricorne* (Fabricus), *Stegobium paniceum* (Linn.), and *Laemophloeus minutus* (Oliv.) infest whole and coarsely broken pepper; *Tribolium castaneum* (L.), *Sitophilus oryzae* (L.), and *Oryzaephilus surinamensis* (L.) persist for 2 to 4 months; psocids and mites attack when moisture content favors mold growth.
2. Cardamom	*Stegobium paniceum*
3. Chilies	*Stegobium paniceum; Lasioderma serricorne* (Fabricus)
4. Cloves	*Stegobium paniceum*
5. Coriander	*Stegobium paniceum*
6. Caraway, cumin, and fennel seeds	*Stegobium paniceum*
7. Curry leaf	*Stegobium paniceum*
8. Dehydrated onion	*Stegobium paniceum; Oryzaephilus* spp.
9. Ginger	*Stegobium paniceum*
10. Tamarind pulp	*Caryedon jonagra; Calendra linearis*
11. Turmeric	*Stegobium paniceum*

White (1957) based his detailed observations on large-scale shipment and storage of black pepper (in 2½- to 5-ton lots) from different countries in Southeast Asia. He isolated as many as sixty-four species of insects or other arthopods, representing fifty-three genera in forty families of twelve orders (Table LVIII). Observations indicated that most of the insects were removed prior to shipment through proper cleaning, washing, and drying. It is probable that either reinfestation took place during bulk storage or in double burlap bags for some period prior to shipping owing to unavoidable circumstances, or reinfestation took place in ships' holds en route to the importing countries. This is especially probable in the case of psocids and mites if the atmosphere in the storage compartment had high relative humidity. Of course, washing does not remove completely the immature stages of insects from within pepper berries. The development and eventual emergence of such insects could have occurred. Further exploratory tests revealed that the cigarette beetle, the drugstore beetle, and a species of flat grain beetle could penetrate the three gauges (200, 300, and 400) of the polythene lining of the double burlap bags. However, it seems doubtful whether all these insects would attack pepper to the same degree as *S. paniceum*. Further, in complex populations of insects that attack the stored product, it is apparent that, even though a species may be unable to penetrate the material, it could gain entrance through the holes made by other species that were able to penetrate the bags or material (White, 1957).

According to Desikachar and Muthu (1962), safe storage of turmeric in

TABLE LVIII INSECTS AND OTHER ARTHROPODS FOUND IN SAMPLES OF WHOLE BLACK PEPPER FROM STORAGE IN THE ORIENT OR FROM SHIPMENT UPON ARRIVAL IN NEW YORK[a]

Order	Family	Species
Acarina (mites)	Acaridae	*Tyrophagus lintneri* (Osb.)
	Cheyletidae	*Cheyletus malaccensis* (Oud.)[b]
	Cunaxidae	*Cunaxa setirostris* (Herm.)[b]
	Erythraeidae	*Balaustrium* sp.
	Laelaptidae	*Haemolaelaps magaventralis* (Str.)
	Phytoseiidae	*Garmania* sp.
	Tydeidae	*Tydeus* sp.[b]
Araneida (spiders)	Not identifiable	
Coleoptera (beetles)	Anobiidae	*Lasioderma serricorne* (F.)
		Lasioderma sp.[b]
		Stegobium paniceum (L.)[b]
	Bruchidae	*Callosobruchus chinensis* (L.)
		Callosobruchus maculatus (F.)
	Chrysomelidae	
	Coccinellidae	
	Cryptophagidae (Near Hapalips)	
	Cucujidae	*Ahasverus advena* (Waltl.)[b]
		Oryzaephilus surinamensis (L.)[b]
		Oryzaephilus mercator (Fauvel)[b]
		Laemophloeus minutus (Oliv.)
		Laemophloeus sp.[b]
	Curculionidae	*Sitophilus oryzae* (L.)
	Dermestidae	*Attagenus piceus* (Oliv.)
		Anthrenus verbasci (L.)
		Thylodrias contractus (Mots.)
		Trogoderma inclusum LeC.
		Trogoderma sp.[b]
	Lathridiidae	*Microgramme* sp.[b]
		Holoparamecus kunzei (Aube)
		Corticaria sp.[b]
	Mycetophagidae	*Typhaea stercorea* (L.)
	Scolytidae	*Xyleborus* sp.[b]
	Tenebrionidae	*Xyleborus* sp.[b]
		Tribolium castaneum (Hbst.)[b]
		Tribolium confusum (Duv.)[b]
Collembola (spring-tails)	Not identifiable[b]	
Corrodentia (book lice)	Liposcelidae	*Liposcelis entomophilus* (End.)[b]
		Liposcelis granicola (B. and H.)
		Liposcelis bostrychophilus (Bad.)[b]
		Liposcelis hospes
		Liposcelis terricolis (Bad.)

(*continued*)

TABLE LVIII (*Continued*)

Order	Family	Species
	Peripsocidae	*Ectopsocus* sp.[b]
Diptera (flies)	Phoridae	
	Chloropidae	
	Drosophilidae	*Drosophila melanogaster* (Mg.)
		Drosophila sp.[b]
	Lonchaeidae	*Lonchaea polita* (Say)
Hemiptera (true bugs)	Anthocoridae	*Cardiostethus rugicollis* (Champ.)[b]
Hymenoptera (ants, bees, etc.)	Formicidae	*Camponotus* sp.
		Lasius sp.
		Pheidole sp.[b]
		Solenopsis geminaia rufa (Jerd.)
		Solenopsis sp.[b]
	Pteromalidae	*Sphegigaster* sp.[b]
	Chalcidoidae	
	Cynipidae	*Eucila* or *Kleidotoma* sp.
	Platygasteridae	
Isoptera (termites)	Rhinotermitidae	*Reticulitermes flavipes* (Kol.)
	Kalotermitidae	*Cryptotermes* sp.[b]
Lepidoptera (moths)	Gelechiidae	*Sitotroga cerealella* (Oliv.)[b]
	Phycitidae	*Ephestia* sp.[b]
	Tineidae	*Setomorpha rutella* (Zell.)
Pseudoscorpionida (scorpions)	Not identifiable[b]	
Thysanura (silverfish)	Leptsmatidae	*Lepisma* sp.

[a] From White (1957).

[b] Specimens taken from sealed samples of pepper from the Orient. Most of the species were also recovered from regular commercial shipments.

godowns becomes a problem particularly during the monsoon season, owing to attacks of *S. paniceum.*

Blumberg (1939) has reviewed the literature on the pharmacognostical aspects of spices and drug infestation by insects and presented experimental data on the role of *Tribolium* beetles as pests. In general, it would seem that carbohydrate-containing spice drugs are quite susceptible to insect attacks. Of course, carbohydrates are not the only substances required; protein or fats or both are also necessary. Onion, ginger, and decorticated cardamom sustained the life of *Tribolium* for varying lengths of time. Contrary to previous reports, *Tribolium* could not be cultured in capsicum, nutmeg, cinnamon, or mustard. According to McCormack (1954), the cigarette beetle, the confused flour beetle, the saw-

toothed grain beetle, and the Indian meal moth are frequently found in spices. The methods of examination of different types of contaminants found in spices, including such techniques as flotation, sedimentation, and microscopic examination, are described.

Harris (1961) stated that three species of insects—*Myzus persicae, Liriomyza pusilla,* and *Plantysenta sutorare*—were the important pests of celery in the Everglades area of Florida.

II. INSECT DISINFESTATION BY HEAT TREATMENT

In the absence of any published report on the control of insect infestation in curry powders, Misra (1962) and Muthu *et al.* (1962) conducted systematic studies to explore the possibilities of adopting the following methods of disinfestation of spice products: (1) thermal sterilization and (2) fumigation with ethylene oxide and methyl bromide, using different techniques such as in-package vacuum fumigation, bulk fumigation, and serial fumigation of artificially infected curry powder with *Stegobium paniceum*. Their main conclusions are summarized here.

A. Thermal Disinfestation with a Spin Pasteurizer

Insects succumb to high-temperature treatments; they die if their body temperature is raised to 60°C (140°F) for 5 or 10 minutes. Owing to the insulating effect of the substance in which the insects are embedded, the exposure time and temperature required in practice are enormously greater than those needed to kill unprotected insects (Muthu and Majumder, 1974). Disinfestation by heat was attempted by (1) direct heating under an infrared lamp, (2) stationary heating in a hot-air oven at 100°C, and (3) agitated heating in a spin pasteurizer (Pruthi, 1957; Pruthi *et al.*, 1959g; Muthu *et al.*, 1962). The thermal death time, determined for 100% mortality of the pest, was found to be 50° to 60°C for 10 to 12 minutes; that is, a time–temperature of 600° centigrade–minutes ($T \times t$ product) was required for 100% mortality. Spin pasteurization of the product in hermetically sealed 4-ounce cans rotated axially at an optimum speed of 250 rpm in atmospheric steam at 93° to 94°C proved to be very efficient for 100% mortaility of the pest (Table LIX). There was no material loss in volatile oil content, nor was any serious heat damage to the flavor or color of the treated product detected. However, the comparative economic feasibility of this method in relation to cold sterilization by fumigation is yet to be established (Misra, 1962; Pruthi, 1964).

B. Heating by Infrared Unit

An *infrared heat-disinfestation unit* has been developed by Majumder *et al.* (1963) in which a battery of infrared lamps of 150-watt capacity are aligned to

TABLE LIX LETHAL HEAT TREATMENT OF CURRY POWDER TO CONTROL *STEGOBIUM PANICEUM* ADULTS IN 4-OUNCE CANS[a]

Temperature (°C)	Exposure (min)	Mortality (%)
50	120	100
60	60	100
70	45	100
80	45	100
100	15	100

[a] From Muthu *et al.* (1962).

heat a rotating aluminum or stainless-steel tube in an inclined plane. The infested materials are fed through a hopper onto the tube, which attains a temperature of 110° to 120°C. The material takes 1.5 to 2.5 minutes to travel from the feeding end to the exit, by which time it attains a temperature of 65° to 68°C. One hundred percent mortality of the insects at all stages of development is possible by this method.

C. Heating in a Hot-Air Oven on a Home Scale

A 10-pound sealed tin containing curry powder required a 2-hour heat treatment in a hot-air oven at 100°C, whereas a 1-pound tin required only 1 hour at 60°C. Similar conditions were required for the material exposed in shallow steel or aluminum trays on a home scale (Muthu and Majumder, 1974). However, the practical or commercial feasibility of this method is yet to be established. Furthermore, large quantities of bulk materials are difficult to treat, as they require a well-insulated room heated by steam or electric heaters.

D. Forced Hot-Air Circulation

Forced hot-air circulation will be useful if a uniform temperature at all levels can be maintained by efficient recirculation, extraction of air from around the top of the room, and injection of it again at the bottom. It is necessary to raise temperatures from 120° to 130°F in all parts and to maintain them for 10 to 12 hours. With a steam pressure of 50 to 75 pounds, a temperature up to 200°F can be quickly obtained. A vault temperature between 180° and 200°F can easily disinfest 100-pound bags of powdered spices in 24 hours (Cotton, 1963).

To summarize, heat treatment may be applicable on a small scale under domestic conditions, when small quantities of spices in domestic larders, which may develop insect infestation, could be easily disinfested by heat.

III. IRRADIATION OF SPICES FOR MICROBIAL AND INSECT DISINFESTATION

Microbial decontamination of spices by cold sterilization or irradiation has already been reviewed and discussed in Chapter 10. Lewis (1974) also briefly reviewed the physical methods of insect disinfestation and microbial decontamination in spices, including microwave treatment and ionizing radiation. From a microbiological standpoint, any process utilizing γ radiation can be categorized into the following three processes.

1. *Radurization Process*. The radurization process normally involves the use of γ radiation in the range of 0.1 to 0.5 Mrad, to bring about reduction in the initial microbial load. This process is synonymous with pasteurization. The effectiveness of the radurization process for spices is correlated with the radiation sensitivity of microorganisms initially present in the spice. Thus, before developing a radurization process for treating spices, one has to examine the types of microorganisms present in a particular spice. After characterizing the microorganisms typical for each spice, the radiation sensitivity of these organisms will have to be determined.

2. *Radicidation Process*. Radicidation refers to destruction of a specific organism, usually a pathogenic species.

2. *Radappertization Process*. Radappertization is analogous to sterilization—that is, total destruction of microorganisms. This process requires the use of high doses of radiation in the range 2.5 to 4.5 Mrads.

Radappertization is the ideal process for microbial decontamination of spices; however, a serious limitation of this process is the destruction of quality attributes such as color, texture, and aroma, which is known to occur in foods subjected to high doses of radiation. Nevertheless, these deleterious effects are pronounced in foods containing high moisture contents and are minimal in dried foods. Thus, the organoleptic qualities of spices that have low moisture contents may not be significantly affected even at radappertization doses.

A. Effect of Ionizing Radiation on Spices

Hall (1955–1958) undertook systematic research work to cover the following three aspects of irradiation of spices: (1) To eliminate insect infestation in twenty-one spices and spice products by ionizing radiation; (2) to study the radiation effects on spices in doses sufficient to produce near or absolute product sterility; and (3) to study the general effects of spices when added to meat products that are irradiated with doses sufficient to produce partial pasteurization or absolute sterility in the meats, with particular interest lying in the effects of the irradiation on the spices themselves, the effect of the spices in preserving the desired flavor of the product, and the effect of the spices in covering or inhibiting the formation of irradiation flavor. Their main findings are as follows:

1. Insect infestation can be eliminated in spice products by ionizing radiations. In practically every case, about twenty irradiated spices examined showed no evidence of live insects after 3 months of incubation; this was at levels of 50,000 and 75,000 rep. The spices treated were allspice (ground), anise seed (whole), black pepper (ground), celery flakes, chili powder, cinnamon (ground), cloves (whole), cumin (ground), ginger (ground and whole), marjoram (French and Chilean, ground), nutmeg (ground), onion (flakes), paprika (ground), red pepper and clary sage (rubbed), savory (powdered), and white pepper (Worseck, 1958).
2. When irradiated with 3×10^6 rep, no significant change in the character of the spice itself occurred, as measured by triangular comparisons with a panel of about twenty members.
3. None of the spices investigated, when combined in frankfurters and pork sausage and irradiated at 0.75 to 3.0×10^6 rep, showed any tendency to significantly mask or prevent ionized flavor and odor. In general, irradiation appears to reduce the odor and flavor of spices in pork sausage.

The effect of ionizing rays on spices has also been discussed by Worseck (1958, 1959). The doses of irradiation required for obtaining the various effects (for example, pest control and inactivation of enzymes) are indicated. The author also deals with the practical application of irradiation and its effects on meat, eggs, milk, butter, spices, fruits, and vegetables. Irradiation with doses of 0.1 Mrep and higher in meat leads to flavor changes caused by fermentation of H_2S and organic sulfur compounds. Details of the de Graft generator suitable for the sterilization of foodstuffs (made by M/s. High Voltage Energy Corp.) are given in the form of a table.

B. Irradiation of Onion Powder and Soup Powder

Heat sterilization of onion powder, which is often contaminated by spore-forming mesophile bacterial flora, is not practicable because of the volatility of the aromatic substances formed by the enzyme system present. After irradiation with 0.8 Mrad, the surviving microbial flora contained *Clostridium, Bacillus, Enterococcus,* and *Micrococcus* strains, which, however, did not develop as spoilage organisms during storage. The unwholesome flora introduced with the onion powder varies with the product in which it is used. In soup powders with short cooking time, a change is caused by the growth of *Lactobacillus* strains. A radiation treatment with 0.4 Mrad gives a sterile soup when boiled for 5 minutes. In meat pastes, spore-forming anaerobes are most dangerous, but they can be eliminated by an irradiation dosage of 0.2 Mrad. In general, onion powder can be decontaminated by treatment with 0.2 to 0.4 Mrad, a dose that has no detrimental effect on the organoleptic quality. The cost of irradiation is compatible with the price of the product treated (Delkinova and Dupuy, 1973).

C. Radurization of Chili Powder

On the basis of his initial experiments, Lewis (1974) stated that γ radiation has a tremendous scope for enhancing the quality of spices. It was found that radurization of chili powder at doses of 0.3 and 0.5 Mrad could substantially reduce both the bacterial load and the mold count. The microbial destruction curve indicates that at doses up to 0.5 Mrad the microorganisms were killed more rapidly than at doses from 0.5 to 1.0 Mrad, indicating that chili powder probably contains some radioresistant organisms. Also, it was found that a dose of 0.5 Mrad did not quantitatively affect the carotenoid or capsaicin content of chili powder.

D. Limitations of Irradiation of Spices

In addition to (1) the high cost of installation and plant involved, (2) the greater technical expertise required in the industry, (3) the likely health hazards due to lack of precise information on safety limits, nutritional aspects, and wholesomeness of many spices not studied so far, and (4) the adverse effect on flavor and color, irradiation at a dosage rate necessary for sterilization very often results in off-flavor, change in color, etc. (Mayr and Suhr, 1972). According to Lewis (1974), the deleterious effects relating to quality attributes such as aroma, color, and texture are more pronounced in foods containing high levels of moisture. The exact chemistry of these changes and processes has not yet been worked out. Also, their side effects are more troublesome than those of fumigation.

IV. INSECT CONTROL BY FUMIGATION

For quick, efficient, economic, and safe disinfestation, fumigation with a suitable toxic gas or vapor is the treatment of choice. As the fumigant can move both into and away from the commodity, the chance of residues of the chemical in the commodity is much less.

A. Common Effective Fumigants and Their Mixtures

More than thirty fumigants have been screened for their ability to kill insects (Food and Agriculture Organization, 1968). Of these, the most common effective fumigants are methyl bromide (MB), ethylene dibromide (EDB), and their mixture, ethylene oxide alone and in admixture with CO_2 or methyl formate (1:1), and aluminum phosphide or phosphine.

Investigations at the Central Food Technological Research Institute (CFTRI), Mysore, have shown that mixtures of EDB and MB (Durofume) in various

proportions are much more effective against insects than either of them is when used alone (Majumder and Muthu, 1964). A 1:1 (w/w) mixture is recommended at a dosage of 32 g/m^3 with an exposure time of 48 hours either in air-tight godowns or under gasproof tarpaulins. A 1:2 mixture of EDB and MB at 48 to 64 g/m^3 with an exposure period of 6 days has been successfully employed in medium-sized bag stacks (5 to 6 feet in height) of turmeric. Single-bag treatments can be done with Minifume tablets (Pillai *et al.*, 1970). Methyl bromide alone at 16 to 32 g/m^3 can be used with an exposure time of 24 hours, provided ideal airtight conditions are available, as MB is very volatile, having a vapor pressure of 20 psi at 30°C (Muthu and Majumder, 1974).

It is claimed by Mayr and Suhr (1972) that no significant differences can be found between an unfumigated and a fumigated spice, provided the fumigation is done properly by using adequate installations and techniques. Differences can, of course, be detected by sensitive physicochemical methods. All the typical quality attributes such as color, aroma, and texture remain unchanged.

Since the early fifties, a tremendous amount of basic and applied research work has been done on fumigants. This research has been reviewed by Thompson (1966), the Food and Agriculture Organization (1968), and Mayr and Suhr (1972) and discussed in several standard books. It has also been shown that mixtures of fumigants such as Etox (ethylene oxide 90% and CO_2 10%) and similar mixtures are more effective and less risky than using ethylene oxide (EO) alone. Addition of CO_2 not only reduces the inflammability of EO slightly, but also has a stabilizing effect, so that the gas may be stored for longer period. Furthermore, the use of pure EO alone can never be recommended because of possible disastrous explosions even at ambient temperature, due to polymerization.

The basic concept of achieving sterilization through fumigation is the destruction of the protein of the bacterial cell, which inhibits the metabolism of the organism. The EO–CO_2 mixture (9:1) is applied generally with the help of vacuum fumigation equipment, which makes it quite safe. This mixture had only a negligible effect on color, pH, etc. (Table LX).

Another important point is that the chemical action of EO takes place in solution. Thus, if one not only makes use of relative humidity during the sterilization process to effect the solution of EO, but also increases the humidity at an earlier stage, this prepares the microorganisms by causing their membranes to swell, which makes them more susceptible to the action of EO, thereby reducing the exposure time. This can be clearly seen from Fig. 22.

B. Methods of Application of Fumigants and Their Dosages

Thompson (1966) has reported an exhaustive review (about one hundred references) of the properties and methods of application of methyl bromide (MeBr) as a fumigant. The important advantages and characteristics of MeBr are (1) ease of

TABLE LX EFFECT OF FUMIGATION ON COLOR, pH, AND BACTERIAL COUNT OF VARIOUS SPICES[a]

Spice	Lovibond tintometer color readings			pH	Moisture (%)	Bacterial count/g
	Red	Yellow	Blue			
Mace						
Unfumigated	8.0	16.0	3.4	4.8	1.0	23×10^4
Fumigated with 1000 g Etoxiat/cbm/6 hours	0.0	13.0	3.4	4.6	1.0	6.0×10^1
Fumigated with 1000 g Etox/cbm/6 hours	7.7	13.0	3.2	5.3	1.0	6.0×10^1
Fumigated with 1000 g Etoxiat/cbm/16 hours	8.0	15.0	3.1	4.8	1.0	6.0×10^1
Fumigated with 1000 g Etox/cbm/16 hours	8.0	10.0	3.1	5.3	1.0	6.0×10^1
Nutmeg						
Unfumigated	6.5	10.0	3.4	5.6	2.5	2.0×10^4
Fumigated with 1000 g Etoxiat/cbm/6 hours	6.5	10.0	4.0	5.6	2.5	2.0×10^1
Fumigated with 1000 g Etox/cbm/6 hours	6.2	8.0	3.2	6.1	2.5	2.0×10^1
Fumigated with 1000 g Etoxiat/cbm/16 hours	6.5	10.0	3.2	5.8	2.5	2.0×10^1
Fumigated with 1000 g Etox/cbm/16 hours	6.3	10.0	3.4	6.5	2.5	2.0×10^1
Ginger						
Unfumigated	2.0	4.6	1.2	6.8	3.5	5.5×10^6
Fumigated with 1000 g Etoxiat/cbm/6 hours	2.0	4.8	1.2	6.7	3.5	1.1×10^4
Fumigated with 1000 g Etox/cbm/6 hours	2.1	5.0	1.2	7.3	3.5	1.2×10^4
Fumigated with 1000 g Etoxiat/cbm/16 hours	2.1	5.1	1.0	6.8	3.5	1.6×10^4
Fumigated with 1000 g Etox/cbm/16 hours	2.5	5.0	1.4	7.8	3.5	3.6×10^3
Allspice						
Unfumigated	6.0	8.2	4.2	5.8	5.0	3.7×10^6
Fumigated with 1000 g Etoxiat/cbm/6 hours	6.0	8.2	4.2	6.5	5.0	4.0×10^1
Fumigated with 1000 g Etox/cbm/6 hours	6.0	8.0	4.2	7.0	5.0	4.0×10^1
Fumigated with 1000 g Etoxiat/cbm/16 hours	6.0	8.5	4.1	6.5	5.0	4.0×10^1
Fumigated with 1000 g Etox/cbm/16 hours	6.0	10.0	5.0	7.3	5.0	4.0×10^1
Paprika ground						
Unfumigated	11.0	11.9	1.5	5.5	6.5	1.6×10^6
Fumigated with 1000 g Etoxiat/cbm/6 hours	11.0	12.0	1.3	5.3	6.5	4.1×10^3
Fumigated with 1000 g Etox/cbm/6 hours	11.0	10.6	1.2	5.7	6.5	2.0×10^4
Fumigated with 1000 g Etoxiat/cbm/16 hours	11.0	12.0	1.3	5.5	6.5	9.8×10^3
Fumigated with 1000 g Etox/cbm/16 hours	11.3	10.7	1.3	5.8	6.5	8.7×10^3

(*continued*)

TABLE LX (*Continued*)

Spice	Lovibond tintometer color readings			pH	Moisture (%)	Bacterial count/g
	Red	Yellow	Blue			
Turmeric						
Unfumigated	4.5	30.0	0.5	6.9	4.2	2.3×10^6
Fumigated with 1000 g Etoxiat/cbm/6 hours	5.0	30.4	0.6	7.0	4.2	8.7×10^2
Fumigated with 1000 g Etox/cbm/6 hours	9.0	20.0	2.6	7.9	4.2	3.5×10^2
Fumigated with 1000 g Etoxiat/cbm/16 hours	5.2	31.0	0.4	7.2	4.2	1.4×10^2
Fumigated with 1000 g Etox/cbm/16 hours	9.2	10.6	1.6	8.2	4.2	1.0×10^2

[a] From Mayr and Suhr (1972).

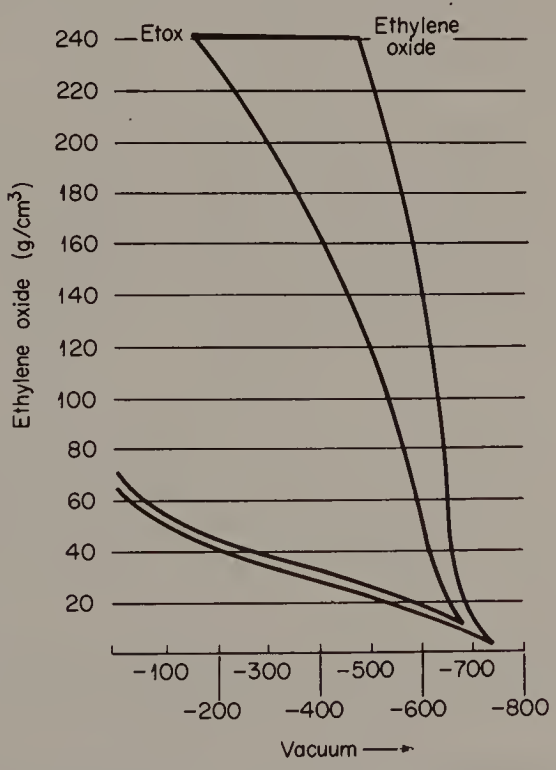


FIG. 22. Vacuum fumigation of spices with ethylene oxide and Etox. Upper and lower explosion limits of pure ethylene oxide and Etox (90% ethylene oxide + 10% CO_2), depending upon ethylene oxide concentration/vacuum. From Mayr and Suhr (1972).

penetration and airing; (2) no effect on most metals except aluminum; (3) low fammability; and (4) no odor at normal concentration. It can be used for most of the agricultural commodities, including spices. Its performance can be improved by mixing it with other fumigants such as EDB (Heseltine *et al.*, 1953; Majumder, 1962).

Fumigation with MeBr can be done successfully under almost all situations—under gasproof sheets, in ships' holds, in grain silos, in warehouses, and in stores fitted with device for recirculation of fumigant. It can also be done in atmospheric or vacuum fumigation chambers.

Phosphine or aluminum phosphide tablets that give off phosphine (PH_3) gas can also be employed. The exposure period should be no less than 7 days for a 100% kill of insects of all stages (Howe, 1972; Muthu and Majumder, 1974). A dosage of three tablets per ton of the commodity is sufficient. Rigid airtight conditions are a prerequisite for successful fumigations. As the permissible limit of PH_3 in air is 0.05 to 0.3 ppm (lower than that of most fumigants used), and as detection of the gas by smell is not desirable and the pressure of the gas is 38.5 atmospheres, making it difficult to be confined in an adequate manner, personnel who use aluminum phosphide should be properly protected with a special type of gas mask containing chemically treated carbons (Muthu *et al.*, 1974). Detector strips have been developed that can signal the threshold concentrations and can be worn as badges by operators (Muthu *et al.*, 1973a).

1. Fumigation of Small Packs of Spice Products

Fumigation of small packs of spice products is best accomplished in atmospheric or vacuum fumigation vaults. Vacuum fumigation is quicker and more efficient, although steel tanks that can withstand 1 atmosphere pressure are required. Methyl bromide is the fumigant of choice. A dosage of 32 g/m^3 with an exposure of 3 hours and an initial vacuum of 2.0 (absolute pressure) is recommended. A controlled leakage is advantageous and gives 100%. mortality throughout the commodity (Monro, 1969). Repeated vacuumization and restoration of atmospheric pressure, known as air washing, will remove residual gas. This residual gas can be recycled if two chambers are available. By increasing the exposure time and lowering the fumigant concentration, the same effective concentration–time (CT) product can be achieved. This gives economy in the use of the gas (Muthu *et al.*, 1961).

According to the Food and Agriculture Organization (1968) manual of fumigation for insect control in spices, the normal dosages and exposure times for five important fumigants are as shown in the tabulation.

2. Disinfestation of Curry Powder by Fumigation

a. Atmospheric Fumigation. Muthu *et al.* (1962) used two fumigants—methyl bromide and ethylene oxide. Organoleptic qualities were not affected by

Fumigant	Dosage	Exposure (hours)
Aluminum phosphide	45 tabs/1000 ft^3	72
Methyl bromide	16–24 g/m^3	24
Aluminum phosphide	45 tabs/1000 ft^3	72 (vac)
EDB–MB (1:1)	32 g/m^3	48
EO–CO_2 (1:9)	640 g/m^3	3

either of the fumigants. Application of methyl bromide resulted in residues up to 121 ppm at the dosage tried (48 mg/liter), which was within the permissible limits of 200 ppm prescribed under United States food laws. The flexible packages containing curry powders were found to be permeable to both the fumigants.

b. In-package Vacuum Fumigation. In-package vacuum fumigation was found to be a practical method for rapid disinfestation of curry powder on the production line.

c. Bulk Fumigation. Bulk fumigation in 5-foot columns was also attempted with both fumigants. Although ethylene oxide penetrated better than methyl bromide in the bulk of curry powder, insect mortality at the bottom of the column could not be obtained with an initial dose of 64 mg/liter. Bulk fumigation of curry powder does not seem to be feasible, since the high sorption at the top layers of the column results in a very low concentration at the bottom. Further studies are needed on this aspect (Misra, 1962; Muthu *et al.*, 1962; Pruthi, 1964).

d. Serial Fumigation. Serial fumigation of packaged curry powder indicated that, owing to high sorption of methyl bromide, the amount of the residual fumigant in the fumatorium after the exposure period could not be determined. Further studies are needed for the standardization of the serial fumigation technique for rapid disinfestation of curry powders (Misra, 1962; Muthu *et al.*, 1962; Pruthi, 1964).

3. Fumigation of Packaged Spice Products

The serial fumigation technique developed at the CFTRI, Mysore, consists in evacuation of fumigation chambers in series and carrying out sequential fumigations by employing the residual fumigant left in a fumatorium after the exposure period. It has been found that, with methyl bromide at a dosage of 48 mg/liter, three fumigations can be carried out, one after the other, each exposure period being terminated by assessing the required concentration–time product in each transfer. Data are presented on the fumigation of different types of commodities, on dosages, and on the resuse value of the residual fumigant concentrations. The closed system in which the entire operation is carried out and the economy in the use of the fumigant are the points to be considered for adopting this technique for packaged spice products (Muthu and Majumder, 1962).

4. Fumigation of Other Spices

The results on the effect of fumigation with an ethylene oxide and methyl formate mixture (1:1) show that, even at the lowest level of fumigant (64 mg/liter), there was complete elimination of coliforms and of yeasts and mold. Exposure to higher dosage of fumigant (96, 128, and 160 mg/liter) considerably reduced the total microbial load but did not bring about complete destruction. A higher degree of sterilization could not be obtained with fumigation, probably because it was carried out on sealed bags of polyethylene films. This might have acted as a barrier to the entry of microorganisms, but it did not affect the penetrability of the fumigant. This possibility needs to be checked by carrying out fumigation in unsealed plastic film bags (Krishnaswamy *et al.*, 1974a,b).

5. Prophylactic Measures

Fumigations offer no residual protective effect against insects. To achieve this kind of prophylaxis, mechanical treatment of jute bags or spraying the stacks with a residual insecticidal spray at 2 ml/ft^2 is recommended. The formulations based on lindane and DDT or malathion are available at the CFTRI. Fumigation treatment coupled with prophylactic measures will ensure insect-free storage for one full year (Majumder and Muthu, 1965).

C. Lethal Doses of Fumigants

The *lethal doses* of the different fumigants have also been determined (Lingdren *et al.*, 1954; Lingdren and Vincent, 1966; Food and Agriculture Organization, 1968). In decreasing order of toxicity (milligrams per liter) toward the adults, they are as follows: phosphine (1), HCN (4), acrylonitrile (15), chloropicrin (32), ethylene dibromide (38), methyl bromide (40), methyl iodide (52), ethylene oxide (78), carbon disulfide (371), and ethylene dichloride (768). It is essential that these concentrations and suitable times of exposure (concentration $\times$ time = CT) be maintained in the ambient intergranular atmosphere, where the insects reside, making due allowances for sorption of the gas by the commodities and leakage from the enclosure. Hence, the working dosages of the fumigants used (based on availability and individual requirements) in practical fumigations will be much higher, usually by a factor of 4 or 5. The aim of the fumigator is to achieve 100% control of all stages of the insects, without damaging the quality of the commodity treated (Muthu and Majumder, 1974).

D. Tolerance Limits for Residues

Fumigation with EDB, MB, or PH_3 will result in residues in the fumigated materials. Tolerances for these have been established by governments of different countries as well as by international organizations (WHO, FAO). With

EDB and MB, the permanent residues will be in the form of inorganic (water-soluble) bromide. The tolerance established for spices with EDB and MB (Food and Agriculture Organization/World Health Organization Joint Expert Committee, 1968) is 400 ppm. The residues obtained at CFTRI, Mysore, in some spices fumigated separately by EDB and MB are given in Table LXI. This shows that the tolerances are not exceeded in any of the spices treated. The Food and Agriculture Organization/World Health Organization Joint Expert Committee (1968) have prescribed tolerance limits for some spices. More work is needed in this area, with special reference to practical residue limits and acceptable daily intake in relation to the toxicological aspects for the remaining spices.

The residue tolerance of PH_3 is fixed at 0.1 ppm for raw produce and 0.01 ppm for processed foods (Monro, 1969). Preliminary work at CFTRI showed the following residues in turmeric and cardamom: With a dosage of 6 tabs per ton and an exposure period of 5 days, turmeric and cardamom showed 0.6 ppm of residue. As this exceeds the established tolerance, fumigation with PH_3 should be approached cautiously. In addition, the significance of the irreversible sorption of PH_3 by four spices (86 to 97%) needs serious examination (Muthu *et al.*, 1973b; Muthu and Majumder, 1974).

The Food and Agriculture Organization/World Health Organization Expert Committee (1968) have also clearly defined various terms such as pesticide residue, negligible residue, unintentional residue, practical residue limit, acceptable daily intake, temporary acceptable daily intake, tolerance, temporary tolerance, and good agricultural practice. They have also discussed methods of estimating tolerances, sampling for estimation of pesticide residue limits, analytical difficulties in total diet studies, significance of interactions of pesticides,

TABLE LXI RESIDUES OF ETHYLENE DIBROMIDE AND METHYL BROMIDE IN SPICES[a]

Material	Moisture (%)	Total inorganic bromide (ppm)	
		Ethylene dibromide	Methyl bromide
Coriander	9.3	15.1	16.9 (32 mg/liter)
Clove	8.3	14.3	23.0
Cumin	8.4	16.0	23.0
Ginger	12.0	16.9	24.7
Pepper	7.0	15.6	24.0
Turmeric	6.1	26.0	26.0
Coffee (plantation A)	—	22.0	—

Tolerance 400 ppm; dosage 6.4 mg/liter
Exposure periods in ethylene dibromide and methyl bromide were 96 and 48 hours, respectively

[a] From Muthu and Majumder (1974).

the interpretation of negligible residue, and future lines of work on pesticide residue evaluation.

Chadha (1973) has discussed the need, present status, and principles governing the establishment of tolerance limits for pesticide residues in foods (including spices). Dalra *et al.* (1973) have critically examined in detail (230 references) the significance of metabolites of insecticides in relation to their tolerance limits.

E. Methods of Residue Analysis

The Food and Agriculture Organization/World Health Organization Expert Committee (1968) have reviewed and briefly discussed the methods of residue analysis, a multidetection system of analysis, methods of fumigant residue analysis, etc., in a 242-page publication entitled "Evaluation of Some Pesticide Residues in Foods." Thornburg (1971) has published an exhaustive review of 424 papers published during just two years (November 1968 to October 1970) on the specific subject of pesticide residues, covering general analysis, GLC and other analytical techniques, chlorinated and organophosphorus pesticides, carbamates, herbicides, fungicides, and other pesticides. Proceedings of the symposium "Progress and Problems in Pesticide Residue Analysis" cover more than fifty papers on different facets of pesticide residues, residue analysis (technique, equipment, chemicals, etc.), and development of residue tolerances. A special background review paper (44 pages, 127 references) by Bindra and Kalra (1973) also covers development of methodology for pesticide residue analysis and the need for establishing simple, accurate, inexpensive, and easily adaptable procedures. Space does not permit a review of individual references on residue analysis; they are also covered in standard works on the subject.

F. Limitations of Fumigation Process

Chemical fumigation has several disadvantages: (1) It is not very effective against bacterial spores or insect eggs; (2) it must be done repeatedly; (3) it causes discoloration in some spices; (4) it produces off-flavors in some spices; (5) use of high concentrations of ethylene oxide results in dissipation and loss of essential oil; and (6) toxic residues of chemicals have also been reported (Wesley *et al.*, 1965; Lewis, 1974).

Under conditions suitable for effective fumigation with ethylene oxide or propylene oxide, these reagents can combine not only with moisture but also with chlorine from the natural inorganic chloride content of foodstuffs (including spices), thus forming the corresponding chlorohydrins. Concentrations of ethylene chlorohydrin up to about 1000 ppm were found in whole spices (cumin, etc.) and ground spice mixtures after commercial fumigation with ethylene oxide. These chlorohydrins are very toxic substances by all accounts. They are also sufficiently involatile and unreactive chemically to persist under food-

processing conditions. The proponents of this treatment should now establish tolerance limits for chlorohydrins.

Similarly, methyl bromide may also react with numerous sulfur-containing products to produce foul odors. However, this may not be the case with all other fumigants. Despite the above limitations, fumigation still continues to be the most important existing economical commercial practice for microbial decontamination and insect disinfestation of foods including spices.

G. Suggestions for Warehousing, Shipping, and Fumigation of Spices

To ensure freedom from insect and microbial infestation in spices during their warehousing, shipping, or transportation, the following measures are suggested:

1. Fumigation of the spices before storage is important, and also whitewashing of the godowns and prophylactic sprayings before the onset of the monsoon.
2. Treated pestproof bags should be used for storage of spices to prevent reinfestation in the godowns. Treatment of bags with pestproof emulsion or the stacks with Durobase will check cross-infestations. Fumigation with Durofume (EDB–MB, 1:1) at 32 g/m^3 for 48 hours will eliminate resident or residual infestations. The stocks of spices thus treated can be kept free of insects for one year.
3. Waterproof and lightproof packings may be used for susceptible spices. Polyethylene-lined double burlap bags or laminated pestproof bags are ideal for storage of spices if the moisture is reduced to a critical level before bagging. If the bags are loosely lined, there will be no difficulty in lifting them by cranes and loading them on ships when required during transshipment for export.
4. Spices should be examined periodically and fumigated again if necessary. Fumigation of the consignments before the onset of monsoon is also necessary. Old and fresh stocks should be stored separately to avoid cross-infestation and to protect against moisture absorption.
5. Even in spices that are free from infestation and are transported for export, instances of their damage by pests on arrival at the destination have been noticed. To prevent this, the ships' holds where the spices are to be stored should also be cleaned and fumigated before the consignment is loaded.
6. The fumigants used should be effective against the particular pests attacking the spices.
7. To avoid any toxic residues of the fumigants being left after fumigation, it is necessary to use the right type of fumigants in optimum quantities.
8. To avoid having the growers and merchants take their stocks to the godowns of private traders, and to ensure that the spices are scientifically stored and well looked after, modern warehouses should be set up at important trade centers.

12

Marketing, Quality Control, and Standardization of Spices

To a layman, "marketing" means merely buying and selling of agricultural produce. These activities look very simple, but when one inquires into the genesis of buying and selling, one comes across a multitude of economic phenomena relating to human wants and their satisfaction. These phenomena, in turn, lead to a plethora of economic activities that lie in the realm of concentration, equalization, and distribution. These activities include assembling, physical movement or transportation of goods, grading, packing, storing, processing, financing, and distributing. Market news or market intelligence is also now included in such functions. These activities are carried out at places known as markets according to certain practices and operational techniques.

Standardization forms an integral part of marketing. Standardization is the scientific yardstick that when properly used, leads to considerable improvement in quality, enhancement of productivity, reduction of costs, and optimum utilization of available resources. Quality standards are absolutely essential for any program of economic and industrial development in any country, whether developing or developed. Thus, standardization is a sine qua non for the development of a national economy (Pruthi, 1972).

The need for standardization in the sphere of agricultural marketing is all the more important, as marketing is an essential bridge between the producer and the consumer. Therefore, standardization and grading of agricultural produce (including spices) are expected to minimize the problems of long distances in national and international trading, to provide a means for intelligent comparison of prices according to quality or grade, and thus to promote greater confidence in

the minds of the consumer. Standardization also speeds up handling and distribution.

To promote orderly marketing, special attention must be paid to quality control and standardization in both manufactured articles and agricultural produce. The following advantages are achieved: (1) improved quality of the products; (2) increased output; (3) reduction in cost of production; (4) reduction in cost through saving in material; (5) reduced inspection cost by eliminating the need to inspect the contents of every package; (6) increased consumption and sales; (7) promotion of quality consciousness; (8) greater consumer satisfaction; and (9) more employment opportunities for technical and scientific personnel and others.

I. MARKETING OF SPICES

Spices, mostly of Asian origin, constituted the major merchandise of international trade in ancient times, when Arabs enjoyed a monopoly of this trade for centuries. The bulk of the Eastern spices reached Europe in the Middle Ages by way of Venice. There was a great demand in Europe for spices, chiefly pepper, needed both as a flavoring for food and as a preservative of meat. A Pepperers Guild was functioning in London in the twelfth century. The principal distribution center for oriental spices, however, remained Venice, which shared with the Arabs the wealth of commerce in spices (Majumdar, 1965). The fame of the Malabar coast of India and the East Indies as producers of quality spices was well established in Europe. The rest of the history of international marketing of spices is well covered by Parry (1953), and recent trends in the volume of world trade in spices have been covered briefly in the first chapter of this work. The available published information on marketing of spices has been summarized and is briefly reviewed below. Gandhi (1979) discussed marketing trends in spices.

A. Capsicum, Chilies, and Paprika

Marketing of chilies usually takes place through four kinds of markets in India, differing from one another with respect to place, quality of produce that is received for sale, market practices, tastes, purchasing power of wholesalers, etc. The markets can be broadly classified as follows:

1. *Village Bazaars* (markets) or village merchants.
2. *Primary Markets.* Generally situated near the producing centers, mostly at taluk or block headquarters. Many of them are regulated markets, where (1) sale is by open auction; (2) weighings and other ancillary functions in marketing are performed according to rules prescribed by the Regulated Market Committees, where all interests connected with marketing are represented; and (3) growers

need not wait for more than a day to realize sale proceeds. Unregulated markets, however, are not strictly governed by rules and regulations.

3. *Secondary Markets.* Either at District Headquarters or in a large town in a district, known for its advanced commercial activities. Like primary markets, they may be regulated or unregulated, the only difference being that transactions are on a more extensive scale, either between traders and growers directly or between traders from different places through the commission agents. Transactions are on a wholesale basis, and the commission agent is the key figure in marketing.

4. *Terminal Markets.* Large markets governed by traders themselves through their trade associations. The municipal authorities provide certain essential facilities such as water, light, and control over weights and measures. Sellers rarely come in direct contact with buyers. Terminal markets may also serve as re-exporting centers (Marketing Research Officer, 1968).

To sum up, the agencies involved in marketing at different levels are (1) growers, (2) village merchants, (3) commission agents, (4) wholesalers, and (5) growers' cooperative societies.

The major problems of marketing of chili relate to quality grading. These problems include (1) discoloration of chilies during excess rainfall and development of patches, (2) insufficient drying, leading to mold attack, etc., and (3) breaking of pods due to their brittleness at low moisture level, thereby releasing loose chili seeds, which lower the grade or even sometimes are responsible for the rejection of the entire consignment (Directorate of Marketing and Inspection, 1968b). However, these problems can be easily controlled by careful monitoring of moisture content. By drying chilies down to optimal moisture content and by moistureproof packaging, none of the above problems could arise. Narasimhaiah (1965) has described such a method of conditioning and packing of chilies for export, and the State Marketing Officer, Madras (1968), has outlined the present system of marketing, grading, and quality control for export of chilies.

The Spices Export Promotion Council, Cochin, India (1968a), has given an impressive coverage of production and marketing systems in the competing chili-exporting countries of the world—Japan, Thailand, Tunisia, Uganda, Pakistan, Ethiopia, China, Indonesia, Nigeria, and last, but not least, India, which until recently topped the list of countries exporting hot chilies. Spain, Yugoslavia, Bulgaria, Hungary, Czechoslavakia, and Rumania produce and export mild chilies and capsicums (commonly known as paprika and paprika powder). Apart from this, in almost every country, and notably in Africa and Asia, chilies are produced for local consumption.

In another informative and authoritative paper, the same council (Spices Export Promotion Council, Cochin, India, 1968b) states that the total world trade in hot chilies is of the order of 30,000 tons per year. The main countries importing

hot chilies are Ceylon (60%) and the United States (20%), which together account for over 80% of the total world imports of chilies. More than 95% of India's total exports of chilies were to Ceylon alone, but only recently the Sri Lanka Government has completely banned the imports of chilies into that country, with the object of boosting up its own production of chilies. India must now find better and more sophisticated markets in the West and also in the Middle East countries, 80% of the requirements of the latter being met only from Ethiopia. Gandi (1968) and Kanan (1968) have urged the diversification of exports for new markets and new products (oleoresins, chili powder, etc.). Sriraman (1970) and Raghubanshi (1971) have described the marketing system for chilies in Tamil Nadu and Saproon Valley (Himachal Pradesh), India, respectively. Earlier, the Directorate of Marketing and Inspection (1957), published an exhaustive and authoritative report on marketing of chilies.

B. Cardamom

India and Guatemala are the two major countries producing and exporting cardamom. However, published information on marketing is rather limited. The Directorate of Marketing and Inspection (1965a) has published an impressive and authoritative report on the marketing of cardamoms.

Raghavachari (1963) and Reddy (1965) have described some aspects of marketing of cardamom. Abraham (1965) and Thomas and John (1966) have described a modern smokehouse for cardamoms, as well as methods of curing, cleaning, sorting, grading, and preserving the natural green color of cardamoms. The Cardamom Board (India) and the Indian Institute of Foreign Trade (1967) jointly organized an impressive seminar on cardamom, as a result of which they crystallized the problems of export marketing, stabilization of prices of cardamom, which are known to fluctuate, at times violently, and diversification of exports of cardamom.

C. Cassia and Cinnamon

Although Ceylon cinnamon (*Cinnamomum zeylanicum*) is generally considered to be the best, cassia barks of different species from Southeast Asia are of considerable commercial importance, since the exports of cassia amount to five to six times those of Ceylon cinnamon. Of these, the important commercial types of cassia are Chinese cassia (*C. cassia*), Saigon cassia (four species found in Indochina), Batavia or Indonesian cassia, and Indian cassia.

Brown (1955) has described in some detail the production and marketing of cinnamon and cassia and their products. Lawrence (1967) has nicely reviewed some of the commercial aspects of cinnamon and cassia with special reference to types and commercial types, their nomenclature, and international trade.

Amarasingha (1968) has reviewed Ceylon's foreign trade, notably in cinnamon. The Directorate of Marketing and Inspection (1968a) also has published a report on the marketing of about twenty minor spices including cinnamon, Chinese cassia, and Indian cassia.

D. Clove

In Zanzibar and other countries where cloves are produced on a large scale, the bagged spice is carried from the growing areas to the towns and sold. In Zanzibar, the buying and selling prices of various grades of cloves are declared every year by the Clove Growers Association. The planters are sure, therefore, that they will get the approved price. The Association purchases cloves from growers and arranges to export the same after sorting them out in their own godown (Pillai, 1973). The best grade of cloves allows the presence of 2% w/w (max) of clove stems and "mother cloves," 2% of "headless" cloves and "Khoker" cloves, and no extraneous matter at all (International Organization for Standardization, 1974). The available published information on the marketing of cloves is scant; it has been briefly reviewed by the Food and Agriculture Organization (1962) and by Pillai (1973).

E. Ginger

World trade in ginger in recent years has amounted to 7 to 8 million dollars annually, of which India alone, being the largest producer, accounts for more than 50%. The other main ginger producers are Jamaica, Nigeria, Sierra Leone, China, and Taiwan. However, the ginger from different countries differs in quality, preparation, appearance, pungency, flavor, texture, and fiber content. On occasions, they are blended to suit certain purposes.

Ginger is marketed mainly as dry ginger—bleached or unbleached, garbled or ungarbled (cleaned or uncleaned)—which is considered a "spice" in the commerce. Ginger is also marketed as fresh ginger or preserved ginger—as candy or crystallized ginger, jam, pickles, sauce, and also in the bakery and confectionary trades. Added to that are its essence and oil, which are used in perfumery, and its oleoresin, which is used in the food and pharmaceutical industries (Tropical Products Institute, London, 1964).

Jamaican ginger is generally considered the best, with Indian (Cochin and Calicut) being the next best. Ginger from Sierra Leone is known as African ginger in the London trade. Nigeria has virtually replaced Sierra Leone in the ginger market. Chinese ginger has big hands, less fiber, less pungency, and less flavor and is used mostly in confectionary. Australia, too, has successfully grown ginger, which has made its appearance in the world market.

The major importers of Indian ginger are the United Kingdom, the Arab

countries (Aden, Saudi Arabia, and Egypt), and the United States. The Western countries generally prefer ginger from Jamaica and Sierra Leone.

The Directorate of Marketing and Inspection (1966a, 1970a), on the basis of their systematic field survey, has brought out publications on the marketing of ginger and quality control in ginger.

F. Onion and Garlic

Jones and Mann (1963) have covered in some detail the production, curing, storage, and marketing of onion and garlic. The Directorate of Marketing and Inspection (1966b) has also published a report on the marketing of onions. These reports cover the same pattern of information on all aspects of marketing, startig with harvesting of produce, assembling, distribution, method of sale, market charges, price spreads, demand and supply, packaging, transport, grading, storage, marketing costs, and margins. At the end, conclusions are drawn and recommendations offered to make marketing more dynamic and fruitful.

G. Pepper, Black and White

The Food and Agriculture Organization (FAO) and the U. S. Department of Agriculture compile and furnish data on production and world trade in pepper. During the period 1962–1967, the leading producing and exporting countries were as follows: India 31.4%, Indonesia 31.5%, Sarawak 15.9%, Ceylon (Sri Lanka) 9.7%, Cambodia 1.6%, Brazil 8.2%, Malagasy Republic (Madagascar) 1.7%.

There are a number of reports on different facets of marketing of pepper, both black and white. Thus, Brown and Reader (1953) cover cultivation and marketing; Narasimhaiah and Rangaswamy (1963) deal with quality control; Gandhi (1967) discusses pepper exports and export markets; Viswanathan (1967) urges the grading of black pepper at the level of the farmer through the growers' cooperatives; Paulose (1967) describes credit and marketing facilities; Arnold (1970) published a note on markets for pepper; Kay (1970) has published a fairly comprehensive account of the production and marketing of pepper; and the Directorate of Marketing and Inspection (1971) have brought out an impressive report on the marketing of pepper (all aspects).

Mehendran (1972), in describing the marketing of Sarawak pepper, states that the bulk of the Sarawak pepper is channeled through Singapore to terminal markets and consuming countries overseas. The links of the marketing chain in Singapore, together with their respective functions or role in trade, are also discussed. Attention is paid to marketing practices and procedures adopted at the various levels of the pepper-marketing chain, particularly with regard to grading, weighing, pricing, buying, and selling. Further, trading and financial relation-

ships between farmers and primary dealers and between the intermediaries at various levels, both in Sarawak and in Singapore, are mentioned. Finally, government intervention in the pepper trade, including brief details of the pepper-marketing scheme to be implemented in the country, is described. The objectives of the scheme, the manner in which it will be implemented, and the end results anticipated, especially with regard to buyers in terminal markets, are elucidated.

White pepper is produced from mature green pepper on a plantation scale. The conversion ratio of green to white pepper is 27%, and for green to black it is 31 to 33%. Mahendran (1972) also mentioned that, with the main objective of stabilizing international pepper prices, Sarawak has joined the Pepper Community, and his country has had fruitful meetings with other members of the Community. The minutes of the meetings are not available unless one has some connection with the FAO or ECAFE of the United Nations Organization.

Marongiu-Fortucci of the FAO, Rome (1972), presented an interesting paper depicting recent trends in world pepper economy. She stated that India, Indonesia, and Malaysia together supply more than 80% of the total world requirements, but increasing quantities now come from Brazil and the Malagasy Republic. Over the past twenty years, net world exports have more than doubled, and the pepper trade is now valued at $50 million (U.S.). This expansion reflects a recovery in Indonesia, formerly one of the largest producers, and the growth in Malaysia, which now accounts for about 30% of world exports compared to about 10% in the early fifties.

World production expanded rapidly during the latter part of the 1950s in response to high post-war prices, but output increased faster than consumption, and, by the end of the decade, prices had fallen sharply. Despite some recovery during the first part of the 1960s, prices continued to fluctuate sharply, and expanded shipments in 1967 and 1968, particularly from Indonesia, resulted in a considerable fall in prices. However, by the end of 1969, prices began to rise as a result of reports of much smaller crops in Indonesia. In 1970, shipments from that country were the lowest in twenty years!

The Pepper Community, established in 1970 by the major producing countries—India, Indonesia, and Malaysia (although open to all producing countries)—aims at coordinating researches on production, development of trade, and elimination of price fluctuations of the commodity. In the meanwhile, studies on the economic problems of pepper are continuing under the auspices of the FAO committee on commodity problems.

H. Pimento Berry or Allspice

Pimento is popularly known as allspice, since it resembles in taste and aroma a mixture of cinnamon, clove, and nutmeg. World trade in pimento is limited to $3 to $4 million (U.S.) annually. It is grown in the Caribbean area of the West

Indies, Jamaica being the leading producer and exporter. Pimento of inferior quality is also produced in Mexico. The decline in pimento exports since World War II has partly been compensated by a considerable increase in pimento leaf oil trade. The United Kingdom, Sweden, The Netherlands, and West Germany are the main importers of pimento. Pimento trade statistics are incomplete and confused by the inclusion of data on various capsicums or pimiento (Food and Agriculture Organization, 1962).

Ward (1961) has published an interesting pamphlet on pimento covering all aspects of production, marketing, economy, trade, etc. The Jamican government is the sole exporter of pimento berries. It fixes the price that growers receive from the purchasing agents, who are obliged to deliver all the pimento to Government's Clearing House, where it is cleaned, bagged, and stored before export. Sales abroad are made through commercial channels under government supervision, and distributing agents are appointed in the principal importing countries. Trade in pimento berries has been long established.

I. Other Spices

Rowan (1942) reported on the cultivation and marketing of cardamom, cinnamon, cloves, capsicum, ginger, nutmeg, pepper, turmeric, and vanilla. The Food and Agriculture Organization (1962) published a monograph on nine spices covering trends in the world market. Pepper, clove, nutmeg, mace, cardamom, cinnamon, cassia, ginger, and pimento together account for 80% of the world trade in spices. Three spices—pepper, cloves, and nutmeg—together constitute about 60%. The Directorate of Marketing and Inspection (1965b), brought out an exhaustive report on marketing of turmeric. Majumdar (1965) discussed the role of spices in the export trade of India. The Directorate of Marketing and Inspection (1968a) published another report on the marketing of eighteen minor spices—aniseed, biship's weed, caraway, Chinese cassia, Indian cassia, celery seeds, cinnamon, coriander, dill, fennel, fenugreek, garlic, kokum, nutmeg, mace, pomegranate seed, saffron, and vanilla. The Commonwealth Secretariat (1969) has brought out a publication on plantation crops, including spices, their production, and international trade. Breckenridge (1969) discussed the market for spices in the United States, and Jensen (1971) described the marketing of two varieties of vanilla beans, *Vanilla plantifolia* and *V. tahitenses*. See also Manning (1969).

Wands (1972) reported an interesting survey of the American spice market. He stated that a survey of spice-buying habits of consumers in the United States has shown that, with the growing use of convenience foods, the role of spices has increased in importance and has led to an increase in the per capita consumption of spices in the United States from 18.4 to 28.8 ounces per year during the last decade. This has led to considerable changes in the marketing of spices and food products in the United States and has also caused a broadening of the definition

of spices for marketing purposes to include any product of plant origin that is used primarily to season food; for example, dehydrated vegetable seasonings including blends and more than fifty spice products are found in food stores. Industrial buyers have a choice of more than one hundred products!

In the United States spice processing firms now offer a wide spectrum of customer services ranging from advice on spices to the development of new food products using spices. This has grown up largely as an offshoot of the greater technical capability that spice companies have acquired to meet the increasing stringent quality control requirements of customers. Another development has been the implementation of an import and domestic inspection program. Quality analysis is conducted on samples in independent laboratories recognized as competent by the American Spice Trade Association (ASTA) and the U. S. Food and Drug Administration. The role of the ASTA in promoting the sales of spices and in developing new spice research programs has been described (Wands, 1972). Venketachalam and Gray (1974) discussed marketing of spices and spice products.

J. Ground Spices

Published information on the marketing of ground spices is rather scant. The Directorate of Marketing and Inspection (1970a) has issued a publication describing the grading and marking of ground spices such as chili powder, coriander powder, turmeric fingers, bulbs, and powder (fine and coarse ground), Malabar pepper (garbled and ungarbled), light pepper, Tellichery garbled pepper (ground), and curry powder.

K. Spice Products

Allen (1970) has published an interesting account of the structure of marketing of essential oils in overseas countries, and Cripps (1972) has discussed the process, the market, and the future of spice oleoresins. He has briefly described the various steps in the production process, with emphasis on the role marketing should play in influencing the processing techniques. Three aspects of the current status of the oleoresin market—how it is measured, reached, and influenced—are discussed in a general way, with special consideration given to the role of the food technologist in the emergence of new markets for spice oleoresins. Questions relating to the future of oleoresins—where and how will future markets develop, where will oleoresins be produced, and who will control production—are also discussed in general terms with special emphasis on the economics involved. The International Trade Centre, Geneva (1974, 1977a,b) has published an impressive publication entitled "Markets for Selected Essential Oils and Oleoresin" and "An Exhaustive Survey of the Selected World Markets for Spices" in 2 volumes.

II. QUALITY CONTROL AND STANDARDIZATION

A. Standardization at the International Level

At the international level, the two organizations actively engaged in the formulation of quality standards relating to methods of sampling and tests for spices and their products are the International Organization for Standardization (ISO) and the Joint FAO/WHO Food Standards Programme. For the formulation of uniform international standards for spices, spice products, and methods of sampling and test, the Indian Standard Institution undertook the responsibility of holding the Secretariat of ISO/TC34/SC7 Spices and Condiments Sub-Committee (SC7) of the Technical Committee on Agricultural Food Products (TC34) of the International Organization for Standardization (ISO). The twenty-eight countries represented on this sub-committee are Australia, Belgium, Bulgaria, Canada, Cuba, Czechoslovakia, Ethiopia, Egypt, France, Germany, Hungary, India, Ireland, Israel, Italy, Lebanon, The Netherlands, Poland, Portugal, the Republic of South Africa, Rumania, Sri Lanka, Sweden, the United Kingdom, the United States, the USSR, Venezuela, and Yugoslavia. This sub-committee generally meets at intervals of 18 months to consider important problems concerning the standardization of spices and condiments.

Work already completed by ISO/TC34/SC7 Spices and Condiments (up to February 1977) on the formulation of thirty-four international standards on (1) nomenclature, (2) methods of sampling, (3) methods of test, and (4) individual spices and condiments are briefly summarized in the accompanying tabulation.

ISO Specification/ Document number	Short title
	(A) Nomenclature
ISO/R 676-1968	Nomenclature, First List
	(B) Methods of Sampling
ISO/R 948-1969	Methods of Sampling
	(C) Methods of Test
ISO/R 927-1969	Determination of Extraneous Matter
ISO/R 928-1969	Determination of Total Ash
ISO/R 929-1969	Determination of Water-Insoluble Ash
ISO/R 930-1969	Determination of Acid-Insoluble Ash
ISO/R 939-1969	Determination of Moisture Content
ISO/R 940-1969	Determination of Alcohol-Soluble Extract
ISO/R 941-1969	Determination of Cold Water-Soluble Extract
ISO/R 1108-1969	Determination of Nonvolatile Ether Extract
ISO 2825-1974	Preparation of a ground sample for analysis
DIS 3513	Determination of Scoville index in chillies

(*continued*)

ISO Specification/ Document number	Short title
DIS 3588	Determination of degree of fineness of grinding by hand sieving
DP 5564	Determination of Piperine Content of Black and White Pepper
DP 5566	Determination of Coloring Power of Turmeric
DP 5567	Determination of Volatile Organic Sulfur Compound in Dehydrated Garlic
	(D) Specifications for Individual Spices and Condiments
ISO/R 882-1968	Cardamom
ISO/R 959-1969	Pepper, black and white, whole and ground
ISO/R 972-1969	Chillies, whole and ground
ISO/R 973-1969	Pimento (allspice), whole and ground
ISO/R 1003-1969	Ginger, whole, in pieces and ground
ISO 1237-1974	Mustard seed
ISO 2253-1974	Curry powder
DIS 2254	Cloves, whole and ground
DIS 2255	Coriander, whole and ground
DIS 2260	Cinnamon and Cassia
DIS 3632	Saffron
DIS 2256	Spearmint, dehydrated
DP 5563	Peppermint Dried
DP 5562	Turmeric, whole and ground
DP 5561	Caraway seed
DP 5559	Dehydrated onion
DP 5560	Dehydrated garlic
DP 5565	*Vanilla fragrans* (Salisbury)

Basically, these standards, national or international, help us to better understand the market, for they establish the requirements of purchasers and also define quality in quantities that can be measured objectively in the quality control testing laboratories. Especially for spices, the concept of quality differs from purchaser to purchaser, particularly on the international level. For instance, to Indian purchasers, quality of spices might mean cleanliness or freedom for extraneous matter, attractive large size or small size, or particular varieties that possess the maximum quantity of aromatic and flavoring constituents characteristic of the spice. But to the overseas buyers, quality might also mean quantitative expression of flavoring constituents by lots, safe levels of moisture, freedom from insecticides, etc. It is from this broad spectrum of quality that different buyers tend to choose the quality in the light of their own specific requirements. For the integration of such different needs, commonly acceptable international standards provide the answer.

B. Standardization at the National Level

According to the readily available information, there are over thirty national standards societies, in addition to the two international organizations mentioned above. Among these, the following are the national organizations that deal especially with specifications for spices and spice derivatives:

1. American Spice Trade Association (ASTA)
2. Association Française de Normalisation (AFNOR)
3. Association of Official Analytical Chemists of the United States (AOAC)
4. British Standards Institution (BSI)
5. Canadian Food and Drug Directorate
6. Central Committee for Food Standards, New Delhi (CCFS)
7. Ceylon Standards Bureau
8. Deutches Normanauschuss, Berlin
9. Directorate of Marketing and Inspection, India (DMI)
10. Essential Oil Association of the United States (EOA)
11. Hungarian Office for Standards
12. Indian Standards Institution, New Delhi (ISI)
13. The Netherlands Normalizative Institute
14. Romanian Standards Organization
15. U. S. Food and Drug Administration (FDA)
16. U. S. Military Standards (EE-SO645b and EE-S-631 F)

Space does not permit even a sampling of the numerous specifications formulated by them for individual spices and their methods of sampling and test. Suffice it to say that the ASTA, the DMI, and the ISI have taken a leading role in the formulation of specifications for spices, etc. The American Spice Trade Association (1968), in active cooperation with the AOAC, has brought out an excellent compilation of a comprehensive list of twenty-two general and specific standard (official) methods of test for spices based on collaborative analytical research programs. The AOAC has played a key role in organizing collaborative analytical research programs, thereby actively assisting the standard formulating and implementing organizations on national and international levels. Its standard publication, "AOAC Methods of Analysis," covers analytical methods for spices and condiments. After basic and collaborative programs have been conducted, this reference book is revised and brought up to date every five years by incorporating new, commonly accepted methods. Useful work on standardization of spices and their derivatives has also been done and is in progress in the other organizations in different countries. The ISI has brought out over thirty-six specifications on spices and their methods of test. The ISO has arranged col-

laborative analytical research programs in order to establish commonly acceptable analytical methods after thrashing out differences in points of view of member countries. The author had the privilege of serving as a coordinator for an international collaborative research program on screening of analytical techniques for the determination of piperine and pungent principles in black and white pepper (Pruthi, 1969b,c,d, 1970a,b), and he participated in another program on the determination of capsaicin (pungency) and color (capsanthin and capsorubin) in ground Hungarian paprika (Pruthi, 1968b, 1969a, 1970c).

C. Microbiological Quality Standards for Spices

Importers of spices have become highly quality conscious and are stipulating rigid microbiological standards for spices—as, for instance, for dehydrated onion and garlic, even though both onion and garlic are antiseptic and garlic is antimicrobial too. The need for microbiological quality standards is acute, as such standards encourage both production and development of trade on both national and international levels. The available microbiological quality standards established by the ISO and by some countries are a valuable guide, but standards for a particular country should be determined essentially on the basis of the actual data collected within that particular country. In a limited way, some studies on the extent of microbial contamination have been conducted in India (Misra, 1962; Rao *et al.*, 1962; Pruthi, 1964, 1974; Krishnaswamy *et al.*, 1974a,b); they have been reviewed in Chapter 10, along with the work done in other countries. Natarajan and Shankaracharya (1972) have also stressed the need for formulation of microbiological quality standards for foods, including spices.

D. Quality Standards for Spice Products

The quality standards for whole and ground spices have already been reviewed and discussed. Pruthi and Misra (1962a) published a comprehensive paper, "Quality Standards for Curry Powders and Other Spice Mixtures," based on first-hand data collected by them. The ISO/TC54 is concerned with the formulation of standards for essential oils. The EOA has made significant contributions in the formulation of standards for essential oils and oleoresins. Several other national bodies have been fairly active in this field, and a number of quality standards have been set up for individual spice essential oils as well as for the precise methods of their physicochemical and sensory evaluation.

Stahl (1972) has reviewed and discussed practically all the important aspects of standardization of oleoresins; he has complained that, although the standards for spices are quite numerous, there are relatively few for oleoresins, as illustrated in the accompanying tabulation.

Source of publication of specifications	Number of specifications	
	Spices	Oleoresins
Essential Oil Association (EOA)	0	9
U.S. Military Standards (MIL)	29	19[a]
British Standards Institution (BSI)	5	0
Indian Standards Institution (ISI)	36	2
International Organization for Standardization (ISO)	32	0

[a] Soluble spices incorporating oleoresins for the most part.

It is interesting to note that the EOA have laid down specifications for oleoresins of black pepper, red pepper, capsicum, paprika, celery seed, ginger, turmeric, clove, and mace. Other individual primary oleoresin manufacturers have their own "in-house" specifications for those oleoresins not covered by the EOA.

There is a multiplicity of methods of analysis in the spice trade. The available analytical methods for oleoresins laid down by different national and international organizations are summed up in Table LXII. They are considered to be useful in the evaluation of quality of some oleoresins. Other methods of oleoresin evaluation are reviewed in Chapter 6.

E. Some Problems of International and National Standardization

1. One of the problems that confronts a user of spices, oleoresins, and essential oils is that of finding "outside" laboratories that are qualified to do "analytical referee" work.

2. There are reported to be interlaboratory variations of 30 to 50% in results of analysis. Stahl (1972) has cited specific instances of estimation of essential oil in cinnamon and cassia in the United States. Thus, basic faults sometimes lie not in the methods per se, but with the workers. This aspect needs very careful consideration—namely, testing the reproducibility as well as the repeatability of results obtained by each worker in each laboratory.

3. Stahl (1972) has also cited intersting and even greater variations in the results of the Scoville test for capsaicin (pungency) on eight varieties of capsicum obtained by three laboratories (Table LXIII), in comparison with the objective (spectrophotometric) method. Since 1946, thirty-five papers have been published (see Chapter 3 under Capsaicin), each claiming a new or an improved method. Likewise, there are thirteen methods for determination of piperine (Pruthi, 1969b,c,d, 1970a,b). Hence, collaborative analytical research programs are needed to screen these methods with the object of selecting the most commonly acceptable one to be adopted uniformly by all national and international bodies, on the lines of AOAC, ISO, ASTA, etc. Otherwise, there will be no end to arguments over conflicting results, and correspondence and litigation at the ex-

TABLE LXII ANALYTICAL METHODS AVAILABLE FOR SPICE OLEORESINS[a]

	Standard					
	ISO	BSI	EOA	ASTA	AOAC	Nonstandard
Alcohol extract	+	+	+	+	+	
Nonvolatile ether extract		+		+		
Nonvolatile methylene chloride extract				+		
Volatile and nonvolatile ether extract					+	
Volatile oil	In preparation	In preparation	+	+	+	
Color power of turmeric			+	+		
Extractable color in paprika			+	+		
Capsaicin (instrumental)						+
Pungency in capsaicin (Scoville heat test)			+	+	+	
Piperine in black pepper (Kjeldahl nitrogen)				+		
Piperine in black pepper (colorimetric)				+		+ Colorimetric and Spectrophotometric
Nitrogen in nonvolatile ether extract					+	
Moisture	+	+	+	+	+	
Solubility						+
Residual solvent						+ Modified Tod GC method
Optical rotation (on volatile oil)			+		+	
Refractive index (on volatile oil)			+		+	

[a] From Stahl (1972).

pense of considerable time, energy, and money on both sides (the buyer and the seller).

4. The major problem in standardization is that of alignment of national and international standards. The urgency for this is best highlighted by projecting just one of the confusions that exist in the measurement of color in capsicum and paprika oleoresin. At various places in the literature ten different color values are cited: (1) ASTA color value, (2) EOA unites, (3) conventional color units of LaWall and Harrison, (4) standard color units of the Mayonnaise Manufacturers Association, (5) Guenther units (Nessler tubes), (6) Gentry units, (7) Lovibond red units, (8) reflectance values, and (9) the Benedek method (ISO).

Much of the confusion could be eliminated if an acceptable method were adopted by all—the exporting as well as the importing countries, the buyers and

TABLE LXIII COMPARATIVE RESULTS OF THE SCOVILLE TEST FOR CAPSAICIN IN CAPSICUMS[a]

Variety	Scoville heat units (thousands)			
	Lab. I	Lab. II	Lab. III	Spectrophotometric method (Lab. III) (heat units)
Cayenne Long Slim	45	19	28	26
College 64L	26	—	7	9
Anaheim	14	—	22	22
Santaka	60	29	55	64
Turkish	14	9	8	12
Hontaka	80	28	55	69
Long Thin Cayenne	14	1	8	12
Carolina Hot	50	17	50	47

[a] From Stahl (1972).

sellers alike. The ASTA, AOAC, and ISO could take the initiative in matters that involve controversy, litigation, etc.

5. In prescribing legal limits for quality characteristics and residue limits, prevailing practical aspects must not be ignored. The specifications must be reasonable and able to be met without difficulty.

6. Grade designations based on established trade practices within a country should be confined to national standards only, unless the same grades have been accepted internationally, in which case they could be incorporated in the international standards.

7. Grade designations should be precise and not confusing. For instance, Fair Average Quality (FAQ) may be interpreted in some countries to mean "ungraded spices," whereas in other countries it means "average between the best and the lowest grades." Therefore, all grades should be numbered I, II, or III, and, where trade practices are well established, these numerical grades could be supplemented by the relevant grade designations.

8. At the international level, sophisticated chemical methods of analysis of spices have been debated for years—for example, carvone content of peppermint, total volatile organic sulfur compounds in dehydrated garlic, piperine in pepper, capsaicin in capsicums, chromatographic methods for pigments in saffron, and the Benedek method for color in paprika. Their validity should be established wherever necessary, and national standards should keep pace with this activity.

9. There is also need for microbiological standards especially for the importing countries in the larger interests of human health, irrespective of the country of origin.

10. There is also an urgent need for simple, rapid, reliable methods—for

example, for lead and aflatoxin—and also for quick field methods of quality evaluation of spices and spice products.

Moudgil (1958) critically compared the standard methods of test for essential oils prescribed by various countries with those of ISO/TC54 and stated that the ISO provides an excellent platform where the member countries may discuss their respective national viewpoints. Such discussions give an instructive insight into the uniformity that prevails within an apparent international diversity.

Pruthi (1966, 1968a, 1969a–d, 1970a,b,d, 1974) has highlighted other problems and has offered some solutions. Hari Bhagwan and Mathur (1974) have also dealt with the subject in fair detail.

F. International (ISO) Specifications for Spice Essential Oils

As of late 1979 the Technical Committee ISO/TC 54 has formulated the following international standards for the following important spice essential oils, in addition to those for about 20 methods of tests for essential oils.

ISO Specification/ Document number	Title of standard	Year published
ISO 3475	Oil of aniseed	1975
ISO 3045	Oil of bay	1974
ISO 3216	Oil of cassia	1974
ISO 3760	Oil of celery seed	1979
ISO 3524	Oil of cinnamon leaf	1977
ISO 3142	Oil of clove bud	1974
ISO 3141	Oil of clove leaf	1975
ISO 3143	Oil of clove stem	1975
ISO 3215	Oil of nutmeg	1974
ISO 3527	Oil of parsley fruit	1975
ISO/R 856	Oil of peppermint, France, Italy, United Kingdom, and the United States	1968
ISO 3043	Oil of pimento berry	1975
ISO/R 1342	Oil of rosemary	1971
ISO 3526	Oil of Spanish sage	1976
ISO 3033	Oil of spearmint	1975
	Draft International Standards (ISO)	
DIS 4733	Oil of cardamom	
DIS 3516	Oil of coriander	
DIS 4734	Oil of mace	
DIS 3061.3	Oil of black pepper	
DP 4729	Oil of pimento leaf	

13

Research Needs

It is clear that spices play an important role in international trade. One of the major obstacles to a satisfactory analysis of the economics of spices is the lack of statistical information on most sectors of the industry. Production figures are sometimes lacking even for major producers of important spices. Time series are often incomplete. Very little is known about stocks. Spices are frequently shown separately in export statistics. The Standard International Trade Classification provides only two headings or groupings for spices. Such groupings may be adequate for analyzing general trade problems, but they are of little use in studying the market for a particular spice. Since trade statistics are usually the only source of information on consumption in importing countries, deficiencies in them make it difficult to analyze demand. There is, therefore, an urgent need for comprehensive statistics, to be maintained by an international agency, on production, internal consumption, mean consumption per capita, exports (amount and value), etc.

Although the literature on various aspects of some major spices of international importance is voluminous, further research is necessary on some of the important areas mentioned below:

1. *Antioxygenic Aspects*. The exact mechanism of the antioxygenic properties of spices—their precise mode of action, and the actual nature of their contribution in exerting a preservative action in some foods—needs further elucidation.
2. *Antibacterial Activity*. Investigations should be made into the precise mode of action of volatile constituents and spice essential oils vis-à-vis the relation between their chemical structure and antibacterial activity.

3. *Medicinal Aspects.* In Chapter 2 it has been clearly shown that most of the spices that are being used in the human dietary possess different physiological and medicinal properties. Very little information is available on the exact mechanism of their action. Some of them are antibacterial. This obviously suggests that they may exert their beneficial effect by suppressing the growth of harmful intestinal bacteria, but, considering the quantity of the spices consumed, the medicinal effects they exert on the human system seem to be out of proportion to their antibacterial properties. It appears, therefore, that there is an alternative mechanism of action by which the spices act as therapeutic agents. Further investigations are necessary to explore this important aspect. The German and Indian groups of workers have made a good start in carrying out a series of research investigations on the physiological and medicinal aspects of some important spices commonly used in flavoring of their food, but there is still considerable scope for intensification of researches in several areas, such as the following:

a. Prior to 1955 physicians had warned patients with peptic ulcers to avoid spices and highly seasoned foods, without presenting any scientific evidence to support their recommendation. This approach needs thorough study. Some attention has been paid by a few scientific workers to paprika, pepper, and mustard only. Other important spices require similar study. The proposed concept of a defensive mechanism—the gastric-mucoso barrier—also should be investigated further.

b. Extracts and preparations from spices such as coriander have been reported to improve the utilization of glucose, as shown by glucose tolerance tests. These preparations should be tested on human subjects with *Diabetes mellitus.*

c. The "circular activity" of other spices not investigated so far may have to be examined. It will be necessary to determine whether "spice effects" depend on the composition of the basic food. In any case, it is possible, in principle, to effect by use of spices the required changes that cannot be produced by any of the known medicaments.

d. The diuretic effect of extracts of caraway, anise, and sweet fennel seeds has been reported. Similar research should be done on other spices.

e. Further studies should be initiated to elucidate the nature of the "active principle" in parsley that has been reported to have a tonic effect on uterine musculature.

f. Garlic causes clinical improvement in the treatment of patients with leprosy. However, its mode of action is not known yet. It is not clear whether the curative property of garlic is due to its antibiotic principle, allicin, or to the allithiamine formed as a result of allicin's having combined with thiamine, or to the "thiamine status" of the patient. These aspects need further investigation.

g. Further studies should also be made on the effect of spices on the adrenocortical functioning.

h. Onion is beneficial to everybody, irrespective of age, as an effective agent

for preventing atherosclerosis in patients with high blood pressure and violations of the secretary function of the intestines. Similar studies are needed on other species of *Allium* and other spices.

i. The precise mode of action of spice essential oils and their aromatic chemicals in perfumes as fungicides, insecticides, and pesticides needs further study.

j. Similarly, the precise role of bacterial and/or fungal enzyme systems in affording protection to the skin by spice essential oils and aromatics against microbial invaders should be thoroughly investigated.

4. *Sensory Evaluation of Spices and Spice Products*. Although sensory evaluation has emerged as an important tool in assessing the quality or flavor of foods, including spices, a major difficulty still remains in the precise measurement of sensory responses. Reliable data on sensory evaluation of spices in particular are rather scanty, and we do not have adequate information from which to draw meaningful conclusions. There is an urgent need for the development of a correlation between the objective and subjective methods of flavor evaluation of different spices—whole, ground, liquid, and "instant" spices, as well as spice mixtures, etc.

A number of workers have stressed the need for Anchored Sensory Evaluation Tests, which could be used in grading spices and in establishing the thresholds of individual grades. In fact, grade limits anchored in terms of physicochemical attributes can be considered as the sine qua non of modern quality control of foods in general and of spices in particular.

5. *Analytical Techniques and Instrumentation*. (*a*) There is a great need for the application of specific, simple, analytical techniques such as TLC and colorimetry, including the determination of characteristic fluorescence of spices. More data are needed on the content of copper, lead, arsenic, and other heavy metals in spices. There is also an urgent need for the development of rapid and reliable techniques for the determination of lead and farinaceous matter in spices and spice mixtures, the determination of pungency in garlic and onion, etc.

b. Systematic GLC studies on a number of spices have been reported, but several other spices have yet to be studied. Among them are asafetida, compounded asafetida, saffron spice mixtures, and new spice products, such as liquid spices, instant spices, and encapsulated spices. The knowledge thus gained may be used for (1) the determination of the geographical origin of spices and spice essential oils; (2) the detection of adulteration; (3) the study of seasonal variations in the quality of spices; (4) the study of the changes and mechanism of deterioration during storage of spices and spice products; (5) an understanding of the biogenesis of the various components thereof; and (6) recognition of the genetic variations among plants and making the best use of such variations in developing new varieties of spices of the desired characteristics.

c. There is also a need for the application of TLC and other simple analytical techniques to study the above aspects of spices.

d. Simple, rapid, reliable, and inexpensive analytical techniques should be

developed for the determination of active principles in spices, such as piperine in black and white pepper, capsaicin in red pepper (capsicum), pungent principles in celery, garlic, ginger, and onions, spice oleoresins, coloring power and pigments in red pepper, saffron, and turmeric, phenols in nutmeg and mace, and sulfur compounds in different spices such as asafetida, mustard, etc.

e. Similar techniques should be developed for detection of different adulterants in ground spices and spice mixtures, liquid spices, instant spices, etc.

f. So far only one good paper has been published on trace elements in some spices. Such useful information should be of specific interest to dieticians in formulating low-sodium diets and to geneticists as well for developing better varieties of spices of desired characteristics.

6. *Agrohorticultural Aspects.* There is a need to identify important problems in production, postharvest technology, processing technology, standardization, and marketing, to devise ways to tackle them, and to evolve a new strategy of marketing, including locations of foreign markets for both raw and finished products at reasonable competitive international prices. Some of these needs are highlighted below for the attention of agriculturists, horticulturists, scientists, technologists, and others concerned in their respective spheres pertaining to spices.

a. Development of new varieties. No successful technological innovation or improvement is possible without the assurance of a steady and regular supply of high-quality raw material. This calls for increased research into the agricultural and horticultural aspects of various spice crops that have not received the attention they deserve. Development of newer, high-yielding and disease-resistant varieties and study of their manurial requirements and other agrohorticultural aspects should be encouraged and intensified. Genetic manipulation to obtain crops with specialized characteristics, such as spices with improved flavor retention and oleoresin content, is one of the prime areas where agricultural research should be initiated. It is also necessary to determine the precise degree of maturity at which spices should be harvested for processing to obtain desired qualities in terms of flavor and pungency in the final products (dry spice, spice powder, oleoresin, spice oils, and other products).

b. Improvement in active principles and yield. The modern techniques of plant breeding, mutation, and cultivation that have proved successful with cereals and other crops have yet to be applied seriously to spice plants. These activities should be intensified at the agricultural universities, wherever feasible. This work may be comparatively easy with respect to annuals such as celery, chilies, coriander, cumin, fennel, fenugreek, garlic, ginger, onion, and turmeric. However, with perennials such as cardamom, cassia, nutmeg, and pepper, the work is bound to be slow, requiring an extensive time-lag to evaluate the results. The main objective is, naturally, to improve the content of active principles in the final product as well as to increase the total yield per plant in terms of the fruit, seed, rhizome, or bark.

c. Intensification of research and development. Considering the impact that spices have on the economy of several spice-producing countries and the magnitude of the problems confronting spice production and processing, the present efforts in spice research are totally inadequate. At least 1% of the value of the total spice production should be designated for this purpose.

d. Reduction of varieties. It has been observed that a number of varieties of important spices such as ginger, pepper, and turmeric are under cultivation in several countries irrespective of their lower yielding capacity or quality characteristics. Through research and extension work, the number of varieties should be reduced to the absolute minimum, keeping in view the known market quality requirements and yield, in order to maintain effective control over the quality of produce for internal and international trade.

e. Planned production and marketing. Owing to lack of planned production and marketing of various spices, particularly cardamom, ginger, pepper, and turmeric, there are sometimes violent fluctuations in the prices, which adversely affect the spice industry, especially in producing countries. An effort should be made to have planned production and marketing of different spices.

f. Large-scale cultivation of tree spices. Some of the spices, such as nutmeg, cassia, cinnamon, cloves, and allspice, are in great demand and in short supply. Cultivation of these spices on a large scale should be encouraged wherever possible. The soil profiles, cultivation techniques, and climatic conditions for growing these spices should be studied and the results made available to interested spice growers the world over.

7. *Postharvest Technology.* After harvest, most spices are subjected to different types of unit operations. Some of the postharvest techniques of curing and bleaching of spices can be standardized by using scientifically controlled procedures to obtain uniformity in product batches. At the same time, there is a need to guard against the indiscriminate use of such chemicals.

a. Curing houses. To avoid loss of volatile oil and to arrest microbial and insect infestations in spices, it is preferable to have "curing houses" located in the producing centers to make use of the improved techniques of processing (curing, cleaning, grading) for the greater benefit of the growers. Such curing houses should also provide facilities for specialized techniques such as abrasive peeling of ginger rhizomes and their subsequent mechanical drying, boiling of turmeric at controlled temperature, alkali treatment and stove drying of cardamom, and reducing the moisture content of spices by mechanical drying to critical levels.

b. Optimization of pretreatments and drying conditions. On the technological side, there is need for optimization of drying conditions and standardization of the necessary pretreatments for different spices, keeping in mind the final quality of the product. It may be more advantagenous to use mechanical drying and also to use mechanical devices for removal of husks, stalks, etc., instead of following traditional methods. The conventional methods of sun drying, smoke curing,

trimming, curing (of cinnamon bark), or bleaching (of ginger and cardamom) should be re-examined and modified if necessary, and the operations should be mechanized. This is essential to maximize flavor and pungency and to minimize contamination with foreign matter, stalks, husks, and microorganisms. It may be necessary to design suitable equipment for these mechanical operations.

8. Processing Technology. (a) Spice grinding. Spice-grinding techniques should be studied in more detail in order to evolve more efficient methods with a view to preventing changes from taking place in ground spices with respect to flavor and pungency. It is known that temperatures above 35°C reached during grinding cause deterioration of delicate flavor. Modern precrushing and milling techniques such as the Cryomill process or sieveless fine grinding by Contraplex wide-chamber mills with rotatable stud disks permit spices to be milled to the desired fineness, without risk of metallic contamination. The cast iron plate grinding mills that yield spice powder contaminated with iron dust, etc., and are still being used in some countries, should be dispensed with.

b. Spice products technology. Spice concentrates, oleoresins, and essential oils have become more in demand as exportable items and also as flavoring agents in pharmaceutical preparations, beverages, and commercial or household food formulations. Methods should be standardized for extraction and recovery of these concentrates, removal of traces of solvents, and stabilization. In countries where spice concentrates are largely employed in food processing, specialty blends are in demand. Developmental work on tailor-made oleoresin blends should be initiated after a survey of national and international markets. In fact, such an approach, which is very common in the perfumery industry, offers a vast opportunity for innovative and developmental work. Newer products such as canned, bottled, or dehydrated tender green pepper deserve greater attention from the exporters and the researchers for further improvement.

9. Packaging Technology. As a means of increasing the shelf life of foods in general and of spices and condiments in particular, packaging has gained considerable importance. The development of new and improved transparent plastic films, foils, laminations, high-speed film-sealing machines, deaerators, gas flushers, etc., has created new opportunities for food packaging. Spices, condiments, and new spice products such as instant spices, liquid spices, spice concentrates, liquid masalas, spice pastes, spice powders, and encapsulated soluble dry seasonings such as Spisoseals, are speciality products and therefore generally costly; they require attractive, efficient, and comparatively inexpensive packaging, particularly for retail marketing in Western countries, where attractively packaged spices fetch a premium price. This aspect should receive greater attention from packaging technologists and exporters.

a. Unit packages. Factors such as the compatibility between the packaging materials and the products, and the migration of components from the packaging materials into the product and vice versa, should be considered before any

packaging material is selected. With increasing consumer insistence on fair practices in packaging and labeling and also with increasing costs of packaging materials, the packages should be of minimum requisite size to contain the products. Since research on the packaging of spices and spice products has been meager, it is imperative to devote more time to this matter.

Cardboard cartons with a flexible bag inside can provide the chemical and mechanical protection required for spices. Such lined, folding cartons can hold liquids, pastes, and powders and are suitable for gas packaging and reclosure facilities. Compared to other forms of rigid containers, these cartons are light in weight and occupy less space, but they require more packaging time. All these aspects should be carefully studied.

For powdery spices and blends, composite containers made of cardboard bodies and seamed metal tops with suitable coatings or linings could also be tried. The production of molded aluminum containers should be increased, as the trend is toward the export of more essential oils and oleoresins.

b. Bulk packages. Bulk packages such as jute sacks are sometimes used without any lining material, but the addition of kraft paper and polyethylene film would prevent sifting, and the film, to some extent, would protect the commodities against moisture and also against insect attack. Woven plastic and multiwall paper sacks with plastic or bituminized liners could be tried for ground and whole spices, as these materials possess adequate physical strength and low permeability to water vapor.

Wooden containers used for the shipment of spices are rather heavy and are subject to insect attack. Corrugated and solid fiberboard containers are available in various sizes, and they can be filled and closed rapidly. Insect infestation could be reduced by the incorporation of insect repellants in the sealing tapes and adhesives between the plies at the conversion stage. Bituminized kraft or polyethylene in the facing liners of the boxes retards the ingress of moisture. The possibility of using plywood containers with appropriate liners should be explored for whole and ground spices.

c. Other packaging requirements. High-density polyethylene film has better barrier properties and more rigidity than low-density polyethylene. Films of polyvinyl chloride, regenerated cellulose, and polypropylene, either singly or laminated with other suitable packaging films, have varying degrees of barrier properties to prevent the passage of water vapor, gases, organic volatiles, and the migration of fats. Since many of the indigenously available packaging materials are not suitable for certain techniques such as gas packaging, the availability of newer packaging materials such as polyester and nylon would facilitate the design of more functional packages. This aspect of packaging of spices and spice powders needs detailed investigation.

d. Standardization of packages. There is also a need for standardization and grading of packing materials, reduction in the variety of containers, rationalized

net weights of packages and their tolerances, uniformity in essential features of labeling, classification of packaging hazards, and objective methods of assessment or testing of packages. Suitable coverage of these subjects in a logical and flexible manner should bring about economy in use of materials and a reasonable growth of relevant industries. This will also help to coordinate the work of package designers, laboratory technicians, exporters and distributors of spices and spice products, standards-setting bodies, and the certifying authorities.

10. *Storage, Transport, and Shipping.* In these areas, studies on spices and spice products are scanty and need greater attention.

11. *Microbiological Aspects.* Although some useful information on the nature and extent of microbial contamination and changes in microbial load during milling and processing operations is available, there is an urgent need for the development of efficient and economically feasible techniques for the inactivation of microflora in spices without significantly affecting the delicate aroma and other quality characteristics of spices. The possibility of thermal inactivation of microflora in large, sealed containers by using a spin pasteurizer or any suitable agitating cooker may be examined further. Other methods, such as atmospheric or vacuum fumigation, with or without heat treatment, and cold sterilization of spices by irradiation, should be further investigated with the objective of developing the most economical and practical method possible on a commercial scale for the inactivation of microflora.

12. *Insect Infestation and Its Control.* Pilot studies on the effect of treatment of burlap bags to provide protection from insect and microbial infestation during storage and shipment should be undertaken on a large scale by shippers at the international level, since such treatment could be of immense use to the spice industry as a whole and could also help in stabilizing prices to a certain extent. Atmospheric or vacuum fumigation and serial fumigation of spices on a large scale should also be undertaken. To accomplish this, the latest information on these innovations should be disseminated among dealers, exporters, various trade associations, export promotion councils, and warehousing, shipping, and other organizations. Efforts should also be made to develop simple, efficient, and reliable methods of decontamination of spices and packed spices, as well as methods for the determination of different pesticide residues.

13. *Quality Control and Standardization.* Spices, spice concentrates, and other spice products must conform to certain specifications, especially in the export market. The specifications are continually becoming more and more stringent. Contamination with microorganisms, insects, insect fragments, and pesticide residues is going to be viewed with increasing seriousness. It is, therefore, necessary that the procedures should be ready along with the methodology for quality evaluation and decontamination. Further, in order to establish standards where necessary and to upgrade the specifications, considerable physical, chemi-

cal, and microbiological data should be collected on spices in different seasons of the year.

Turmeric is one of the most common ingredients in the diets of most of the Southeast Asian countries, having being consumed for centuries. While formulating quality standards for foods and food additions, the World Health Organization is considering the banning of curcumin as a food colorant. Centuries of experience backed by research data do not support the claims of some parties that curcumin is harmful. This aspect—the effect of curcumin on human physiology—needs further scientific investigations to substantiate the claim or to establish the true facts before enforcement of such standards.

It is desirable to collect data on the quality requirements of spices and semi-preserved spice products in the importing countries.

14. *Marketing.* Marketing of spices and spice products depends to a large extent on the rapport established between the producer and the marketing agencies. It is highly necessary that production be tailored to suit the requirements of the markets. Suitable steps may be taken to assess the market requirements of spices on national and international levels so that production of spices can be adjusted accordingly.

a. Global market research. Global demands for different spices should be projected in a rational and dependable manner over a length of time based on systematic market research. Market intelligence, which covers collection and compilation of statistics, product development, market development, and sales promotion, needs to be improved. Sales and study teams should regularly visit existing and potential world markets in order to help regulate the factors of supply and demand.

b. Study of quality changes in spices during shipping and storage: There is an urgent need for studies on changes in spices during shipping or transport to distant destinations in importing countries.

c. Need for better shipping facilities: Inadequate sailings, refusal of space, and shutting out of cargo on alleged grounds of incompatibility of spices and spice products with other cargo or lack of proper storage have, in increasing measure, affected exports and hence the world supply of spices. This problem should be tackled immediately to help the flow of spices, both exports and imports, without interruption.

15. *Spices as Food Additives.* Recently, the Western world has shown an increasing interest in newer flavoring agents. With the growing anxiety about the toxicity of synthetic food additives, emphasis is shifting toward the discovery of newer flavorants, colorants, and texturizing agents of natural origin. There is a vast scope for exploitation in this new wave of interest. Several herbs and plant materials have been used for such varied applications in different parts of the world but have not yet received serious attention. Collaborative research pro-

grams should be initiated to survey such plant materials for essential oils, oleoresins, pigments, gums, mucilages, or polysaccharides. In fact, portions of spice plants themselves should be studied for such applications. Residues left after extraction of the active principles have in some cases, like chilies, already been proposed for alternative uses.

16. *Search for New Applications.* It is the responsibility of the producer and the seller to invent new strategies to boost the demand for spices. In the field of spices and flavoring extracts, such a need is already apparent. It may be worth exploring the possibility of formulating new beverages—alcoholic and nonalcoholic—snacks, stuffings, and a variety of such products. Investigations on the pharmacological properties of the active principles in spices may suggest new applications as well.

To promote such newer food applications, it may be possible to introduce modifications in spice concentrates to improve the flavor characteristics, to suppress discordant notes, and also to impart dispersibility. Basic investigations on the chemistry and chemical alterations may help in achieving these objectives.

17. *International Collaborative Research and Liaison.* Research activities in the field of spices can be fruitful only if a close liaison is established among growers, processors, exporters, and export promotion authorities on the one hand, and scientific research organizations and universities on the other. Such a well-knit interrelationship and a continuous feedback of information from the exporting and marketing organizations to the scientists are absolutely essential. The FAO could take the initiative in setting up a committee to promote international marketing and research programs on sound scientific lines for the benefit of all concerned and for humanity at large.

18. *Other Aspects.* Further spice research is necessary to provide new opportunities in the diversification of trade in spices, spice blends, and their products and to find new uses of spices in human physiology.

Our knowledge of spices in relation to evaluation of their flavor quality is fragmentary. Flavor is a conglomeration of many contributing factors. We know more about these factors now than we did yesterday. However, further knowledge must be continuously acquired as a first priority in the various disciplines engaged in this complex field of spice flavor, if we are to develop methods for its correct and rapid evaluation.

References

Aas, A. (1966). Cold storage of onions and leeks. *Gartneryket* **56,** 972.

Abdel-Hafez, F. A., Hamid, A. S., and Eid, M. N. (1964). Studies on basil oil. I. The physico-chemical properties of basil oil. *Ann. Agric. Sci.* **9,** 69.

Abdel-Hafez, F. A., Hamid, A. S., and Eid, M. N. (1966). Studies on basil oil. II. Acetylation of basil oil. *Ann. Agric. Sci.* **9,** 79.

Abdel-Hafez, F. A., Hamid, A. S., and Eid, M. N. (1967a). Basil oil. III. Benzoylation of basil oil. *Ann. Agric. Sci.* **10,** 68.

Abdel-Hafez, F. A., Hamid, A. S., and Eid, M. N. (1967b). Basil oil. IV. Formylation of basil oil. *Ann. Agric. Sci.* **10,** 117.

Abdel-Hafez, F. A., Abdel Baki, M. M., El-Waraki, A., and Abbas, S. (1967c). Studies on essential oil of rosemary. I. Isolation of rosemary oil. *Ann. Agric. Sci.* **10,** 137.

Abdel-Hafez, F. A., Abdel Baki, M. M., El-Waraki, A., and Abbas, S. (1968). Studies on the essential oil of rosemary. *Ann. Agric. Sci.* **11,** 99.

Abraham, K. O., Shankaranarayana, M. L., Raghavan, B., Sastry, L. V. L., and Natarajan, C. P. (1973). Asafetida. I. Oxidimetric determination of the volatile oil. *Flavour Ind.* **4,** 310.

Abraham, K. O., Shankaranarayana, M. L., Raghvan, B., and Natarajan, C. P. (1976). Alliums—varieties, chemistry and analysis. *Lebensm.-Wiss.+Technol.* **9,** 1973.

Abraham, P. (1965). "The Cardamom in India," *Farmers' Bull.* (New Ser.), 37. Indian Counc. Agric. Res., New Delhi.

Adam, J. R., and Merz, A. R. (1929). Hygroscopicity of fertilizer materials and mixtures. *Ind. Eng. Chem.* **21,** 305.

Adamanis, F., and Kaczmarek, F. (1955). Coriander flakes as raw materials for oil and protein. *Przem. Spozyw.* **9,** 411.

Adamson, A. D. (1971). Oleoresins: Production and markets with particular reference to the United Kingdom. *Trop. Prod. Inst.* [*Rep.*] *G (G.B.)* **G56.**

Ahmad, S. M., Gupta, M. R., and Majumder, S. K. (1976). Effect of oleoresins and powders of some dried fruits and rhizomes on the residual toxicity of pyrethrins. *Botyu-Kagaku* **41,** 135.

Ahuja, M. M., and Nigam, S. S. (1970). Chemical investigation of the essential oil obtained from the

seeds of *Apium graveolens Linn.* (Celery seed oil.) Part I. *Riechst, Aromen, Koerperpflegem.* **20,** 339.

Aiyar, A. S., Kapur, O. P., Kokatnur, M. G., Subbarao, D., Sreenivasan, A., and Subrahmanyan, V. (1964). Effect of turmeric extract on cholesterol level in rats. *Indian J. Exp. Biol.* **2,** 104.

Akman, A. (1942). "Sarape, Sirke ve dayankle sira (Wine, Vinegar and Preserved Fruit Juices)," p. 183. Y.Z.E. Matbassi, Ankara, Turkey.

Alexander, M. M., and Subbelle, G. A. (1973). Pectic substances in onion and garlic skins. *J. Sci. Food Agric.* **24,** 611.

Alfonso, N., and Lopez, H. (1959). *Centrib. Cient. Inst. Mex. Invest. Technol.* **59-2** (cited by Schwimmer and Weston, 1961).

Allen (cf. Sadler *et al.*, 1948). "Allen's Commercial Organic Analysis." Macmillan, New York.

Allen, H. E., and McCaleb, A. G. (1937). Celery salt preparations. British Patent 473,314.

Allen, J. L. (1970). The structure of marketing of essential oils in overseas countries. *Flavour Ind.* **1,** 64.

Ally, M. M. (1961). The pharmacological action of *Zingiber Officinale. Proc. Pan Indian Ocean Sci. Congr., 4th., 1960* Sect. G, p.11.

Aloy and Valdiguie, (1925). Identification of the constituents of laudanum (detection of saffron, etc.). *J. Pharm. Chim.* **8,** 369; *Chem. Abstr.* **19,** 2389 (1925).

Amarasingha, D. K. J. (1968). Review of Ceylon's foreign trade for the year 1968. *Ceylon Trade J.* **34,** 247.

American Meat Institute (1953). "Sausages and Ready to serve Meats," Rev. ed. University of Chicago, Chicago, Illinois.

American Spice Trade Association (1964). "What You Should Know About Pepper", Leaflet. ASTA, New York.

American Spice Trade Association (1967). "What You Should Know About Dehydrated Onions," Leaflet. ASTA, New York.

American Spice Trade Association (1968). "Official Analytical Methods for Spices," 2nd ed. ASTA, New York.

Amerine, M. A., Pangborn, R. M., and Rosessler, E. S. (1965). "Principles of Sensory Evaluation of Foods." Academic Press, New York.

Ames, G. R., and Mathews, W. S. A. (1968). The distillation of essential oils. *Trop. Sci.* **10,** 136.

Ames, G. R., Barrow, M., Borton, C., Casey, T. E., Mathews, W. S., Nabney, J., and Coward, L. D. G. (1970). Bay oil distillation in Dominica. *Trop. Sci.* **13,** 13.

Amonkar, S. V., and Banerji, A. (1971). Isolation and characterisation of larvicidal principle of garlic. *Science* **174,** 1343.

Amonkar, S. V., and Reeves, E. L. (1970). Mosquito control with active principle of garlic (*A. sativum*). *J. Econ. Entomol.* **63,** 1172.

Analytical Methods Committee (1973). Application of GLC to analysis of essential oils. II. Determination of 1, 8-cineole in oils of cardamom. *Analyst (London)* **98,** 616.

Anand, J. C., and Johar, D. S. (1957). Effect of condiments on control of *Aspergillus niger* in mango pickle. *J. Sci. Ind. Res., Sect. A* **16,** 370.

Ananey, V. A. (1959). The effect of hydrogen peroxide upon tick-borne encephalitis. *Vinus Vopr. Virusol.* **4** (6), 47.

Ananthakrishna, S. M., and Govindarajan, V. S. (1974). Evaluation of spices and oleoresins. IV. Estimation of pungent principles of ginger oleoresin. *Lebensm.-Wiss. +Technol.* **7,** 220.

Ananthakrishna, S. M., and Govindarajan, V. S. (1975). Evaluation of spices and oleoresins. V. Estimation of pungent principles of pepper. *J. Food Sci. Technol.* **12,** 253.

Anderson, E. E., Ruder, L. F., Esselen, W. B., Jr., Nebesky, E. A., and Labbee, M. (1951). Pasteurized fresh whole pickles. II. Thermal resistance of micro-organisms. *Food Technol.* **5,** 364.

Anderson, E. E., Esselen, W. B., Jr., and Handleme, N. A. R. (1953). The effect of essential oils on the inhibition and thermal resistance of micro-organisms in acid food products. *Food Res.* **18,** 40.

Anderson, L. (1970). Flavors and nonalcoholic beverages. Allyl isothiocynate in mustard seed. *J. Assoc. Off. Anal. Chem.* **53,** 1.

André, L., and Mile, L. (1975). Analysis of capsaicin content of paprika. *Acta Aliment. Acad. Sci. Hung.* **4,** 113.

Andrews, P., Hough, L., and Jones, J. K. N. (1952). The galactomannan of fenugreek seed. *J. Chem. Soc.* p. 2744.

Anjaneyulu, V. S. R., and Krishnamurthy, D. (1968). Turmeric in Andhra Pradesh. *J. Indian Spices* **5** (1), 8.

Anonymous (1948). Bacteriostatic oils. *J. Am. Med. Assoc.* **138,** 858.

Anonymous (1954). Cold sterilization process using ethylene oxide gas kills bacteria in spices and other dry food ingredients. *Food Process.* **15,** 20.

Anonymous (1955). Moisture content: Measurement of. *Process Control Autom.* **2,** 59.

Anonymous (1956). The microscopy of spices used in flavour confectionary. *Food* **25,** 138.

Anonymous (1957). Clove oil, its production and importance in cosmetics and perfumery. *Perfumerie (Vienna)* **11,** 6.

Anonymous (1962a). Food additives. Tamarind seed kernel powder. *Fed. Regist.* **27,** 2362; *Chem. Abstr.* **56,** 14671i (1962).

Anonymous (1962b). Changes in official methods of analysis made at the 75th annual meeting, Oct. Nov. 1961. *J. Assoc. Off. Agric. Chem.* **45,** 200.

Anonymous (1965). Dimethyl 2,3,5,6-tetrachlorotexepthalate, tolerances, for residues. *Fed. Regist.* **30,** 218.

Anonymous (1969). Spices—for what they are. *Indian Spices* **6**(2), 2.

Anonymous (1970a). Continuous process for celery oil. *Agric. Res.* **18**(11), 15.

Anonymous (1970b). Peppermint. *Flavour Ind.* **1,** 452.

Anonymous (1970c). Evaluation of oleoresins (review). *Flavour Ind.* **1,** 459.

Anonymous (1970d). Naardenized essential oils. *Gard. News* **21** (213), 2.

Anonymous (1970e). Caraway. *Flavour Ind.* **1,** 524.

Anonymous (1972). Oleoresin from Hungarian paprika. *Lebensm.-Ind.* **19,** 96.

Ansel, H. R. (1955). Spices from black pepper berries. U.S. Patent 2,713,000.

Aoi, T. (1960). The changes in skin function provoked by curry powder. Dermatological study on spices ingestion. (Report VI.) *Igaku Kenkyu* **30,** 520.

Apeland, J. (1971). Effect of scale quality on physiological process in onions. *Acta Hortic.* **20,** 72.

Arai, V. *et al.* (1957). Amino acids in *Allium* Spp. *Tokyo Toritsu Eisei Kenkyusho Kenkyu Nempo* **9,** 117.

Arjona, E. M. (1945). "Cultivation of Saffron and its Uses," Inf. Bull. No. 10. Dept. of Agriculture, Madrid.

Arnold, J. P. (1970). Note on the market for pepper. *Agron. Trop. (Paris)* **25,** 294.

Arora, R., and Pandey, G. N. (1977). The application of essential oils and their isolates for blue mould decay in *Citrus reticulata* Blanco. *J. Food Sci. Technol.* **14,** 14.

Asakawa, Y., Komatsu, T., Hayashi, S., and Matsuura, T. (1971). Chemical components of the benzene extract of *Cinnamomum loureirii* (Lauraceae). *Flavour Ind.* **2,** 114.

Ashurst, P. R., Firth, A. R., and Lewis, O. M. (1972). A new approach to spice processing. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 209.

Association of Official Analytical Chemists (AOAC) (1970). "Official and Tentative Methods of Analysis," 8th ed. A.O.A.C., Washington, D. C.

Association of Official Analytical Chemists (1975). "Official and Tentative Methods of Analysis," 9th ed. A.O.A.C., Washington, D.C.

Atal, C. K., and Sethi, J. K. (1961). Occurrence of amino acids and alliin in the Indian *Allium* (garlic). *Curr. Sci.* **30,** 338.

Atal, C. K., and Shah, K. C. (1964). Thin layer chromatographic patterns of umbelliferous drugs and their adulterants. *Indian J. Pharm.* **26,** 265.

Atal, C. K., and Sood, N. M. (1967). Study of Indian caraway and its substitutes. 1. Essential Oil of *Carum Carvi*. *Indian J. Pharm.* **29,** 42.

Atal, C. K., Girotra, R. N., and Dhar, K. L. (1966). Occurrence of sesamin in *Piper longum*. *Indian J. Chem.* **4,** 256.

Atkinson, 1. S., Cecil, S. R., Woodroof, J. G., and Shelor, E. (1947). Extending keeping quality of frozen pork sausage. *Food Ind.* **19,** 1198.

Avakova, L. S., Kustova, S. D., and Titova, N. V. (1963). Composition of the oxygen containing part of low menthol peppermint oil. *Tr. Vses. Nauchnoz Issled. Inst. Sint. Nat. Dushistykh Veshchestv* p. 125; *Chem. Abstr.* **61,** 8125c (1964).

Aycock, R., and Jenkins, J. M. (1960). Methods of controlling certain diseases of shallots. *Plant Dis. Rept.* **44,** 934.

Babayan, V. K., Kootungal, D., and Halaby, G. A. (1978). Proximate analysis, fatty acid and amino acid composition of *Nigella sativa* L. seeds. *J. Food Sci.* **43,** 1314.

Babichev, I., and Lukovnikova, G. (1961). Biochemistry of turnips, rutabagas, radishes, and horseradish. *Biokhim. Ovoshchn. Kul't.* p. 468.

Baboob, S. (1961). Cinnamon oil industry in India—a survey. *Res. Ind.* **6,** 229.

Baboob, S., and Bharmaramba, A. (1963). Studies on Indian cinnamon leaf oil. *Perfum. Essent. Oil Rec.* **54,** 30.

Bachmann, F. (1916). The inhibiting action of certain spices on the growth of microorganisms. *Ind. Eng. Chem.* **8,** 620.

Bains, G. S., Kapur, N. S., and Bhatia, D. S. (1958). Colorimetric determination of micro-quantities of vanillin. *J. Sci. Ind. Res., Sect. B* **17,** 462.

Baker, J. A. (1921). Bleached coriander. *J. Am. Pharm. Assoc.* **10,** 453.

Balansard, J., and Flandrin, P. (1945). Preparation of the saponin of fenugreek. *Med. Trop. (Marseille)* **5,** 324.

Balaraman Nair, M. (1974). "Export of Spices from India," *Proc. Symp.* Dev. Prospects Spice Ind. India, p. 68. Jointly organized by AFST (India) and CFTRI on 28th Feb.–2nd March, 1974.

Balasubrahmanyam, N., Kumar, K. R., and Anandaswamy, B. (1978). Packaging and storage studies of ground black pepper (*Piper nigrum* L.) in flexible consumer packages. *J. Indian Spices* **15**(4), 6.

Balasubrahmanyam, N., Kumar, K. R., and Anandaswamy, B. (1979). Packaging and storage studies on ground turmeric (*Curcuma longa*, L.) in flexible consumer packing. *J. Indian Spices* **16**(2), 10.

Balbaa, S. I., Karawya, M. S., and Girgis, A. N. (1968). The capsaicin content of capsicum fruits at different stages of maturity. *Lloydia* **31,** 272.

Balcar, E., and Kozlowski, J. (1962). Effect of storage period on the content of volatile oil in some pharmacopical drugs. *Biul. Inst. Przem. Zielarskiego* **8,** 1.

Baltimore Spice Co. (1957). Flavouring composition: Special food by vacuum absorption process. U.S. Patent 2,786,766; *Food Eng.* **29,** 131.

Bandyopadhyay, C., Srirangarajan, A. N., and Sreenivasan, A. (1970). Studies on flavour components of onion (*Allium capa*). 1. Thin layer chromatographic investigation of onion oil. *J. Chromatogr.* **47,** 400.

Bandyopadhyay, C., Pewari, G. M., and Sreenivasan, A. (1972). Studies on some chemical aspects of gamma-irradiated onion. *FAO/IAEA Int. Symp. Radiat. Preserv. Food, 1972.*

Bandyopadhyay, C., Gholap, A. S., and Tewari, G. M. (1973). Comparative studies on some chemical aspects of white and red globe onions. *J. Food Sci. Technol.* **10,** 169.

Barber, M. S., Jackman, L. M., Warren, C. K., and Weedon, B. C. 1. (1960). The structure of the paprika ketones. *Proc. Chem. Soc., London* p. 19.

Barber, M. S., Jackman, L. M., Warren, C. K., and Weedon, B. C. 1. (1961). Carotenoids and related compounds. 9. The structures of capsanthin and capsorubin. *J. Chem. Soc.* p. 4019.

Barnyai, M., and Szabolcs, J. (1976). Determination of the red pigment in paprika products. *Acta Aliment. Acad. Sci. Hung.* **5,** 87.

Barothy, J., and Neukom, H. (1965). Synthesis of p-hydroxbenzyl isothiocyanate and its isolation from white mustard seeds. *Chem. Ind. (London)* p. 308.

Barrington De Puyster (1919). Color. *Tr. J.* **5,** 175; *Chem. Abstr.* **14,** 745 (1920).

Baruah, A. K. S., Bhagat, S. D., and Saikka, B. K. (1973). Chemical composition of Alleppey Cardamom oil by GC. *Analyst (London)* **98,** 168.

Baslas, B. K., and Baslas, R. K. (1971). Chemical studies into the essential oils from the plant of *Anethum graveolens* and *Anethum sowa* (Dill oils). *Indian Perfum.* **15,** Part I, 27.

Baslas, K. K. (1967). Chemistry of Indian essential oils. I, 11, and III. *Perfum. Essent. Oil Rec.* **58,** 437, 622, and 782.

Baslas, K. K. (1968). Chemistry of Indian essential oils. IV and V. *Perfum. Essent. Oil Rec.* **59,** 12 and 110.

Baslas, R. K. (1970). Studies on the influence of various factors on the essential oil content from the plants of *Mentha arvenses* (Japanese Mint). *Flavour Ind.* **1,** 188.

Baslas, R. K., and Baslas, K. K. (1968a). Essential oils from some exotic plants raised in Kumaon. *Perfum. Essent. Oil Rec.* **59,** 110.

Baslas, R. K., and Baslas, K. K. (1968b). Essential oils from some exotic plants raised in Kumaon. Part II. *Perfum. Essent. Oil Rec.* **59,** 180.

Baslas, R. K., and Baslas, K. K. (1969). Essential oil from some exotic plants raised in Kumaon. Part III. *Mentha arvensis. Perfum. Essent. Oil Rec.* **60,** 337.

Baslas, R. K., Gupta, R., and Baslas, K. K. (1971). Chemical examination of essential oils from plants of genus Anethum (Umbelliferae)—oil of seeds of *Anethum graveolens.* Parts I and II. *Flavour Ind.* **2,** 241 and 363.

Battaile, J. (1960). Biosynthesis of terpenes in mint. *Dis. Abstr.* **21,** 442 (Univ. Microfilm, Ann. Arbor, Michigan. L.C. Card No. Mic. 60-3327).

Baumgartner, J. G., and Hersom, A. C. (1956). "Canned Foods—An Introduction to Their Microbiology," 4th ed. Churchill, London.

Bazorova, V. 1., Solov'eva, T. Ya., and Grimm, A. I. (1964). Oligosaccharides in some vegetable foods. *Ukr. Biokhim. Zh.* **36,** 735 (in Russian).

Beauquesne, L. B., and Delelis, A. (1967). Flavenoids from Allium as calonicum bulbs. *C.R. Hebd. Seances Acad. Sci., Ser. D* **265,** 2118.

Bedour, M., Munajjed, E., Fayez, B., and Girgis, 1. (1964). Steriod sapogenins. VII. Identification and origin of 25D-Spirosta-3,5-diene among the fenugreek sarogenins. *J. Pharm. Sci.* **53,** 1276.

Bejnarowicz, E. A., and Kirch, E. R. (1963). Gas chromatographic analysis of oil of nutmeg. *J. Pharm. Sci.* **52,** 988.

Belcher, W. F. M. (1965). The distillation of clove oils. *Perfum. Essent. Oil Rec.* **56,** 148.

Benedek, L. (1959a). Determination of capsaicin in red peppers. *Kiserletugyi Kozl.* **52,** 33.

Benedek, L. (1959b). After ripening in red pepper. *Kiserletugyi Kozl.* **52,** 69.

Benedek, L. (1968). *Z. Lebensm.-Unters.-Forsch.* **107,** 2238 (cited by Rosebrook *et al.,* 1968).

Benn, M. H. (1962). Biosynthesis of mustard oils. *Chem. Ind. (London)* p. 1907.

Bennet, E. (1945). A note on the presence of pyruvic acid in Ebenezer onions. *Plant Physiol.* **20,** 461.

Beresey, L. (1934). Effect of paprika on gastric secretion. *Orv. Hetil.* **78,** 397.

Berisso, B. (1940). Physical methods of examining certain drug spices for purity and adulteration. *Rev. Col. Farm. Nac.* **7,** 828.

Bernhard, R. A. (1957). Separation and identification of some tarpenes by gas partition chromatographic analysis. *J. Assoc. Off. Agric. Chem.* **40**, 915.

Bernhard, R. A., Wijesekera, R. O. B., and Chichester, C. O. (1971). Terpenoids and cardamom oil and their comparative distribution among varieties. *Phytochemistry* **10**, 177.

Bernstein, H. I., and Quimby, W. C. (1943). Physico-chemical determination of transcinnamic acid. *J. Am. Chem. Soc.* **65**, 1845.

Beroza, M. (1954). Micro-determination of methylenedioxyl or combined formaldehyde groups. *Anal. Chem.* **26**, 1970.

Berry, H. (1935). Examination of the extractives of capsicums. *Q. J. Pharm. Pharmacol.* **8**, 479.

Berry, H., and Samways, E. (1937). Determination of capsaicin content of capsicum and its preparations. *Q. J. Pharm. Pharmacol.* **10**, 387 (cited by the Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry, 1959).

Betts, T. J. (1964). Umbelliferous fruit identification by thin layer chromatography. *J. Pharm. Pharmacol.* **16**, Suppl., 161.

Betts, T. J. (1965a). Carvone in the developing fruits of *Anethum graveolens* and *Carum carvi*. *J. Pharm. Pharmacol.* **17**, suppl., 41.

Betts, T. J. (1965b). Examination of cinnamon by direct thin layer chromatography. *J. Pharm. Pharmacol.* **18**, 520.

Beyrich, T. (1964a). Paper chromatographic separation of furocoumarine. *J. Chromatogr.* **13**, 181.

Beyrich, T. (1964b). Furocoumarins. II. Determination of furocoumarine by paper chromatography in celery, parsley and parsnip. *Pharmazie* **19**, 700.

Bhagya (1977). Detection of capsaicin adulteration in ginger oleoresin. *J. Food Sci. Technol.* **14**, 176.

Bhargava, P. P., and Haskar, C. N. (1959). Chemical examination of ajowan oil. *Perfum. Essent. Oil Rec.* **50**, 209.

Bhargava, P. P., and Haskar, C. N. (1962a). Chemical examination of ajowan oil. *Perfum. Essent. Oil Rec.* **53**, 379.

Bhargava, P. P., and Haskar, C. N. (1962b). Essential oil from ajowan seeds and its byproducts. *Indian Oil Soap J.* **27**, 147.

Bhat, J. V., Broker, R., and Iyer, V. (1953). The role of saffron and nutmeg in indigenous milk preparations. *Indian J. Dairy Sci.* **6**, 121.

Bhat, J. V., Bhat, M. G., and Broker, R. (1954). Role of asafetida in the elementary tract: Effect of microbial fermentation and enzymes of digestion. *J. Sci. Ind. Res., Sect. B* **13**, 765.

Bhatia, I. S., Satyanarayana, M. N., and Srinivasan, M. (1955). Carbohydrates in garlic. (*Allium. sativum* Linn.) *J. Sci. Ind. Res., Sect. C* **14**, 93.

Bhatnagar, J. K., and Gupta, O. P. (1965). Microscopic detection of papaya seeds in powdered black pepper. *Res. Bull. Panjab. Univ. Sci.*, **16**, 323.

Bhatnagar, J. K., Thomas, P. L., and Pal, A. K. (1974). Studies on the insecticidal activity of garlic oil. II. Mode of action of the oil as a pesticide in *Musca domestica* Nebulo Fabr and *Throgoderma granarium* Everts. *J. Food Sci. Technol.* **11** (4), 153.

Bhatti, M. B., and Asghar, A. (1965). Certain dehydration studies on Pakistan onions. I. Composition and change in various constitutents. II. Pilot-plant dehydration. III. Reconstitution, quality and organoleptic evaluation of dehydrated varieties. *Agric. Pak.* **16**, 299, 307, and 311.

Bhramaramba, A., and Sidhu, G. S. (1963). Chromatography studies on Indian cinnamon leaf oil. *Perfum. Essent. Oil Rec.* **54**, 732.

Bickoff, E. M. (1951). Phenolic antioxidants for carotene. *J. Am. Oil Chem. Soc.* **28**, 65.

Biffoli, R. (1955). Coloring matter in food extracts. *Boll. Lab. Chim. Prov.* **6**, 23.

Bindra, O. S., and Kalra, R. L. (1973). A review of work done in pesticide residue. *Proc. Symp. Prog. Probl. Pestic. Residue Anal., 1971* p.1.

Bitter, B. (1950). Polarography of some essential oils. *Collect. Czech. Chem. Commun.* **15**, 677.

Bitting, A. (1909). *U.S., Dep. Agric. Bur., Chem. Bull.* **119** (cited by Baumgartner, and Hersom, 1956).

Bitting, K. G. (1920). "The Effect of Certain Agents on the Development of Some Moulds." Nat. Capital Press, Inc., Washington, D.C.

Bjeldanes, L. F., and Kim, I. S. (1978). Sedative activity of celery oil constituents. *J. Food Sci.* **43,** 143.

Blanco, H. G., and Leiderman, L. (1968). The use of maleic hydrazide in the storage of onion bulbs. *Biologico* **34,** 90.

Blazovich, M., and Spanyar, P. (1969). A method for determining capsaicin in oleoresin and some other preparations with high capsaicin content. *Elelmiszervizsgalati Kozl.* **15,** 358.

Blum, H. B., and Fabian, F. W. (1943). Spices, oils and their components for controlling microbial surface growth. *Fruit Prod. J.* **22,** 326 and 347.

Blumberg, B. L. (1939). Insect infestation of drugs. *J. Am. Pharm. Assoc.* **28,** 483.

Blumberger, W., and Glatzel, H. (1963). *Dtsch. Z. Verdau.-Stoffwechselkr.* **23,** 97 (cited by Glatzel, 1968).

Blumberger, W., and Glatzel, H. (1965). Physiology of spices and condiments. V. Salivary effects of paprika. *Nutr. Dieta* **7,** 222.

Blyth, A. W., Blyth, M. W., and Cox, H. E. (1927). "Foods: Their Composition and Analysis—A Manual for the Use of Analytical Chemists and Others." Griffin, London.

Bohnsack, H. (1965). Essential oils, perfumes and flavouring. XII. Constituents of pepper flavours and relation between chemical constituents and pungency. *Riechst., Aromen, Koerperpflegem.* **15,** 77.

Bohnsack, H. (1971a). Contribution to information on essential oils, odour and flavour substances. XXV. On the components of the Borbian vanilla pod. Part 5. *Riechst., Aromen, Koerperpflegem.* **21,** 125.

Bohnsack, H. (1971b). Contribution to information on essential oils odour and flavour substances. XXVI. Neutral oil on the components of the Borbian vanilla pod. Part 6. *Riechst., Aromen, Koerperpflegem.* **21,** 163.

Bollinger, H. R. (1965). *In* "Thin Layer Chromatography" (E. Stahl, ed.), p. 124. Academic Press, New York.

Bonnet, G. (1968). Gas and thin layer chromatographic separation of compounds with a vanilla flavor. Distinctive criteria of Bourbon and Tahiti vanilla extracts. Determination of vanillin, anisic alcohol and p-hydroxy benzaldehyde in vanilla extracts. *Ann. Falsif. Expert. Chim.* **61,** 360.

Booth, V. H. (1962). A method for separating lipid components of leaves. *Biochem. J.* **84,** 444.

Bose, A. N., and Roy, A. K. (1959). Heat resistance of *Bacillus substilis* spores. (i) Effect of different chemicals. *J. Sci. Ind. Res.,* Sect. C, **18,** 248.

Bose, P. K. (1974). Sport test for detection of *Argenone medicana* seeds in mustard seeds. *J. Food Sci. Technol.* **11,** 28.

Bose, S., and Shrivastava, A. N. (1961). Soluble Carbohydrates from onion (*Allium cepa*). *Sci. Cult.* **27,** 253.

Bose, S. M., BhimaRao, C. N., and Subramanyan, V. (1949). Influence of organic matter on bactericidal efficiency of Indian-essential oils. *J. Sci. Ind. Res.* **8,** 157.

Bose, S. M., BhimaRao, C. N., and Subramanyan, V. (1950). Some factors affecting the bactericidal property of lemongrass oil emulsions. *J. Sci. Ind. Res., Sect. B* **9B** 12.

Botta, A., and Morassutti Botta, M. (1956). *Montha spicata,* Scottish variety. Influence of some factors on the crop and on the oil properties. *Riv. Ital. Essenze, Profumi, Piante Off., Olii Veg., Saponi* **38,** *403; Chem. Abstr.* **51,** 3093 (1957).

Bottcher, H. (1967). Recent knowledge on the practical storage of onions. *Dtsch. Gartenbau* **14,** 316.

Bottcher, H. (1969). Results from the onion harvests of recent years. *Dtsch. Gartenbau* **16**, 189.

Bottcher, H. (1970). Importance of ventilation during the post-harvest ripening of onions. *Dtsch. Gartenbau* **17**, 245.

Bough, W. A. (1973). Characterization of waste effluents from a commercial pimiento canning operation. *J. Milk Food Technol.* **36**, 371.

Bough, W. A., and Badenhop, A. F. (1974). A comparison of roasting vs. lye peeling of pimientos for generation of wastes and quality of canned products. *J. Food Sci.* **39**, 1107.

Boyle, W. (1955). Spices and essential oils as preservatives. *Am. Perfum. Eseent. Oil Rev.* **66**, 25.

Brahamachari, H. D., and Augusti, K. T. (1961). Hypoglycemic agent from onions. *J. Pharm. Pharmacol.* **13**, 128.

Brauer, O., and Schoon, J. (1962). Determination of the flavour constituents of paprika fruits, particularly capsaicin. *Angew. Bot.* **36**, 25.

Breag, G. R., Coward, L. D. G., Nabney,, J., and Robinson, F. V. (1972). Artificial drying of Jamaican pimento. *Proc. Int. Conf. Spices Trop. Prod. Inst. London, 1972* p. 149.

Breckenridge, N. (1969). The market for spices in the United States of America. *Ceylon Trade J.* **34**, 267.

Brennand, C. P., and Heinz, D. E. (1970). Effects of pH and temperature on volatile constitutents of cardamom. *J. Food Sci.* **35**, 533.

Bretin, P. H., Manceau, P., and Namdar, M. (1931). Possible adulteration of saffron by annato. *J. Pharm. Chem.* **13**, 369; *Chem. Abstr.* **25**, 5736 (1931).

Bricker, C. E., and Johnson, H. R. (1945). Spectrophotometric method for determining formaldehyde. *Ind. Eng. Chem., Anal. Ed.* **17**, 402.

Bricker, C. E., and Veil, W. A. (1950). Microdetermination of formaldehyde with chrometrophic acid. *Anal. Chem.* **22**, 720.

Brieskorn, C. H., and Dalferth, S. (1964). Constituents of *Salvia officinalis* L. Part 15. The occurrence of 2-methyl-3-methylene-5-heptene in the essential oil of *Salvia Officinalis* L. *Justus Liebigs Ann. Chem.* **676**, 171.

Brieskorn, C. H., and Wenger, E. (1960). Constitutents of Salvia officinalis. XI. The analysis of ethereal Sage Oil by means of gas and thin bedded Chromatography. *Arch. Pharm. (Weinleim, Ger.)* **293**, 21; *Chem. Abstr.* **54**, 15846h (1960).

Brieskorn, C. H., and Zimmerma, N. K. (1965). Occurrence of pristane in the anise fruits. *Experientia* **21**, 385.

Broda, B., Wojciechowska, B., and Kombrowicz, E. (1966). Determination of glycoalkaloids in seeds of Spanish pepper (*Capsicum anuum*). *Diss. Pharm. Pharmacol.* **18**, 501.

Brodnitz, N. H., and Pollack, C. L. (1970). Gas chromatographic analysis of distilled onion oil. *Food Technol.* **24**, 78.

Brodnitz, N. H., Pollack, C. L., and Vallon, P. P. (1969). Flavour components of onion oil. *J. Agric. Food Chem.* **17**, 760.

Brodnitz, M. H., Pascale, J. V., and Derslice, L. V. (1971). Flavor components of garlic extract. *J. Agric. Food Chem.* **19**, 273.

Brown, B. I. (1973). Ginger storage in acidified metabisulphite solutions. *Food Technol.* **7**, 153.

Brown, E., and Reader, D. E. (1953). The cultivation and marketing of pepper. *Colon. Plant Anim. Prod.* **3**, 195.

Brown, E., Coomes, T. J., Islip, H. T., and Mathews, W. S. A. (1954). Oils of cinnamomum spp. from Sarawak as a source of safrole. *Colon. Plant Anim. Prod.* **4**, 239.

Brown, E. B., Thomas, J. M., and Bina, A. F. (1946). Nicotinic acid values by chemical and microbiological methods. Effects of H_2O_2 and infra-red rays on nicotinic acid. *J. Biol. Chem.* **162**, 221.

Brown, E. G. (1955). Cinnamon and Cassia: Sources, production and trade. *Colon. Plant Anim. Prod.* **5**, 257.

Browne, C. W. (1951). Packaging. *In* "Chemistry and Technology of Foods and Food Products" (M. B. Jacob, ed.), 2nd ed., Vol. 3, p. 1992.

Bruno, S. (1961). Gas chromatographic analysis of some essential oils in biological matter. *Farmaco, Ed. Prat.* **16,** 481.

Bruto, G. L., and Calvarano, M. (1959). Essential oils from Sicilian aromatic plants (incl. *Ocimum basilicum*). *Essenze Deriv. Agrum.* **29,** 182.

Buchi, J., and Hippenmeier, F. (1948). Evaluation of pure capsaicin, drugs and preparations containing capsaicin. *Pharm. Acta Helv.* **23,** 327.

Buchi, J., and Huber, R. (1961). Determination of the most important secondary alkaloids of opium. *Pharm. Acta Helv.* **36,** 571.

Bullerman, L. B. (1974). Inhibition of aflatoxin production by cinnamon. *J. Food Sci.* **39,** 1163.

Bullerman, L. B., Lieu, F. Y., and Seiier, S. A. (1977). Inhibition of growth and aflatoxin production by cinnamon and clove oils, cinnamic aldehyde and eugenol. *J. Food Sci.* **42,** 1107.

Bunt, J. S. (1964). Analysis of algal pigments by thin layer chromatography. *Nature (London)* **203,** 1261.

Burger, A. (1958). Curcuma root and its essential oils. *Perfum. Essent. Oil Rec.* **49,** 801.

Buttery, R. G., Seifer, R. M., Lundin, R. G., Guadgni, D. G., and Ling, L. C. (1969). Characteristics of an important aroma component of bell pepper. *Chem. Ind. (London)* **15,** 490.

Buzas, C., and Sqendrei, K. (1964). Identification of plant drugs by ultra-violet fluoroscence. *Pharmazie* **19,** 771.

Buzinov, P. A. (1962). Action of ionizing radiation on crop and content of essential oils in basil (*Ocimum basilicum*) and anise (*Pimpinella anisum*). *Maslob.-Zhir. Prom-st.* **28,** 28.

Cade, A. R. (1954). *In* "Antiseptics, Dissinfectants, Funguides, and Chemical and Physical Sterilization" (G. F. Riddish, ed.), Chapter 15. Lea & Febiger, Philadelphia, Pennsylvania.

Cairncross, S. E., and Sjöström, L. B. (1950). Flavour profiles—a new approach to flavour problems. *Food Technol.* **4,** 308.

Calzolari, C., Stancher, B., Pertoldi, I., and Marletta, G. (1968). Origanum oils and their investigation by gas chromatographic and infra-red spectroscopic analysis. *Analyst (London)* **93,** 311.

Cambell, C. H. (1920). Sweet cider—some methods of handling for bottling or barreling. *Canner* **51,** 51.

Capek, A. (1956). Phytoncides in anise and fennel. *Prum. Potravin* **7,** 260.

Capelle, G. B., and Monique De Serres (1967). Distribution of arginase in bulbous plants. Allium species. *C. R. Hebd. Seances Acad. Sci., Ser. D* **264,** 54.

Carr, D. S., and Harris, B. L. (1949). Solutions for maintaining constant relative humidity. *Ind. Eng. Chem.* **41,** 2014.

Carson, J. F. (1967). Onion flavour. *Proc. Symp. Foods, 4th, 1965* p. 390.

Carson, J. F., and Wong, F. F. (1961a). Onion flavor and odor: The volatile flavor components of onions. *J. Agric. Food Chem.* **91,** 40.

Carson, J. F., and Wong, F. F. (1961b). Isolation of (+) -S. methyl - L. cystein sulfoxide and of (+) -S- Propyl-L-cysten sulfoxide from onions as their N- (2,4-dinitrophenyl) derivatives. *J. Org. Chem.* **26,** 4997.

Carson, M. A. (1950). Report on volatile oil in spices. *J. Assoc. Off. Agric. Chem.* **33,** 575.

Carson, N. A. (1953). Report on volatile oil in spices. *J. Assoc. Off. Agric. Chem.* **36,**752.

Carson, N. A. (1954). Volatile oils in spices. *J. Assoc. Off. Agric. Chem.* **37,** 390.

Carson, N. A. (1955). Volatile oils in spices. *J. Assoc. Off. Agric. Chem.* **38,** 548.

Cartwright, L. C., and Nanz, R. A. (1948). Comparative evaluation of spices. *Food Technol.* **2,** 330.

Casetti, P. (1962). Garlic powder. French Patent 79,158.

Castell, C. H. (1944). Thermophilic bacteria in foods and in various ingredients entering into the manufacture of foods. *Food Res.* **9,** 410.

Caul, J. F. (1957). The profile method of flavour analysis. *Adv. Food Res.* **7**, 1.

Caul, J. F. (1960). Geruchs und Geschmacksanalysen. Mitt der Profile method. *Einaehrungswirtschaft* **7**, 398.

Cavallito, C. J., and Bailey, J. H. (1944). Allicin, the antibacterial principle of Allium sativum. 1. Isolation, physical properties and antibacterial action. *J. Am. Chem. Soc.* **66**, 1950.

Central Food Technological Research Institute, Mysore (1963). Studies on packaging and storage of dry green cardamom. *Spices Bull.* **3** (3-5), 3.

Central Food Technological Research Institute, Mysore (1964). Development of compressed package for dried chillies. *Spices Bull.* **3**, 91.

Chadha, D. S. (1973). Pesticide residues in foods—their need, present status and principles for laying down tolerances. *Proc. Symp. Prog. Probl. Pestic. Residue Anal., 1971* p. 267.

Chakraborty, D. P., Sen, M., and Bose, P. K. (1961). Antibiotic properties of some natural coumarins. *Trans. Bose Res. Inst., Calcutta* **24**, 31.

Chakravarti, K. K., and Bhattacharya, S. C. (1954). Chemical examination of Indian spearmint oil. Part 1. Separation and characterisation of various constituents. *Perfum. Essent. Oil Rec.* **45**, 217.

Chakravarti, K. K., and Bhattacharya, S. C. (1955a) Chemical examination of Indian spearmint oil. Part II. Elucidation of the structure of mintglyoxal. *Perfum. Essent. Oil Rec.* **46**, 258.

Chakravarti, K. K., and Bhattacharya, S. C. (1955b). Chemical examination of Indian dill oil—isolation of dihydrocarvone. *Indian Pharm.* **9**, 218.

Chang, L. (1940). Pharmacological action of cinnamon. *Proc. Jpn. Pharmacol. Soc.* **13**, 52.

Chang, S. S. (1966). Flavour characterization. *Kirk-Othmer Encycl. Chem. Technol., 2nd ed.* Vol. **9**.

Chang, S. S., Matijasavic, B. O., Hiseh, O. A. L., and Huang, C. L. (1977). Natural antioxidants from rosemary and sage. *J. Food Sci.* **42**, 1102.

Chauhan, U. P. S., and Ray, B. C. (1964). A microchemical method for the determination of strontium in some plant materials. *Indian J. Chem.* **2**, 175.

Chawan, T., and Pflug, I. J. (1968). Controlled atmosphere storage of onions. *Mich., Agric. Exp. Stn., Q. Bull.* **50**, 449.

Chen, S. L., and Gujmanis, F. (1968). Auto-oxidation of extractable colour pigments in chilli pepper with special reference to ethoxyquiim treatment. *J. Food Sci.* **33**, 274.

Chernysheva, S. L. (1966). Determination of linalcol in coriander oil. *Maslo-Zhir. Prom-st.* **32**, 29.

Chipault, J. R., Mizuna, G. R., Hawkins, J. M., and Lundberg, W. O. (1952). The antioxidant properties of natural spices. *Food Res.* **17**, 46.

Chipault, J. R., Mizuna, G. R., Hawkins, J. M., and Lundberg, W. O. (1955). The antioxidant properties of spices in oil-in-water emulsions. *Food Res.* **20**, 443.

Chipault, J. R., Mozuna, G. R., Hawkins, J. M., and Lundberg, W. O. (1956). The antioxidant properties of spices in foods. *Food Technol.* **10**, 209.

Cholnoky, L., and Szabolcs, J. (1960a). Structure of capsorubin. *Experientia 16*, 483.

Cholnoky, L., and Szabolcs, J. (1960b). Structure of colouring matter of paprika. *Acta Chim. Acad. Sci. Hung.* **22**, 117.

Cholnoky, L., Gyorgyfy, K., Magy, E., and Panczel, M. (1958). Investigations of caratenoid pigments. III. Pigments of the yellow paprika. *Acta Chim. Acad. Sci. Hung.* **16**, 227.

Cholnoky, L., Szaboles, J., Cooper, R. D. G., and Weedon, B. C. L. (1963). The structure of kryptocapsin. *Tetrahedron Lett.* **19**, 1257.

Chopra, I. C., Khajuria, B. N., and Chopra, C. L. (1958). *Alpinia galanga* Willd. Antibacterial spectrum. *Proc. Symp. Antibiot., 1958*, p. 43.

Chopra, M. M. (1965). Oils of cardamom and pepper. *Indian Perfum.* **9**, 3.

Chopra, M. M., Vashist, V. N., and Handa, K. L. (1964). The essential oils of the species of genus Mentha. *Perfum. Essent. Oil Rec.* **55**, 323.

Chopra, M. M., Nigam, M. C., and Rao, P. R. (1968a). Pharmacopoial grade oil of dill from Anethum sowa. *Perfume Essent. Oil Rec.* **59**, 604.

Chopra, M. M., Nigam, M. C., and Rao, P. R. (1968b). Production in India of celery seed oil. *Perfum. Koset.* **49**, 223.

Chopra, R. N., Chopra, I. C., Handa, K. K., and Kapur, L. D. (1958). "Indigenous Drugs of India," 2nd ed. U.N. Dhar & Son Ltd., Calcutta.

Chow, P. N., Motl, O., and Lukes, V. (1965). Terpenes. CLXXIII. Hydrocarbons from the oil of laurel leaves. *Collect. Czech. Chem. Comnum.* **30**, 917.

Christensen, R. E., Beckman, R. M., and Birdsall, J. J. (1968). Some mineral elements of commercial spices and herbs as determined by direct reaching emission spectroscopy. *J. Assoc. Off. Anal. Chem.* **51**, 1003.

Cieri, U. R. (1969). Characterization of steam—non-volatile residue of bergamot oil and some other essential oils. *J. Assoc. Off. Anal. Chem.* **52**, 719.

Cingova, B. (1967). Results from trials with some coriander varieties. *Rastenievud. Nauki* **4** (9), 11.

Cipriani, C. (1959). Additives for liquid or gasseous fuels. German Patent 1,054,273.

Clark, W. R. E. (1970). Modern trends in the application of spice. *Food Manuf.* **45**, p. 53.

Clayton, C. (1918). Electrical conductivity of emulsions. *Br. Assoc. Colloid Rep.* **2**, 114.

Clevenger, J. F. (1928). An apparatus for the determination of volatile oil in plant and plant products. *J. Am. Pharm. Assoc.* **17**, 345.

Clevenger, J. F. (1933). Report on the determination of volatile oil in spices. *J. Assoc. Off. Agric. Chem.* **16**, 557.

Clevenger, J. F. (1935). Report on the analysis of spices. *J. Assoc. Off. Agric. Chem.* **18**, 417.

Clevenger, J. F. (1942). Volatile oil in spices. Assay of caraway and fennel. *J. Assoc. Off. Agric. Chem.* **25**, 700.

Coffman, I. R., and Schwecke, W. M. (1962). Gas-liquid chromatography of vanillin and related substances. *Gas Chromatogr. Int. Symp. (U.S.)* **3**, 471.

Colombo, E., and Manitto, P. (1971). Star anise oil. *Parfums, Cosmet., Savons Fr.* **1**, 347.

Commonwealth Secretariat (1969). "Plantation Crops—Review." HM Stationery Office, London.

Connel, D. W. (1969). Pungent principles of ginger and their importance in ginger products. *Food Technol.* **21**, 570.

Connel, D. W. (1970a). Natural pungent compounds. III. The paradols and associated compounds. *Aust. J. Chem.* **23**, 369.

Connel, D. W. (1970b). The chemistry of the essential oil and oleoresin of ginger. *Flavour Ind.* **1**, 677.

Connel, D. W., and Jordan, R. A. (1971). Composition and distinctive volatile flavour characteristics of essential oil from Australian grown ginger. *J. Sci. Food Agric.* **22**, 93.

Connel, D. W., and Sutherland, M. D. (1969). Reexamination of gingerol, shogoal, and zingerone, the pungent principles of ginger (*Zingiber officinale*). *Aust. J. Chem.* **22**, 1038.

Considen, W. J., and Pearl, B. (1960). Vanilla extract. II. Stability of lead test solution. *Food Technol.* **14**, 204.

Constabel, F. (1963). Quantitative studies of the extracellular hydrolysis of the carbohydrates by tissue cultures of *Juniperus communis*. *Planta* **59**, 330.

Cook, W. B., and Haward, A. S. (1966). The essential oil of *Illicium anisatum* Linn. *Can. J. Chem.* **44**, 2461.

Cook, W. B., and Haward, A. S. (1967). The sesquiterpene hydrocarbons of *Illicium anisatum* Lin. *Can. J. Chem.* **45**, 2002.

Cooper, R. D. G., Jackman, L. M., and Weedon, B. C. L. (1962). Stereochemistry of capsorubin and synthesis of the optically active epimers. *Proc. Chem. Soc., London* p. 25.

Coretti, K. (1955a). Bacterial content of spices. *Fleischwirtschaft* **7**, 305.

Coretti, K. (1955b). Sterilization of spices—a hygienic necessity for sterilization of spices by ultraviolet radiation. *Fleischwirtschaft* **7**, 386.

Coretti, K. (1957). Cold sterilization of spices by ethylene oxide. *Fleischwirtschaft* **9**, 183.

Coretti, K., and Inal, T. (1969). Ruckstandproblems bei der kattentkeimune von yewiirzen mit. *Fleischwirtschaft* **49**, 599.

Corran, J. W. (1934). Emulsification by mustard. *Food Manuf.* 9 (1), 1.

Corran, J. W., and Edgar, S. H. (1933). Preservation action of spices and related compounds against yeast fermentation. *J. Soc. Chem. Ind.* **52**, 149T.

Cort, W. M. (1974). Haemoglobin peroxidation test screens antioxidants. *Food Technol.* **28**, 60.

Cotton, R. T. (1963). "Pests of Stored Grain and Grain Products," p. 306. Burgess, Minneapolis, Minnesota.

Council of Scientific and Industrial Research, Publication Directorate (1949-1976). "Wealth of India," 1st ed., Vols. I-XI. CSIR, New Delhi, India.

Courtois, J. E., and Percheron, F. (1961). Galactosidase of germinated seeds of fenugreek. *Bull. Soc. Chim. Biol.* **43**, 167.

Covello, M. (1943). Trigonellins and nicotinic acid in fenugreek and their relation to antipellagra activity. *Boll. Soc. Ital. Biol. Sper.* **18**, 159.

Cowan, J. W., Saghir, A. R., and Saiji, J. P. (1967). Antithyroid activity of onion volatiles. *Aust. J. Biol. Soc.* **20**, 683.

Cowley, E. (1972). Vanilla and its uses. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 79.

Cox, H. E., and Pearson, D. (1962). "Chemical Analysis of Foods." Chem. Publ. Co., New York.

Cripps, H. D. (1967). Oleoresin turmeric—application in pickle production. *Glass Packer/Process.* **46**, 24.

Cripps, M. H. (1972). Spice oleoresins, the process, the marketing and the future. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 237.

Crocker, E. C. (1945). "Flavor." McGraw-Hill, New York.

Crossley, E. L. (1938). *J. Hyg.* **38**, 205 (cited by Baumgartner and Hersom, 1956).

Csedo, K., and Kopp, E. (1964). Red paprika varieties applicable in therapeutics. *Pharmazie* **19**, 541.

Csedo, K., Horvath, M. P., and Nagy, S. (1960). Determination of capsaicin contents of red pepper (*Capsicum frutescens*). *Farmacia (Bucharest)* **8**, 435.

Csiba, L., and Kilb, Gy. (1959). The damage caused by *Rhyzopus nigricans* in paprika stored in strings. *Konzerv. Hutoipari Kutato Ihtezet Kozle.* Nos. I-II, p. 33.

Curl, A. L. (1962). The carotenoids of red bell peppers. *J. Agric. Food Chem.* **10**, 504.

Curl, A. L. (1964). The carotenoids of green bell peppers. *J. Agric. Food Chem.* **12**, 522.

Currier, H. B. (1945). Photometric estimation of volatile sulfur in onions as a criterion of pungency. *Food Res.* **10**, 177.

Czaja, A. T. (1949). A recent cinnamon imitation and microchemical method of detecting cinnamic aldehyde. *Dtsch. Lebensm.-Rundsch.* **45**, 396.

Czaja, A. T. (1953). The detection of roller dried potato meal in clour and spice mixes. *Dtsch. Lebensm.=Rundsch.* **49**, 231.

Czaja, A. T. (1962). Polarisation—microscopical detection of palm kernel meal in condiments *Z. Lebensm.-Unters.-Forsch.* **118**, 32.

Czernicki, B., and Weiser, H. (1962). Influence of pure carotenoids, especially those of paprika on yolk. *Zentralbl. Veterinaermed.* **9**, 899.

Czionkowska, H., Rutkowska, U., and Wojsa, K. (1966). Polarographic copper, lead and zinc estimation in plant material. *Herba Pol.* **11**, 194.

Dac, (1966). Oral contraceptive. British Patent 1,025,372.

Dakshinamurthy, K. (1962). Methionine content of some south Indian foods. *J. Food Sci.* **27**, 367.

Damjanic, A., Borcic, I., and Vrscaj, P. (1969). Chromatographic determination of the oil of Thymus vulgaris. *Riv. Ital. Essenze, Profumi, Piante Off., Aromi, Saponi, Cosmet., Aerosol* **51,** 65.

Damrau, F., and Ferguso , E. A. (1949). Garlic in functional gastrointentinal disorders. *J. Am. Med. Assoc.* **141,** 626.

Danos, B., and Sarkany, S. (1961). Novel results of the poppy hybrid experiments directed to increasing morphine production. *Acta Pharm. Hung.* **31-B,** Suppl., 108.

Daoud, H. N., and Luh, B. S. (1967). Packaging of foods in laminate and film combination pouches IV. Freeze dried red bell peppers. *Food Technol.* **22,** 21A.

Dastur, J. J. (1951). "Medicinal Plants of India and Pakistan," p. 225. D. B. Taraporwal & Co., Bombay.

Date, W. B. (1960). Effect of preharvest foliar sprays of maleic hydrazide on the cold storage behaviour of onions and garlic. *Food Sci.* **9,** 203.

Datta, P. R., and Susi, H. (1961). Quantitative differentiation between natural capsaicin (8-methyl-N-Vanillyl-l-nomenamide) and vanillyl-n-nondnamide. *Anal. Chem.* **33,** 148.

Datta, P. R., and Susi, H. (1962). Gas chromatographic separation of oxygen containing terpene compounds on low temperature columns. *Anal. Chem.* **34,** 1028.

Datta, P. R., Susi, H., Higman, H. C., and Filipic, V. J. (1962). Use of gas chromatography to identify geographical origin of some spices. *Food Technol.* **16,** 116.

David, H. N., and Luh, B. S. (1967). Packaging of foods in laminate and aluminum film combination pouches. lV. Freeze-dried red bell peppers. *Food Technol.* **21,** 21A.

Davies, B. H. (1965). Analysis of carotenoid pigments. *In* "Chemistry and Biochemistry of Plant Pigments" (T. W. Goodwin, ed.), p. 489. Academic Press, New York.

Dayal, B., and Purohit, R. M. (1971). Chemical examination of the essential oil from the seeds of *Majorana hortensis* Moench. *Flavour Ind.* **2,** 477.

Deal, E. C. (1967). Report on spices and other condiments. *J. Assoc. Off. Anal. Chem.* **50,** 831.

Deb, A. R., Ramanujam, S., Krishnamurti, C. S. R., and Tirumalachar, D. K. (1963). Capsaicin content of some new Pusa varieties of chilies. *Indian J. Technol.* **1,** 59.

Debrawere, J., and Verzele, M. (1975). New constituents of the oxygenated fraction of pepper essential oil. *J. Sci. Food Agric.* **26,** 1887.

De Gavina Mugica, M. *et al.* (1969). Oil of Spanish sage (*Salvia lavendulaefolia Vehl.*). *Parfums, Cosmet., Savons* **12,** 334.

Delage, J. C. (1923). Use of mustard meal and of mustard oil in enology. *Ann. Falsif. Fraudes* **16,** 483.

De La Mar, R., and Francis, F. J. (1969). Carotenoid degradation in bleached paprika. *J. Food Sci.* **34,** 287.

Delkinova, S. M., and Dupuy, P. (1973). Preservation of onion powder. *Acta Aliment. Acad. Sci. Hung.* **2,** 181.

Delonca, H., Puech, A., and Tirat, P. (1963). Estimation of allyl isothiocyanate in black mustard flour. *Trans. Soc. Pharm. Montpellier* **23,** 271.

De Martinez Mandal, N. G. (1959). Study of the non-terpenic fraction of Puerto Rican bay oil, *Myrcia acris. Am. Perfum.* **73,** 22.

Demole, E. (1958). Application of absorption micro-chromatography in thin layers. *J. Chrômatogr.* **1,** 24.

Demole, E. (1959). Chromatostrips and chromatoplates. *Chromatogr. Rev.* **1,** 1.

Deryng, J., Strzelecka, H., and Walewska, E. (1966). Volatile oils of the polish Pharmacopeici. 1. *Oleum coriandri & Oleum carvi. Farm. Pol.* **22,** 7; *Chem. Abstr.* **65,** 13451e (1966).

Deryng, J., Strzelecka, H., Walewska, E., and Sorocyzynska, T. (1967). Volatile oils of the polish pharmacopeia. ll. *Oleum foenieuli and Oleum anisi. Farm. Pol.* **23,** 32; *Chem. Abstr.* **67,** 93904a (1967).

Desikachar, H. S. R., and Muthu, M. (1962). Processing and preservation of turmeric. *Proc. Symp. Spices, Role Natl. Econ. 1st, 1962,* Abstr. 19, p. 11.

Desikachar, H. S. R., Srinivasan, M., and Subrahmanyan, V. (1959). Processing of turmeric. *Res. Ind.* **4,** 172.

Devetak, Z., and Cenci, A. (1967). The essential oil of *Salvia officinalis* from Montenegro, Yugoslavia. *Riv. Ital. Essenze, Profumi, Piante Off., Aromi, Saponi, Cosmet., Aerosol* **49,** 86.

Dewin, H. (1955). Natural and synthetic pepper substances (problems of constitution synthesis in relation to pepper flavour). *Seifen, Oele, Fette, Wachse* **81,** 489 and 511; *Chem. Abstr.* **50,** 8936a (1956).

Dhar, K. L., and Atal, C. K. (1967). Occurrence of N-isobutyldeca-trans-2-trans-4-dienamide in *Piper longum* Linn. and *Piper populoides Royale. Indian J. Chem.* **5,** 588.

Dhaul, R. A., Rao, K. M., and Bhave, V. M. (1958). Examination of the essential oil from siyah zeera (*Carum bulbocastanum*). *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 120.

Dhesi, N. S., Randhawa, K. S., and Nandpuri, K. S. (1966). Losses in weight of onions during storage as influenced by certain plant grown regulators. *J. Res., Ludhiana* **3,** 23.

Dhingra, S. N., Dhingra, D. R., and Gupta, G. N. (1954). Essential oils of *Ocimum* basilicum. *Indian Soap J.* **19,** 251.

Diemair, W., and Bauierh, H. (1964). Interference in the determination of sulfurous acid in mustard oil containing products. *Z. Lebensm.-Unters.- Forsch.* **124,** 109.

Directorate of Arecanut and Spices Development, Government of India, Calicut (1970). "Turmeric," Farmers' Bull. No. 2. Dept. Ext., Minist. Agric. & Food, Govt. of India, New Delhi.

Directorate General of Commercial Intelligence and Statistics (1979). "Foreign Trade of India," DGCIS Sect. 81, p. 6. Govt. of India.

Directorate of Marketing and Inspection (1957). "Report on the Marketing of Chillies in India," Market. Ser. No. 83, pp. 1–232. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1965a). "Report on the Marketing of Cardamom in India," Market. Ser. No. 144, pp. 1–134. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1965b). "Report on Marketing of Turmeric in India," Market. Ser. No. 148, pp. 1–233. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1966a). "Report on Marketing of Ginger in India," Market. Ser. No. 151, pp. 1–74. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1966b). "Report on Marketing of Onion in India," Market. Ser. No. 157, pp. 1–72. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1968a). "Report on Marketing of Minor Spices in India," Market. Ser. No. AMA 163, pp. 1–288. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1968b). The present system of marketing, grading and quality control of chilies for export. *Indian Spices (Chilies Semin. No.)* **6** (3), 28.

Directorate of Marketing and Inspection (1970a). "Instructions for Grading and Marketing of Ground Spices under Agmark," Market. Ser. No. 173, pp. 1–87. Manager of Publications, Delhi.

Directorate of Marketing and Inspection (1970b). Marketing of ginger and its quality control in India. *Indian Spices* **7** (2), 5–9.

Directorate of Marketing and Inspection (1971). "Report on Marketing of Pepper in India," Market. Ser. No. 171, pp. 1–177. Manager of Publications, Delhi.

Divakar, N. G., Nagarajarao, U. N., and Hartman, C. P. (1974). Detection of ragi (*Eleusine coracana*) in mustard seeds (*Brassica nigra*). *Curr. Sci.* **43,** 529.

Dmitrieva, L. L., Bychkova, Z. N., and Deeva, I. D. (1967). Stability of citral from coriander oil. *Maslo-Zhir. Prom-st.* **33,** 30.

Dobrynina, T. I. (1958). Effect of combined action of low temperatures and gama-irradiation of the activity of certain enzymes of vegetable products. *Nov. Fiz. Metody Obrab. Pishch. Prod.* **2,** 51.

Dold, H., and Knapp, A. (1948). The antibacterial action of spices. *Z. Hyg. Infektionskr.* **127,** No. 2, 51.

Doring, F. (1952). Spice concentrate. German Patent 828,343.

Dranitsyna, Yu. A. (1961). Two oil plants of the family Umbelliferae from the Central Sayan Mountains. *Tr. Bot. Inst., Akad. Nauk SSSR, Ser. 5* No. 9257.

Du, C. T., and Francis, F. J. (1975). Anthocyanins of garlic (*Allium sativum* Linn). *J. Food Sci.* **40,** 1101.

Du, C. T., Wang, L. L., and Francis, F. J. (1974). Cyanidin-3-laminariobioside in Spanish red onion (*Allium cepa L*). *J. Food Sci.* **39,** 1265.

Du, C. T., Wang, P. L., and Francis, F. J. (1975). Anthocyanins of pomegranate. *J. Food Sci.* **40,** 417.

Dublyanskaya, N. F. (1964). Fatty oils and essential oils in cultivated plants. *Maslob.-Zhir. Prom-st* **30,** 24.

DuBois, C. W., and Tressler, D. K. (1943). Seasonings, their effect on maintenance of quality in storage of frozen ground pork and beef. *Proc. Inst. Food Technol.* p. 202.

DuBois, H. L. (1949). Spices stand up in frozen storage. *Food Ind.* **21,** 116.

Duquenois, P. (1962). Standardization of vegetable drugs. IV. Comparison of methods for the determination of essential oil. *Ann. Pharm. Fr.* **20,** 43.

Duro, F. (1961). Volumetric determination of pipperine in *Piper nigrum* fruits. *Boll. Sedute Accad. Gioenia Sci. Nat. Catania* **7,** 120.

Dutta, A. B. (1961). A chemical method for distinguishing cinnamon from cassia. *J. Assoc. Off. Agric. Chem.* **44,** 639.

Dwarkanath, C. T., and Ramachandra Rao, T. N. (1963). Chemical analysis of pepper varieties, stalks and musks. *Spices Bull. (Annu. No.)* **2,** 21.

Dwarkanath, C. T., Ramachandra Rao, T. N., and Johar, D. S. (1958). Chemical analysis of some varieties of Indian pepper (*Piper nigrum*). *Food Sci.* **7,** 285.

Dwarkanath, C. T., Ramachandra Rao, T. N., and Johar, D. S. (1959). Chemical analysis of trade grades and by-products of pepper (*Piper nigrum*). *Food Sci.* **8,** 351.

Dwarkanath, C. T., Ramachandra Rao, R. N., and Johar, D. S. (1961). Chemical analysis of some varieties of Indian pepper (*Piper nigrum*). Part II. *Food Sci.* **19,** 1.

Dytkowska, C. (1964). Heavy metal content of certain plant materials. *Acta Pol. Pharm.* **21,** 497.

Egan, H., and Laws, E. Q. (1962). The analysis of pesticide residues in foods. A review of recent developments in the analytical methods. *Proc. Br. Insectic. Fungic. Conf., 1961* p. 395.

Eisenberg, W. V. (1949). Report on extraneous material in drugs, spices, and miscellaneous materials. *J. Assoc. Off. Agric. Chem.* **32,** 319.

Eisenberg, W. V. (1961). Determination of light filth in ground cinnamon. *J. Assoc. Off. Agric. Chem.* **41,** 336.

Eiserle, R. J. (1966). The role of oleoresin turmeric in the pickling process. *Glass Packer/Process.* **45,** 48.

Eisissi, H. 1., Saheh, N. A. M., Elsherbeiny, A. E. A., and Elansary, M. A. I. (1965). Local plants as potential sources of tannins and the isolation of their free and combined sugars. *Qual. Plant. Mater. Veg.* **12,** 262 (in English).

El-Deeb, S. R., Karawya, M. S., Wahba, S. K., and Edeid, F. M. (1962a). The chemistry of oil of caraway. *J. Pharm. Sci. U.A.R.* **3,** 63.

El-Deeb, S. R., Karawya, M. S., and Wahba, S. K. (1962b). Chromatographic analysis of caraway and lemon oils. *J. Pharm. Sci. U.A.R.* **3,** 81; *Chem. Abstr. 60,* 6697f (1964).

El-Hamidi, A., and Ahmed, S.S. (1966). Content and composition of some umbelliferous essential oils. *Pharmazie* **21,** 438.

El-Hamidi, A., and Richter, G. (1967). Preliminary investigation of Egyptian cumin oil by thin-layer chromatography. *Lloydia* **28,** 252; *Chem. Abstr.* **63,** 1779lh (1965).

Ellis, A. (1960). "The Essence of Beauty—A History of Perfume and Cosmetics." Macmillan, New York.

Elvehjem, C. A., and Burns, C. H. (1952). *J. Am. Med. Assoc.* **148,** 1033.

Embong, M. B., Hadziyev, D., and Molnar, S. (1977a). Essential oils from herb and spices grown in Alberta. Sage oil, *Salvia officinalis (Labiatae). Can. Inst. Food Sci. Technol. J.* **10,** 201.

Embong, M. B., Hadziyev, D., and Molnar, S. (1977b). Essential oils from spices grown in Alberta. Dill oil, *Anethum graveolens,* L. (Umbelliferae). *Can. Inst. Food Sci. Technol. J.* **10,** 208.

Endo, S., and Tanji, K. (1960). Physiological chemistry of *Acorus gramineus.* V. Studies on amino acids and sugar components. *Bot. Mag.* **73,** 427.

Eolkin, D., and Bouthilet, R. J. (1967). Sterilizing dry particulate material. U.S. Patent 3,341,335 (C.99-150).

Epshtein, M. M., Bogatsakaya, L. N., and Shchigol, M. B. (1959). Biochemical changes in nervous tissues by the action of substances reacting with sulfhydryl compounds. *Tiolovye Soedin. Med., Tr. Nauchn. Konf., 1957* p. 72.

Ertan, U., and Duvekot, W. S. (1969). A Study on the storage of onions grown in the Marmara region. *Yalleva-Bahco Kelt. Arast. Egit. Merk. Derg.* **2,** 32 (English summary).

Eschenmoser, A., and Schinz, H. (1950). Constitution of zingiberene. *Helv. Chim. Acta* **33,** 171.

Esses, C. D., and Schelton, J. H. (1956). Food microscopy. Parts 4 to 9. Spices used in flour confectionary. *Food* **25,** 138, 184, 225, 266, 307, and 345.

Essential Oil Association (1965). "EOA Specifications and Standards." Essent. Oil Assoc., New York.

Fabian, F. W., Kreh, C. F., and Little, N. W. (1939). Role of spices in pickled food spoilage. *Food Res.* **4,** 269.

Fagen, H. J., Kolen, E. P., and Hussong, R. V. (1955). Spectrophotometric method for determining piperine in oleoresins of black pepper. *J. Agric. Food Chem.* **3,** 860.

Faigle, H., and Karrer, P. (1961). Constitution and configuration of capsanthin and capsorubin. *Helv. Chim Acta* **44,** 1257.

Farber, L. (1949). Chemical evaluation of odour intensity. *Food Technol.* **3,** 390.

Farber, L. (1957). The chemical evaluation of pungency of onions and garlic by the content of volatile reducing substances. *Food Technol.* **11,** 621.

Farooq, M. O. (1958). Chemical examination of some less familiar essential oils. *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 130.

Favro, L. (1968). The evolution of essential oil production in Morocco. *Proc. Int. Conf. Essent. Oils, Trop. Prod. Inst. London, 1967* p. 13.

Fazli, F. R. Y., and Hardman, R. (1968). The spice, fenugreek (*Trigonella foenumgraecum*) its commercial varieties of seed as a source of diosgenin. *Trop. Sci.* **10,** 66.

Fazlullah Khan, K. (1967). "Cultivation of Cinnamon," Farmers' Bull. (New Ser.), No. 40. Indian Coun. Agric. Res., New Delhi.

Fefer, I. M. (1959). Liquid part of edible oil from fennel seeds. *Farm. Zh. (Kiev)* **14,** 61.

Fernandes Costa, A., and Cardoso Do Vale, J. (1959). Observations in the study of portugese aromatic plants; essential oil of *Foeniculum vulgare* Mill. *Perfum. Essent. Oil Rec.* **50,** 291.

Fernandez Pizarro, M. I., and Samper, M. N. (1953). Saffron and its adulterants. *An. Bromatol.* **5,** 349.

Ferns, R. S. (1961a). Extraction of oleoresin from pimiento (*capsicum*). *Galenica Acta* **13,** 381.

Ferns, R. S. (1961b). Solvents for the extraction of oleoresin from pimiento (*capsicum*). *Galenica Acta* **13,** 391.

Ferrara, A. (1958). Technology of spices: Cinnamon and ginger. *Riv. Agric. Subtrop. Trop.* **52,** 50.

Ferre, L. (1925). Action of allyl mustard oil as wine and must preservative. *Ann. Falsif. Fraudes* **18,** 34; cf. *Chem. Abstr.* **19,** 1323 (1925).

Fibranz, L., Blake, M. I., and Miller, C. E. (1958). Colorimetric determination of thymol in thyme oil. *J. Am. Pharm. Assoc., Sci. Ed.* **17,** 133.

Ficicchia, F. (1966). Chromatographic separation and determination of essences in mixtures and emulsions. *Corr. Farm.* **21,** 330; *Chem. Abstr.* **66,** 68831k (1967).

Firstenberg, R., Mannheim, C. H., and Cohen, A. (1974). Microbiological quality of dehydrated onions. *J. Food Sci.* **39,** 685.

Fischer, R. (1965). Aging of linseed and poppy seed groats. I. Introduction and literature about oxidative fat spoilage through aging. II. Further literature report on oxidative fat spoilage through aging and on the influence of radiation of fats. *Dtsch. Apoth.-Ztg.* **105,** 173 and 290.

Fitelson, J. (1960). Method used for F.E.M.A. Collaborative studies on vanilla resins. *Proc. Annu. Conv. Flavoring Extr. Manuf. Assoc. U.S.* **51,** 58.

Fitelson, J. (1961). The detection of foreign plant material in vanilla extract. *J. Assoc. Off. Agric. Chem.* **44,** 531.

Fitelson, I. (1962a). Paper chromatography of the organic acids of vanilla extracts. *J. Assoc. Off. Agric. Chem.* **45,** 246.

Fitelson, J. (1962b). Qualitative test for vanilla resins. *J. Assoc. Off. Agric. Chem.* **45,** 250.

Fitelson, J. (1964). Organic acids in vanilla extract. *J. Assoc. Off. Agric. Chem.* **47,** 558.

Fitelson, J. (1965). Analysis of vanilla powders. *J. Assoc. Off. Agric. Chem.* **48,** 911.

Fitelson, J. (1967). Further collaborative studies on methods for vanillin and ethyl vanillin in flavouring material. *J. Assoc. Off. Anal. Chem.* **50,** 859.

Fitzpatrick, F. K. (1954). Plant substance active against *Mycobacterium tuberclosis. Antibiot. Chemother. (Washington, D.C.)* **4,** 528.

Flick, W. (1949). Sterilization in high pressure autoclaves. *Ind. Obst-Gemueseverwert.* **34,** 102.

Food and Agriculture Organization (1962). "Spices: Trends in World Markets," Commodity Bull. Ser. No. 34. F.A.O., United Nations, Rome.

Food and Agriculture Organization (1968). The 1967 "Evaluation of Some Pesticide Residues in Foods," Monogr. F.A.O., U.N.O., Rome, and World Health Organization, Rome. 1–224.

Food and Agriculture Organization/World Health Organization Joint Expert Committee (1968). "Report of the 1967 Joint Meeting of the FAO Working Party of Experts on Pesticide Residue and the WHO Expert Committee on Pesticide Residues," No. OL:1967/M/11-No.391, pp. 1–43. FAO,UNO, Rome.

Food Processing Staff (1960). Longer storage life for spices. *J. Food Process.* p. 44.

Forrest, J. E. and Heacock, R. A. (1972). Identification of the major components of the essential oil of mace. *J. Chromatgr.* **69,** 115.

Foter, M. J. (1940). Bactericidal properties of allyl isothiocyanate and related oils. *Food Res. 5,* 147.

Frank, A. M. (1942). "German Spices and their Effect on Gastric Secretion." Med. & Nutr. Council of the Inst. For Culinary Art of H.Q. Staff of Herman, Esser, Hosp. of Holy Ghost and Franciscan Klinic.

Frank, A. M. (1961). Purpura resulting from artificial abortion. *Dtsch. Med. Wochenschr.* **86,** 1618.

Franklin, D. F., Works, D. W., and Williams, L. G. (1966). Experiments in harvesting, curing and storing yellow sweet Spanish onions. *Idaho, Agric. Exp. Stn.* **479,** 35.

Freeman, G. G. (1975). Distribution of flavour components in onion (*Allium cepa* L.), Leek (*Allium porrum*) and Garlic (*Allium sativum*). *J. Sci. Food Agric.* **26,** 471.

Freeman, G. C., and Whenham, R. J. (1974a). Changes in onion flavour components resulting from some post-harvest processes. *J. Sci. Food Agric.* **25,** 499.

Freeman, G. G., and Whenham, R. J. (1974b). Flavour changes in dry bulb onions during overwinter storage at ambient temperature. *J. Sci. Food Agric.* **25,** 517.

Freeman, G. G., and Whenham, R. J. (1975a). The use of synthetic (+)-S-1-propyl-L-cysteine sulphoxide and of alliinase preparations in studies of flavour changes resulting from processing of onion (*Allium cepa* L). *J. Sci. Food Agric.* **26,** 1333.

Freeman, G. G., and Whenham, R. J. (1975b). A survey of volatile components of some allium spices in terms of S-alk(en)yl-l-cysteine sulphoxides present as flavor precursors. *J. Sci. Food Agric.* **26,** 1869.

Freeman, G. G., and Whenham, R. J. (1976). Nature and origin of volatile flavour components of onion and related species. *Int. Flavours Food Addit.* **7,** 222.

Friedrich, H., and Rangoonwala, R. (1965). Separation of capsaicin and non-anoic acid vanillyl amide. *Naturwissenschaften* **52,** 514.

Fujita, M., Furuya, T., and Kawana, A. (1954). Japanese capsicum. I. Colorimetric determination of capsaicin by paper partition chromatography. *J. Pharm. Soc. Jpn.* **74,** 766.

Fungairifio, L. V., Samper, M. N., Portoles, A., and Pizano, F. (1954). Analysis of condiments. *An. Bromatol.* **191,** 399.

Gabel, E., Mueller, K. H., and Schnoknecht, I. (1962). Analysis of herbage, oil and tincture of thyme. *Dtsch. Apoth.-Ztg.* **102,** 293; *Chem. Abstr.* **57,** 953a (1962).

Gaind, K. H., Dar, R. N., and Popli, S. D. (1965). Spectrophotometric estimation of alliin in allium species. *Indian J. Pharm.* **27,** 199.

Gal, I. E. (1972). Extraction of the antibiotic capsaicidin from mature capsicum seeds by a simple method. *Z. Lebensm.-Unters.-Forsch.* **148,** 286.

Galetto, W. G., and Bednarzyk, A. A. (1975). Relative flavor contribution of individual volatile components of the oil of onion (*Allium cepa*). *J. Food Sci.* **40,** 1165.

Galetto, W. G., and Hoffman, P. G. (1976a). Synthesis and flavour evaluation of some alkylthiophenes volatile components of onion. *J. Agric. Food Chem.* **24,** 852.

Galetto, W. G., and Hoffman, P. G. (1976b). Synthesis and flavour evaluation of several alkylfuranones found in Allium species (onions, shallots and leeks). *J. Agric. Food Chem.* **24,** 854.

Galperin, L. M. M., and Dunaeva, V. F. (1954). The effect of volatile oils and of some of their fractions on the catalase of paranocia and helminths. *Ukr. Biokhim. Zh.* **26,** 454 (cited by Maruzzella, 1962).

Galperin, L. M. M., and Dunaeva, V. F. (1955). Effect of volatile oils and their fractions on the dehydrogenase activity of paramecia and ascrids. *Ukr. Biokhim. Zh.* **27,** 197.

Gandhi, A. D. (1967). Pepper export and export markets. *Indian Spices* **3,** 8.

Gandhi, J. M. (1968). Diversification of exports for (a) new markets, and (b) new products. *Spices Bull. (Chillies Semin.)* No. 6, p. 68.

Gandhi, J. M. (1974). Marketing trends and potential for exports growth for important Indian spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 70.

Garcia, F. M. (1959). Paprika quality determination by its capsanthin and capsorubin content. *An. Fac. Farm. Porto* **19,** 5.

Garcilase, S. G. (1961). Microdetermination of cobalt in native Venezuelan foods. *Arch. Venez. Nutr.* **11,** 109.

Garnero, J., and Chrétien-Bessiere, Y. (1968). Contribution to the study of chemical composition of parsley leaf oil from Yugoslavia. *Fr. Parfums* **11** (60), 332.

Gastineau, F. M. (1941). Occupational dermatitis. *J. Indiana State Med. Assoc.* **34,** 184.

General Medical Council (1963). "British Pharmacopoeia," p. 1062. Pharmaceutical Press, London.

Genest, C., Smith, D. M., and Chapman, D. G. (1963). A critical study of two procedures for the determination of piperine in black and white pepper. *J. Agric. Food Chem.* **11,** 508.

Genova, E., and Balinov, A. (1970). Studies on the essential oil content of certain spices of the genera Saturcia and Calamintha. *Izv. Bot. Inst., Bulg. Akad. Nauk.* **20,** 151 (Ref. 13: Bulgarian, with English summary.).

George, M., and Pandalai, K. M. (1958). Antibacterial activity of a few common essential oils. *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 154.

Georgiev, E. (1965). Changes in essential oils of anise fruits under normal storage conditions. *Wiss. Z., Karl-Marx-Univ., Leipzig, Math.-Naturwiss. Reihe* **14,** 447.

Gerhardt, U. (1968). Routine analysis of volatile oils and other materials in spices—I. Pepper, paprika, mace and pimiento. *Fleischwirtschaft* **48,** 1207.

Gerhartdt, U. (1969). Entkeimung von gewiirzen. *Gordian* **69** (9), 427 (cited by Heath, 1972).

Gholap, A. S., Tewari, G. M., and Bandhyopadyay, C. (1974). Isolation and characterization of onion wax. *J. Food Sci. Technol.* **11,** 289.

Gibbs, H. D. (1927). Phenol tests. III. The indophenol test. *J. Biol. Chem.* **72,** 649.

Gilbertson, G. (1970). Evaluation of oleoresins. *Flavour Ind.* **1,** 459 (rev.).

Gilbertson, G. (1971). Oleoresins as flavour ingredients. *Flavour Ind.* **2,** 403.

Gildermeister, E., and Hoffmann, F. (1956). "Die Aetherischen Oele," 4th ed., Vol.1, p. 307. Adademie-Verlag, Berlin.

Gildermeister, E., and Hoffmann, F. (1956–1961). "Die Aetherischen Oele," 4th ed., Vols. 4–7. Akademie-Verlag, Berlin.

Gill, L. S., Lawrence, B. M., and Morton, J. K. (1973). Variation in *Mentha arvensis* L (Labiatae) I. The North American populations. *Bot. J. Linn. Soc.* **67,** 213.

Gisske, W. (1954). Sterile spice preparation for the preparation of meat dishes and sausage goods. *Fleischwirtschaft* **6,** 280.

Gisske, W., and Heidtmann, R. (1958). Liquid spice extracts: Mixture of—for Frankfurter type of sausages and cooked sausages. *Fleischwirtschaft* **10,** 87.

Gladkikh, A. S., Gubano, U.I.A., and Meshcheryakov, A. A. (1965). Content of saponins in plants of Turkmenia. *Izv. Akad. Nauk Turkm. SSR, Ser. Biol. Nauk* **1,** 22.

Glatzel, H. (1965). Spices and organ function. Results of recent studies. *Muench. Med. Wochenschr.* **107,** 332.

Glatzel, H. (1968a). Physiological aspects of flavouring compounds. *Indian Spices* **5** (2), 13.

Glatzel, H. (1968b). "Die Gevviirze: Ihre Wirkungan 'auf den Gesunden Und' Kranken Menschen," 1st ed. Nicolaische Verlagsbuchhandlung, Herford, Germany.

Glatzel, H., and Hackenberg, K. (1966). X-ray investigation on the action of capsicums and mustard on the motoric and secretary organs. *Dtsch. Z. Verdau.-Stoffwechselkr.* **26,** 114.

Glatzel, H., and Rettenmaier, G. (1965). Kruslarifregulation und Frnahrung. *Z. Kreislaufforsch., B* **54,** No. 10, 63.

Gletto, C. A. P. (1973). Synthetic onion oil. U.S. Patent 3,764,709 (W. G. Galetto, C. A. Pace; McCormic and Co., Inc.).

Gmelin, R. (1959). Biochemical Institute, Helsinki (private communication to Schwimmer and Wetson, 1961).

Goerling, P. (1962). Influence of storage conditions on quality conservation of dried products II. Studies on storage character of air-dried potatoes, green beans, onions, green kale, white leek and freeze-dried chicken meat. *Ind. Obst.- Gemueseverwert.* **47,** 673 and 703.

Gogrof, G. (1957). The volatile oil sesquiterpenes and glandular trichomes of patchouli (Pogostemon spices). *Pharmazie* **12,** 38; *Chem. Abstr.* **51,** 18488h (1957).

Gold, H. J., and Wilson, C. W. (1963a). The volatile flavour substances of celery. *J. Food Sci.* **28,** 454.

Gold, H. J., and Wilson, C. W. (1963b). A colorimetric ester test as a measure of total flavour in celery. *Proc. Fla. State Hortc; Soc.* **76,** 240.

Goldman, A. (1949). How spice oleoresins are made. *Perfum. Essent. Oil Rec.* **53,** 320.

Goldovskii, A. M. (1962). The principal types of quantitative interrelationships for the fatty acids in vegetable oils. *Maslob.-Zhir. Prom-st.* **28** (8), 10.

Goleiz, K. (1969). *Med. Weter.* **23,** 362. (cited by Krishnaswamy *et al.,* 1971).

Gorsica, H. J. (1959). Stabilized seasoning compositions. U.S. Patent 3,008,832 (B. Heller & Co.).

Goryaev, M.I., Sharipova, F. S. Tikhonova, L. K., and Elchibekova, L. A. (1967). Analysis of mint essential oil. *Moslob.-Zhir. Prom-st.* **33** (9), 27.

Gosh, A. K., and Rajagopal, K. (1951). Riboflavin content of some common Indian foods. *Indian J. Med. Res.* **39**, 473.

Gough, I. A., and Goodhead, K. (1975). Occurrence of volatile nitrosamines in spice pre-mixes. *J. Sci. Food Agric.* **26**, 1473.

Gould, R. F., ed. (1966). "Flavour Chemistry," Adv. Chem. Ser. No. 56. Am. Chem. Soc., Washington, D. C.

Govindarajan, V. S. (1973). Evaluation of spices and oleoresins. *J. Plant. Crops* **1**, Suppl., 195.

Govindarajan, V. S. (1977). Pepper: Chemistry, technology and quality evaluation. *Crit. Rev. Food Sci. Nutr.* **9**, 115.

Govindarajan, V. S., and Ananthakrishna, S. M. (1970). Observations on the separations of capsaicin from capsicum and its oleoresin. *J. Food Sci. Technol.* **7**, 212.

Govindarajan, V. S., and Ananthakrishna, S. M. (1974). Paper chromatographic determination of capsaicin. *Flavour Ind.* **5**, 176.

Govindarajan, V. S., and Raghuveer, K. G. (1973). Evaluation of spice oils and oleoresins Part 1. A three-dimentional procedure for thin layer chromatography of essential oils and oleoresins. *Lab. Pract.* **22**, 414.

Govindarajan, V. S., Dhanaraj, S. and Shanthi, N. (1973). Evaluation of spices and oleoresins. III. Evaluation of some horticultural varieties and trade types of pepper. *J. Plant. Crops* **1**, 8.

Govindarajan, V. S., Narasimhan, S., and Dhanaraj, S. (1977). Evaluation of spices and oleoresins. II. Pungency of capsicums by Scoville heat units—a standardized procedure. *J. Food Sci. Technol.* **4**, 28.

Graf, E., and Hoppe, W. (1964). Ingredients and preparation of rosemary. *Dtsch. Apoth.-Ztg.* **104**, 287; *Chem. Abstr.* **61**, 1703e (1964).

Graham, H. D. (1965a). Reaction of piperine with nitric acid. Adaptation to quantitative assay of the piperine content of pepper. *J. Pharm. Sci.* **54**, 319.

Graham, H. D. (1965b). Quantitative determination of piperine. I. Komarowsky reaction. *J. Food Sci.* **30**, 644.

Graham, H. D. (1965c). Quantitative determination of piperine with the Labat reagent. *Nature (London)* **207**, 526.

Graham, H. D. (1965d). Quantitative method of piperine. II. Direct determination with phosphoric acid. *Nature (London)* **207**, 651.

Granger, R., Passet, C., and Verdier, R. (1963). Variety of essential oils of *Thymus vulgaris* L. *Parfums* **6**, 225.

Granger, R., Passet, J., and Verdier, R. (1965). Linalool in the *Thymus vulgaris* L. Oil. *C.R. Hebd. Seances Acad. Sci.* **260**, 2619.

Granger, R., Passet, J., and Arbousset, G. (1970). Optical activity of the oil of *Rosmarinus officinalis* L. *Fr. Parfums* **13**, 62.

Granroth, D. B. (1970). Biosynthesis and decomposition of cystein derivatives in onion and other Allium species. *Soum. Kemistil. A* **154** 71.

Gray, D. M., and Southwick, C. A., Jr. (1929). Scientific research affords means of measuring stability of mayonnaise. *Food Ind.* **1**, 300.

Greenstock, D. D. (1970). "Garlic as a Pesticide." Henry Doubleday Res. Assoc., Braintree, England.

Griebel, C., and Hess, G. (1940). The keeping qualities of ground spices. *Z. Unters. Lebensm.* **79**, 184.

Grieve, M. (1933). "Culinary Herbs and Condiments." Med. Books, London.

Grimm, A. I. (1959). Use of antiseptics and antibiotics during storage of carrots. *Sb. Leningr. Inst. Sov. Torg. F. Engle Sci.* **5**, 17.

Grimme, C. (1953). The maintenance of condiment capacity of spices on fine grinding. *Z. Lebensm.-Unters.-Forsch.* **97,** 191.

Grimme, C. (1954a). The binding capacity of amulin for volatile oils. *Z. Lebensm.-Unters.-Forsch.* **98,** 440.

Grimme, C. (1954b). Preservation of seasoning strength of spices during fine grinding. *Brot Gebaeck* **8,** 118.

Gross, A. F., and Ellis, P. E. (1969). Lipase activity in spices and seasonings. *Cereal Sci. Today* **14,** 332.

Grove (1918). *Annu. Rep. Agric. Hortic. Res. Stn., Long Ashton, Bristol* (cited by Corran and Edgar, 1933).

Grunsteidl, E. (1933). Examination of spices by means of luminescence microscopy 1. Fruit spices. *Z. Unters. Lebensm.* **63,** 425.

Grunsteidl, E., and Stobicke, J. (1936). New microstatic methods for the detection of adulterants of powders adulterated by the addition of powdered black pepper hulls. *J. Micro-kem. Festchr. von Hans Molisch.* p. 164; *Chem. Abstr.* **31,** 4450 (1937).

Guadagni, D. G., Buttery, R. G., and Okano, S. (1963a). Odour threshold of organic compounds associated with food flavours. *J. Sci. Food Agric.* **14,** 761.

Guadagni, D. G., Buttery, R. G., Okano, S., and Burr, H. K. (1963b). Additive effect of sub-threshold concentrations of some organic compounds associated with food aromas. *Nature (London)* **200,** 1288.

Guenther, E. (1943). Oil of coriander. *Drug & Cosmet. Ind.* **53,** 34.

Guenther, E. (1948–1952). "The Essential Oils," Vols. 1–VI. Van Nostrand-Reinhold, Princeton, New Jersey (reprinted in 1973).

Guenther, E., Kulka, K., and Rogers, J. A. (1959). Essential oils and related products. *Anal. Chem.* **31,** 679.

Guenther, E., Kulka, K., and Rogers, J. A. (1961). Essential oils and related products. *Anal. Chem.* **33,** 37R.

Guenther, E., Kulka, K., and Rogers, J. A. (1963). Essential oils and related products. *Anal. Chem.* **35,** 39R.

Guenther, E., Rogers, J. A., Jr., Gilbertson, G., and Koenig, R. T. (1965). Essential oils and related products. *Anal. Chem.* **37,** 46R.

Guenther, E., Burckhart, U., and Costinga, 1. (1966). Composition of pepper varieties. *Dtsch. Lebensm.-Rundsch.* **62,** 212.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1967). Essential oils and related products. *Anal. Chem.* **39,** 48R.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1969). Essential oils and related products. *Anal. Chem.* **41,** 40R.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1971). Essential oils and related products. *Anal. Chem.* **43,** 45R.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1973). Essential oils and related products. *Anal. Chem.* **45,** 45R.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1975). Essential oils and related products. *Anal. Chem.* **47,** 139R.

Guenther, E., Gilbertson, G., and Koenig, R. T. (1977). Essential oils and related products. *Anal. Chem.* **49,** 83R.

Guenther, L. (1966). Umbelliferous fruits in liquor industry. 111. Angelica fruits. Caraway. *Riechst., Aromen, Koerperpflegem.* **16,** 517.

Gujral, M. L., Varma, D. R., and Sareen, K. N. (1960). Oral contraceptives. Part 1. Preliminary observations on the anti-fertility. Effect of some indigenous drugs. *Indian J. Med. Res.* **48,** 46.

Gulati, B. C., and Duhan, S. P. S. (1976). The essential oil of *Mentha citata*. *Enrh. Riechst Arom. Korperflegem.* **26,** 180.

Gulati, B. C., Duhan, S. P. S., and Bhattacharya, A. K. (1967). Trial cultivations of Davana (*Artemisia pallens*) Wall. in Uttar Pradesh. *Perfum. Essent. Oil Rec.* **58,** 523.

Gulati, B. C., Duhan, S. P. S., and Bhattacharya, A. K. (1969). Quality of seed and herb oil produced from *Anethum graveolens* Linn. grown in Tarai of Uttar Pradesh. *Perfum. Essent. Oil Rec.* **60,** 277.

Gupta, G. N., and Patwardhan, V. M. (1957). Chemical examination of cumin oil. *J. Proc. Oil Technol. Assoc.* **12,** Parts II, III.

Gupta, G. N., Ganesh Chandra, and Zaidi, S. T. H. (1958). Chemical examination of dill seed and herb oils. *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 123.

Gupta, J. C., and Gupta, G. N. (1959). A study of peppermint oils from *Mentha piperita* and *Mentha arvensis*. *Indian Perfum.* **3,** Part I, 5.

Gupta, O. P., Atal, C. K., and Gaind, K. N. (1972). Occurrence of methyl piperate in *Piper officinarum* Cas. DC. *Indian J. Chem.* **10,** 874.

Gurvich, N. L., and Neparidze, N. I. (1966). Nephelometric determination of essential oils in small coriander samples. *Dokl. Akad. Nauk Az. SSR* **22** (7), 54 (in Russian).

Gusev, S. P., and Grishina, L. V. (1963). The chemical composition of different varieties of garlic. *Tr., Mosk. Inst. Nar. Khoz.* **24,** 30.

Hadorn, H. (1954). Determination of etherial oils in spices. *Mitt. Geb. Lebensmittelunters. Hyg.* **45,** 71.

Hadorn, H. (1962). Comparative estimation of (the method for) the determination of ethereal oils in condiments. *Mitt. Geb. Lebensmittelunters. Hyg.* **53,** 43.

Hadorn, H., Junqkunz, R., and Biefer, K. W. (1954). A new method for the determination of volatile oils in spices and drugs. *Mitt. Geb. Lebensmittelunters. Hyg.* **45,** 200.

Haffof, E. J. (1954). Dry solubles—versatile seasoning agents. *Food Eng.* **26**(6), 83.

Halbert, A., and Weeden, D. G. (1966). Lipase activity in black pepper. *Nature (London),* **212,** 1603.

Hall, J. R. (1969). Enhanced recovery of *Salmonella montevideo* from onion powder by addition of potassium sulfite to lactose broth. *J. Assoc. Off. Anal. Chem.* **52,** 940.

Hall, L. A. (1951). Ethylene oxide process reduces spoilage organisms. *Food Packer* **32** (12), 26.

Hall, R. L. (1955–1958). "Effect of Ionizing Radiations on Spices," Reports I–VIII. U.S. Army Quartermaster Corps. Contract *7-84-01-002 No. 28* McCormick & Co. Inc., New York.

Hamann, E. H. (1966). Flavors. *Kirk-Othmer Encycl. Chem. Technol., 2nd Ed.* Vol. 9.

Hamed, M. G. E., and Foda, Y. H. (1966). Freeze-drying of onions. *Z. Lebensm.-Unters. Forsch.* **130,** 220.

Hanc, O., and Santavy, F. (1946). Polarographic determination of piperidine in pepper *Cas. Cesk Lek.* **59,** 40; *Chim. Ind.* **58,** 178 (1947).

Handa, K. L., and Kapoor, L. D. (1958). Some aromatic plants of Jammu & Kashmir. *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 37.

Handa, K. L., Sharma, M. L., and Nigam, M. C. (1963a). The essential oil of *Piper longum*. Characterization of the constituents and isolation of two monocyclic sesquiterpenes. *Perfum. Kosmet.* **44,** 233.

Handa, K. L., Nigam, M. C., and Sharma, M. L. (1963b). Degradation studies on the sesquiterpenes from the oil of Piper longum. Perfum. Kosmet. **44,** 261.

Handa, K. L., Smith, D. M., Nigam, I. C., and Levi, L. (1964). Microscopical detection of spices and other plant elements in foods. *Z. Lebensm.-Unters.-Forsch.* **105,** 361.

Hanssen, E., Helenfeldt, R., and Florian, G.Z. (1957). Microscopical detection of spices and other plant elements in foods. *Z. Lebensm.-Unters.-Forsch.* **105,** 361.

Hanssen, E. B., Nielsen, B., and Utzinger, G. E. (1965). Opium alkaloids. II. Isolation and characterization of codamine. *J. Pharm. Sci.* **54** (10), 153.

Harald, H. (1919). Cloves of commerce. *Schweiz. Apoth.-Ztg.* **57**, 599.

Harborne, J. B. (1965). Plant polyphenols. XIV. Characterization of flavonoid glycosides by acidic and enzymic hydrolyses. *Phytochemistry* **4**, 107.

Harborne, J. B., and Saleh, N. A. M. (1971). Flavonol glycoside variation in fennel, *Foeniculum vulgare*. *Phytochemistry* **10**, 399.

Hardman, R. (1972). Spices and herbs, their families, secretary tissues and pharmaceutical aspects. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 23.

Hari Bhagwan, and Hathur, V. S. (1974). Standards for spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 42.

Harihara Iyer, C. R., and Sastri, B. N. (1933). The mucilage from Fenugreek (*Trigonella foenum-graecum*). *J. Indian Inst. Sci., Sect. A* **16**, 88.

Harrington, L. (1951). Spice of industry. *Food Can.* **11**, 24.

Harris, E. D., Sr. (1961). Control of some insects attacking celery in the Everglades. *Fla. Entomol.* **44**, 25.

Harrison, S., and Elder, L. W. (1950). Some applications of statistics to laboratory taste testing. *Food Technol.* **4**, 434.

Harth, V. (1943). German condiments and their influence on secretion of gastric juice. *Dtsch. Z. Verdau.-stoffwechselkr.* **6**, 263.

Hartman, K. T. (1970). A rapid gas liquid chromatographic determination of capsaicin in capsicum spices. *J. Food Sci.* **35**, 543.

Hasan, A., and Basha, M. K. A. (1932). Preliminary note on prolamin isolated from legume seed. *Biochem. J.* **26**, 1843.

Hassan, A., and Morgan, A. (1964). Direct semi-micro method for determination of fenugreek mucilage. *Egypt. Pharm. Bull.* **44**, 55.

Hasselstorm, T. (1957). Composition of volatile oil of black pepper—*Piper nigrum*. *Am. Perfum.* **70**, 43.

Hasselstorm, T., Hewitt, E. J., Konigsbacher, K. S., and Ratter, J. J. (1957). Composition of volatile oil of black pepper (*Piper nigrum* L). *J. Agric. Food Chem.* **5**, 53.

Hata, K., and Kozawa, M. (1961). Pharmacognostical studies on umbelliferous plants XVIII. Constituents of the roots of *Angelica keiskei*. *Yakugaku Zasshi* **81**, 1647.

Hattiangdi, G. S., and Nimbalkar, N. A. (1958). An evaluation of some indigenous perfume bearing materials. *Proc. Symp. Essent. Oils Aromat. Chem. 1958* p. 106.

Havinghorst, C. R. (1955). Continuous operations break batch—bottlenecks. *Food Eng.* **27** (5), 63.

Hayden, A., and Jordan, C. B. (1941). Report on the vanadium oxitrichloride colorimetric method for the detn. of capsaicin in capsicum. *J. Am. Pharm. Assoc.* **30**, 107.

Heath, H. B. (1964). Bacteria free spices in food processing. *J. Food Process. & Packag.* **33**, 144.

Heath, H. B. (1972). Herbs and spices for food manufacture. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 39.

Heath, H. B. (1973). Herbs and spices—a bibliography. *Flavour Ind.* **4**, 24, 65, 123, 169, 179, 217, 263, 346, and 384.

Heath, H. B., Fost, C. F., Macdonald, C. A., Middleton, A. J., Short, G. R., Smales, G. I., Wells, G. M., and Woodgate, A. J. (1964). The determination of capsaicin content of capsicum and its preparation. *Analyst (London)* **89**, 377.

Hefendehl, F. W. (1962). The use of gas chromatography for the analysis of the essential oil of the *Mentha piperita*. *Planta Med.* **10**, 179.

Hefendehl, F. W., Underhill, E. W., and Rudloff, E. V. (1967). The biosynthesis of the oxygenated monoterpenes in mint. *Phytochemistry* **6**, 823.

Heide, R. (1972). Qualitative analysis of the essential oil of cassia (*Cinnamomum cassia* Blume). *J. Agric. Food Chem.* **20,** 745.

Heintze, K. (1964). Polyphenols in vegetables. *Ind. Obst.- Gemueseverwert.* **49,** 471.

Hemingway, J. G., Schofield, H. J., and Vaughan, J. G. (1961). Volatile mustard oils of *Brassica juncea* seeds. *Nature (London)* **192,** 993.

Henderson, S. M. (1952). A basic concept of equilibrium moisture. *Agric. Eng.* **33**(1), 29.

Herndon, J. F., Touchstone, J. C., White, G. R., and Davis, C. M. (1963). Horizontal chromatography accelerating apparatus. Separation of dyes and indicators. *Anal. Chem.* **35,** 238.

Herrmann, K. (1962). Antioxidative action of seasonings and the labiatic acid (labiat tannin material) contained therein *Z. Lebensm.-Unters. -Forsch.* **116,** 224.

Heseltine, H. K., Hamilton, G. K., and Waller, J. B. (1953). Fumigation of grain in silo bins. *Pest Infest. Res., 1952* p. 29.

Heupke, W. (1932). Effect of spices on gastric secretion. *Dtsch. Arch. Klin. Med.* **172,** 583.

Heusser, D. (1964). Evaluation of various medicinal plants or their preparations with the help of thin-layer chromatography. *Planta Med.* **12,** 237.

Hewitt, E. J. (1963). Enzymatic enhancement of flavor. *J. Agric. Food Chem.* **11,** 14.

Hides Terado (1952). *Vitamins* **5,** 585 (cited by Krishnamurthy and Sreenivasamurthy, 1956).

Higgins, J. W., and Black, H. C. (1944). A preliminary comparison of the stabilizing effect of several recently proposed antioxidants for edible fats and oils. *Oil Soap (Chicago)* **21,** 277.

Hight, W. N., Pratt, D. E., and Powers, J. J. (1954). Thermal processes for pimientos canned in glass jars. *Food Technol.* **8,** 298.

Hoffmann, A., and Evans, A. C. (1911). The use of spices as preservatives. *Ind. Eng. Chem.* **3,** 835.

Hohmann, B. (1969). Cinnamon leaves in curry powder. *Z. Lebensm.-Unters. -Forsch.* **139,** 152.

Hollander, F. (1948). The pH of gastric secretion. *Am. J. Physiol.* **152,** 645.

Hollander, F., and Lauber, F. (1948). Eugenol as a stimulus for gastric mucous secretion. *Proc. Soc. Exp. Biol. Med.* **67,** 34.

Hollander, F., Lauber, F. U., and Stein, J. (1947). Some characteristics of gastric secretion induced by mustard oil suspension. *Am. J. Physiol.* **148,** 724.

Hollo, J., Jurucz, E., and Bodor, J. (1969). G.L.C. estimation of capsaicin in paprika oil. *Lebensm.-Wiss.-Technol.* **2,** 1.

Hooker Chemical Corporation (1967). Flame retardants for elastomers. The Netherlands Patent Appl. 6,612,481 (Cl. C. 08 d).

Hora, A. (1961). From vanilla beans to aro-vanillon. *Prum. Potravin* **12,** 629.

Horhammer, L., Wagner, H., and Lay, B. (1960). New methods in the teaching of pharmacognosy. 1. Detecting adulteration of *Pimpinella sascifraga* Root by *Heracleum sphondylium* Root by means of thin-layer chromatography. *Pharmazie* **15,** 645.

Horhammer, L., Wagner, H., and Eyrich, W. (1963). Contents of the fruit of *Angelica sylvestris*. *Z. Naturforsch. Teil B* **18** 639; *Chem. Abstr.* **62,** 6343g (1965).

Horhammer, L., El-Hamidi, A., and Richter, G. (1964a). Investigation of Egyptian basil essential oils by a simple chromatographic method *J. Pharm. Sci.* **53,** 1033.

Horhammer, L., Wagner, H., Richter, G., Konig, H. W., and Heng, (1964b). New methods in the teaching of pharmacognosy. IX. Thin-layer chromatography of pharmaceutically used umbilliferous oils. *Dtsch. Apoth.-Ztg.* **104,** 1398; *Chem. Abstr.* **62,** 1510e (1965).

Horhammer, L., Wagner, H., and Kraemerheydweiller, D. (1966). New methods in pharmacognostic instruction. XII.Identification of umbelliferous roots and detection of imp. falsification by means of thin-layer chromatography. *Dtsch. Apoth.-Ztg.* **106,** 267.

Hornstein, I., and Teranishi, R. (1967). The chemistry of flavour. *Eng. Ring News* p. 93.

Horose, V., Nishimura, K., and Sakai, T. (1960). The constituents of essential oils. I. Juniper berry oil. *Nippon Kagaku Zasshi* **81,** 1966.

Horst, P., and McGlumphy, J. H. (1962). Adulteration in vanilla extracts. *Ann. Falsif. Expert. Chim.* **55,** 264.

Hort, T. P. (1968). The essential oil content of *Thymus dzevanorskyi* in relation to condition of habitat. *Bull. Gos. Nikitsk. Bot. Sad* **1** (7), 36.

Horvath, I. (1932). Decrease of ethereal oil content of cinnamon during storage. *Kiserletugyi Kozl.* **35**, 114.

Howe, R. W. (1972). The susceptibility of the immature and adult stages of *Sitophilus granarius* to phosphine. *J. Stored Prod. Res.* **8**, 241.

Hu, T. Y., Huang, C. C., Wu, H. S., and Lu, F. H. (1964). Action of *Illicium majus* on the central nervous system excitation of cats, rats and mice. *Yao Hsueh Hsueh Pao* **2**, 521.

Huffman, V. L., Schadle, E. R., Villalon, B., and Burns, E. E. (1978). Volatile components and pungency in fresh and processed Jalapeno peppers. *J. Food Sci.* **43**, 1809.

Hunter, G. L. K., Lee, P. L., and Swords, G. (1975). Volatile constituents of tamarind (*Tamarindus indica* L). *J. Agric. Food Chem.* **23**, 1195.

Idel, F. (1976). Investigations of the aroma of fried onions. *Z Lebensm.-Unters. -Forsch.* **157**, 229.

Ikeda, R. M., Stainley, W. I., Vannier, S. H., and Spilor, E. M. (1962). The monoterpene hydrocarbon composition of some essential oils. *J. Food Sci.* **27**, 455.

Ilieva, S., and Zolotovitch, G. (1959). Research on sweet marjoram (*Majorana hortensis* Moench) cultivated in Bulgaria and the essential oil produced from it. *Perfum. Essent. Oil Rec.* **50**, 300.

Imlhoff, M. L. (1956). The chemical composition of essential oils of *Anothum graveolens. Fette, Seifen, Anstrichm.* **58**, 122.

Imlhoff, M. L. (1957). The composition of the volatile oils of *Anethum graveolens* L. in relation to various factors. *Food Sci. Abstr.* **29**, 1151.

Inagaki, S., and Ino, T. (1960). Pungent principle of red pepper. VI. Chemical constituents of the pungent principles—3 hydrogenation products, Bromine and decomposition products. *Nippon Nogei Kagaku Kaishi* **34**, 811.

Indian Council of Agricultural Research (1963). Report on "Coordinated Scheme on Essential Oil Bearing Crop Plants (1955–1960)." Indian Agric. Res. Inst. (ICAR), New Delhi.

Indian Council of Agricultural Research (1967). "Handbook of Agriculture," p. 264. ICAR, New Delhi.

Indian Institute of Foreign Trade (1967). "Cardamom-Seminar Conclusions and Recommendations." IIFT, New Delhi.

Indian Pharmaceutical Codex (1953). Council of Scientific & Industrial Research, New Delhi.

Indian Standards Institute (1973). "Methods of Sampling and Test for Spices and Condiments," Indian Stand. IS: 1797–1973 (rev.). ISI, New Delhi.

Indian Standards Institution (1976). "Methods of Test for Pungency of Chilies by Scoville Heat Units," Indian Stand. IS: 8104–1976. ISI, New Delhi.

International Organization for Standardization (1968). "Spices and Condiments. Nomenclature. ISO Recommendation 1st List." ISO, Budapest.

International Organization for Standardization (1970). "Specification for Dehydrated Onions and Garlic," ISO/TC 34-SC/7. ISO,

International Organization for Standardization (1971). "Spices and Condiments. Mustard Seed Specification," ISO/TC-34 (Secr. 554). Draft Recommendation No. 1237. ISO, Budapest.

International Organization for Standardization (1972). "Spices and Condiments, Nomenclature Finalized Draft Proposal," TC-34/SC-7 (Secretriat-107) 252E, 2nd list. ISO, Budapest.

International Organization for Standardization (1974). "Spices and condiments. Specification for Clove," ISO/TC-34/SC-7, D.R. 2254 (R2825, 1974). ISO, Budapest.

International Organization for Standardization (1979). The determination of trace residual solvents in oleoresins by gas chromatographic headspace analysis. Doc. No. ISO/TC 54/WG 2 N83, Oct. 1979.

International Trade Centre, Geneva (1974). "Markets for selected essential oils and oleoresins," pp. 1–297. International Trade Centre, United Nations Conference on Trade and Development/ General Agreement on Tariffs and Trade, Palais des Nations, CH-1211, Geneva-10, Switzerland.

International Trade Centre, United Nations Conference on Trade and Development/General Agreement on Tariffs and Trade, Geneva (1977a). "Spices—A Survey of the World Markets. Vol. 1, Selected Markets in Western Europe," pp. 1–281. International Trade Centre, UNCTAD/GATT, Palais des Nations, CH-1211, Geneva-10, Switzerland.

International Trade Centre, UNCTAD/GATT, Geneva (1977b). "Spices—A Survey of the World Markets. Vol. II, Selected Markets in North America, Asia, and the Pacific region, the middle East and the Socialist Countries of Eastern Europe," pp. 1–234. International Trade Centre, UNCTAD/GATT, Palais des Nations, CH-1211, Geneva-10, Switzerland.

Ishibashi, K., Katsuhara, J., Hashimoto, K., and Kobayashi, M. (1967). Components of star anise oil and fennel oil. *Kogyo Kagaku Zasshi* **70,** 1195.

Ishiguro, K. (1963). Pantothenic acid intake. 1. Pantothenic acid content in Japanese foods. *Tohoku J. Exp. Med.* **78,** 375.

Ito, M. (1954). Japanese mint. X. Sterioisomers of menthol. *Nippon Kagaku Zasshi* **78,** 1720.

Ito, M., Wakahatsu, S., and Kawahra, H. (1953). Japanese mint. VI. Chromatostrips of the constituents of Japanese Mint oil. *J. Chem. Soc. Jpn., Pure Chem. sect.* **74,** 699; Chem. Abstr. **48,** 3540b (1954).

Ito, M., Wakamatso, S., and Kawahra, H. (1954). Japanese mint. VII, Chromatostrips of the constitutents of Japanese peppermint oil. *J. Chem. Soc. Jpn., Pure Chem. Sect.* **75,** 413; *Chem. Abstr.* **48,** 13172d (1954).

Ivanov, D., Chorbadzhiev, S., and Jordanov, T. (1964). On the composition of Bulgarian basil oil. *Perfum. Essent. Oil Rec.* **55,** 717.

Izmailov, N. A., Mushinskaya, S. Kh., and Danel'Yants, V. A. (1963). Paper chromatography for controlling the manufacture of alkaloids from poppy capsules. III. Determination of narcotine, thebaine and papaverine. *Med. SSSR* **18**(3), 47; *Chem. Abstr.* **59,** 12594c (1963).

Jackson, H. W. (1966). Gas-chromatographic analysis of vanilla extracts. *J. Gas Chromatogr.* **4,** 196.

Jacobs, M. B. (1951). A study of the garlic flavour. *Am. Perfum. Essent. Oil. Rev.* **42,** 301.

Jacobs, M. B. (1958). "Chemical Analysis of Food and Food Products", 3rd ed. Van Nostrand-Reinhold, Princeton, New Jersey.

Jaffer Sait, A. R. (1974). Nutmeg cultivation. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 23.

Jager, H. (1955). Determination of allicin in fresh garlic. *Arch. Pharm. (Weinheim, Ger.)* **288,** 145; *Chem. Abstr.* **49,** 12772b (1955).

Jain, T. C., Varma, K. R., and Bhattacharya, S. C. (1962). Terpenoids. XXVIII. Gas-liquid chromatography of monoterpenes and its application to essential oils. *Perfum. Essent. Oil Rec.* **53,** 678.

James, A. T., and Martin, A. J. P. (1952). Gas-liquid partition chromatography. The separation and micro-estimation of volatile fatty acids from formic acid to dodecanoic acid. *Biochem. J.* **50,** 679.

James, L. H. (1931). Just how antiseptic are spices. *Food Ind.* **3,** 524.

James, L. H. (1938). Reducing microbial count of spices. *Food Ind.* **10,** 428.

Janaki, N., and Bose, J. L. (1967). An improved method for the isolation of curcumin from tumeric (*Curcuma longa*). *J. Indian. Chem. Soc.* **44,** 985.

Janke, A. (1923). The preservative effects of allyl mustard oil. *Fruit Prod. J.* **3,** 17.

Jansen, H., and Vander Hold, H. (1963). Gas chromatographic identification of rue oil and peppermint oil. *Med., Sci. Law* **3,** 77.

Jaspersen-Schib, R. (1961). Testing and standardisation of *Oleum menthal* by means of thin-layer chromatography. *Pharm. Acta Helv.* **36,** 141; *Chem. Abstr.* **55,** 15831f (1961).

Jaspersen-Schib, R., and Flueck, H. (1961). Identification and purity determination of essential oils by means of thin-layer chromatography. *Congr. Sci. Pharm., Conf. Commun., 21st, 1961* p. 608; *Chem. Abstr.* **60,** 1533f (1964).

Jaspersen-Schib, R., and Flueck, H. (1962). Identification of some essential oils by thin layer chromatography. *Boll. Chim. Farm.* **101,** 512.

Jaurenguiberry, G., and Wolff, R. (1962). The presence of (+) ≠ sabinene in the essential oil of nutmeg and some properties of this hydrocarbon. *Bull. Soc. Chim. Fr.* p. 1985.

Javela, B. O., and Rodriguez, Z. E. A. (1966). The rate and time of application of M.H. for checking vegetative growth and reproduction in onions. *Agric. Trop.* **22,** 493.

Jennings, W. G. (1969). Studies on oil of black pepper (*Piper nigrum*). *Abstr. Pap., 158th Meet., Am. Chem. Soc.* AGFD 36.

Jennings, W. G., and Wrolstad, R. D. (1961). Volatile constituents of black pepper. *J. Food Sci.* **26,** 499.

Jensen, A. (1960). Chromatographic separation of carotenes and other chloroplasts on paper containing aluminum oxide. *Acta Chem. Scand.* **14,** 2051.

Jensen, A., and Jensen, S. L. (1959). Quantitative paper chromatography of carotenoids. *Acta Chem. Scand.* **13,** 1863.

Jensen, W. V. (1971). Marketing of vanilla beans - *Vanilla plantifolia* and *V. tahitenis*. *Acta Hortic.* **21,** 158.

Jesson, L. B., Wood, I. H., and Jenson, C. E. (1934). Spoilage of canned chopped ham by organisms carried by spices. *Ind. Eng. Chem.* **26,** 1118 (cited by Baumgartner and Hersom, 1956).

Johnson, M. G., and Vaughn, R. H. (1969). Death of *Salmonella typhimurium* and *Escherichia Coli* in the presence of freshly reconstituted dehydrated garlic and onion. *Appl. Microbiol.* **17,** 903.

Joint Committee of the Pharmaceutical Society and the Society for Analytical Chemistry on Methods of Assay of Crude Drugs (1959). Recommended methods of assay of crude drugs. 1959. The determination of the capsaicin content of capsicum and its preparations. *Analyst (London)* **84,** 603.

Joint committee of the Pharmaceutical Society and the Society for Analytical Chemistry on Methods of Assay of Crude Drugs (1964). The determination of the capsaicin content of capsicum and its preparations. Second report of the panel. *Analyst (London)* **89,** 377.

Jones, E. C. S., and Pyman, F L. (1925). The relation between chemical constitution and pungency in acid amines. *J. Chem. Soc.* **127,** 2588.

Jones, H. A., and Mann, L. K. (1963). "Onions and Their Allies. Botany, Cultivation and Utilization," 1st ed. Leonard Hill, London.

Jones, J. K. N., and Thomas, G. H. S. (1961). Structure of the gum asafoetida polysaccharide *Can. J. Chem.* **39,** 192.

Joppien, P. H. (1961). Detection and determination of vanillin and ethyl vannillin in foods. *Z. Lebensm.-Unters. -Forsch.* **115,** 20.

Josef, Z. (1956). Determination of volatile oils and moisture in spices. *Prum. Potravi* **7,** 815.

Joshi, D. G. (1962). White pepper. Indian Patent 70,439.

Joslyn, M. A. (1970). "Methods in Food Analysis," 2nd rev. ed. Academic Press, New York.

Juhasz, K., Tyihak, E., Kerekes, J., and Udvardy, A. (1965a). Separation of essential oil from steam distillation residues and investigation of the components.II. *Lavandula officinalis* and *Mentha piperita* buds. *Riechst., Aromen, Koerperpflegem.* **15,** 248.

Juhasz, K., Udvardy, A., Tyihak, E., and Kerekes, J. (1965b). The recovery of the essential oils from the cohobation waters and investigation of the components. III. Recovery of the essential oil from the cohobation liquid. *Riechst., Aromen, Koerperpflegem.* **15,** 335.

Julseth, R. M., and Deibel, R. H. (1974). Microbial profile of selected spices and herbs at import. *J. Milk Food Technol.* **37,** 414.

Kaczmarek, F., Kowalewski, Z., Lutomski, J., and Wrocinski, T. (1961). Preparation of diuretic fractions from dried onion scales. *Biul. Inst. Rosl. Lecz.* **7,** 157.

Kaczmarek, F., Ostrowska, B., and Szpunar, K. (1962). Spasmolytic and diuretic activity of more important components of *Petroselinum sativum*. *Biul. Inst. Roslin. Lecz.* **8**, 111.

Kaess, G. (1950). Odorous substances passage through packaging materials and packaging of spices and aromatic foods. *Z. Lebensm.-Unters. -Forsch.* **90**, 101.

Kahan, G., and Stahl, W. H. (1973). Differentiation of geographic origin of spices. III. Lampong and Sarawak black pepper by emission spectroscopy. *J. Assoc. Off. Anal. Chem.* **56**, 341.

Kahan, S., and Fitelson, J. (1964). Chromatographic detection of flavour additives in vanilla extract. *J. Assoc. Off. Agric. Chem.* **47**, 551.

Kahan, S., and Fitelson, J. (1965). Chromatographic detection of flavor additives in vanilla extract. *J. Assoc. Off. Agric. Chem.* **48**, 507.

Kalra, R. L., Chawla, R. P., and Bindra, O. S. (1973). Significance of metabolites of insecticides in relation to tolerance. *Proc. Symp. Prog. Probl. Pestic. Residue Anal., 1971* p. 280.

Kalyanasundram, S., and Venkataratnam, K. P. (1965). Oil of rosemary—potentialities of large scale production. *Indian Oil Soap J.* **30**, 271.

Kanan, R. (1968). Marketing of chillies for export. *J. Agric. Market.* **10**(3), 13.

Kaneko, K. (1960a). Biogenetic studies of natural products. IV. Biosynthesis of anathole by *Foeniculum vulgare*. *Chem. Pharm. Bull.* **8**, 511.

Kaneko, K. (1960b). Biogenetic studies of natural products. V. Biosynthesis of anethole by *Foeniculum vulgare*. *Chem. Pharm. Bull.* **8**, 875.

Kaneko, K. (1961). Biosynthetic studies of natural products. VI. Biosynthesis of anethole by *Foeniculum vulgare*. *Chem. Pharm. Bull.* **9**, 108.

Kaneko, K. (1962). Biogenetic studies of natural products. VIII. Biosynthesis of anethole by *Foeniculum vulgare*. *Chem. Pharm. Bull.* **10**, 1085; *Chem. Abstr.* **58**, 5928g (1963).

Kannan, K., and Pillai, G. G. (1966). Vanilla—a new venture for Kerala farmers. *Indian Farming* **16**(2), 4.

Kanner, J., Mendel, H., and Budowski, P. (1976). Carotene oxidizing factors in red pepper fruits (*Capsicum annuum* L.). Ascorbic acid. *J. Food Sci.* **41**, 183.

Kanner, J., Mendel, H., and Budowski, P. (1977). Carotene oxidising factors in red pepper fruits: Peroxidase activity. *J. Food Sci.* **42**, 1549.

Kanner, J., Mandel, H., and Budawski, P. (1978). Carotene oxidising factors in red pepper fruits: Oleoresin-cellulose solid mode. *J. Food Sci.* **43**, 709.

Kapoor, L. D., and Abkol, B. K. (1961). Post-fertilization development study of dill fruit in relation to its oil content. *J. Sci. Ind. Res., Sect. C* **20**, 54.

Kapur, N. S., Narayanan, K. M., Bains, G. S., and Bhatia, D. S. (1957). Colorimetric determination of vanillin. *Chem. Ind. (London)* p. 1272.

Karaev, A. I., Aliev, R. K., Guseinov, G. A., and Dadashev, A. G. (1958). Effect of preparation of certain plants on sugar tolerance in animal organism. *Izv. Akad. Nauk. Az. SSR, Ser. Biol. S'kh. Nauk* No. 3, p. 81.

Karawya, M. S., and Balbao, S. I. (1967). A micro-method for the determination of capsaicin in capsicum fruits. *Analyst (London)* **92**, 581.

Karawya, M. S., and Wahba, S. K. (1961–1962). Chromatographic analysis of oil of peppermint. *Bull. Fac. Pharm., Cairo Univ.* **1**, 125; *Chem. Abstr.* **60**, 13092e (1964).

Karawya, M. S., and Wahba, S. K. (1962a). Analysis of oil of peppermint by an alluminium oxide—silicic acid double column. *J. Pharm. Pharmacol.* **14**, 611.

Karawya, M. S., and Wahba, S. K. (1962b). Chromatographic analysis of oil of eucalyptus and oil of rosemary. *J. Pharm. Sci. U. A. R.* **3**, 63; *Chem. Abstr.* **61**, 9351a (1964a).

Karawya, M. S., and Wahba, S. K. (1962c). Chromatographic analysis of anise, eucalyptus and rosemary. *J. Pharm. Sci. U. A. R.* **3**, 181; *Chem. Abstr.* **61**, 9351b, (1964b).

Karawya, M. S., and Wahba, S. K. (1962d). Chromatographic analysis of anise oil and sandalwood oil. *Egypt. Pharm. Bull.* **44**, 23; *Chem. Abstr.* **62**, 10288e (1965).

Karawya, M. S., and Wahba, S. K. (1967). Colorimetric estimation of eugenol in oils of clove, allspice and bay. *Am. Perfum. Cosmet.* **82,** 55.

Kardose, E. (1960). A study into the storage of red pepper. *Konzerv- Paprikaip.* **15,** 11.

Karel, M., Aikawa, Y., and Proctor, B. E. (1955). New approach to humidity equilibrium data. *Mod. Packag.* **29,** 213.

Karel, M., Proctor, B. E., and Cornell, A. (1957). Flavour permeability in food packaging and its evaluation. *Food Technol.* **11,** 141.

Karlsen, J., and Svenden, A. B. (1967). Constituents of Norwegian juniper oil. The sesquiterpene hydrocarbons. IV. Studies on terpenes and related compounds. *Medd. Nor. Farm. Selsk.* **29,** 13.

Karrer, P., and Jucker, E. (1950). "Carotenoids," p. 241. Am. Elsevier, New York.

Kartha, A. R. S., and Sharma, D. N. (1967). Detection of colophony in asafoetida (*Hing.*). *Res. Ind.* **12,** 248.

Karting, T. H. (1966). Some component lipids from the fruits of *Anethum graveolens* and *Coriandrum sativum*. *Fette, Seifen, Anstrichmi.* **68,** 131.

Karting, T. H., Wagner, H., and Hoerhammer, L. (1965). Some liquid containing materials from the root and fruit of *Foeniculum vulgare*. *Fette, Seifen, Anstrichmi.* **67,** 10.

Kashchenko, G. F., Akinov, Yu. A., Volchenkov, V. F., and Boitsov, E. N. (1971). Temperature conditions for the decantation of some essential oils. *Maslo-Zhir. Prom-st.* **37** (10), 24.

Kasimova, T. K. (1957). The production of medicinal media from the fruit and essential oil of the ajowan. *Tr. Tashk. Farm. Inst.* **1,** 212.

Kasting, R., Anderson, J., and von Sydow, E. (1972). Volatile constituents in leaves of parsley. *Phytochemistry* **11,** 2277.

Katayama, T. (1960). Chemical significance of the volatile components of spices in food preservation. V. A simple procedure for qualitative assay of spices. *Nippon Suisan Gakkaishi* **26,** 814.

Katayama, T., and Nagai, I. (1959a). Significance of the volatile components of spices from the food preservative viewpoint. II. Anti-bacterial activity of volatile components of coriander seed. *J. Fac. Fish. Anim. Husb., Hiroshima Univ.* **2,** 349.

Katayama, T., and Nagai, I. (1959b). Chemical significance of the volatile components of spices from the food preservative viewpoint. III. Antibacterial activity of volatile components of nutmeg. *J. Fac. Fish. Anim. Husb., Hiroshima Univ.* **2,** 355.

Katayama, T., and Nagai, I. (1960). Chemical significance of the volatile components of spices from the food preservative view point. IV. Structure and antibacterial activity of some terpenes. *Nippen Suisan Gakkaishi* **26,** 29.

Kato, S. (1965). Analysis of pigments in natural foods. I. Method for extraction, separation and absorption spectra. *Eisei Shikenjo Hokoku* **83,** 68.

Kato, T. (1966a). Physiological studies on the bulbing and dormancy of onion plants. VII. Effects of some environmental factors and chemicals on the dormant process of bulbs. *J. Jpn. Soc. Hortic. Sci.* **35,** 49.

Kato, T. (1966b). Physiological studies on the bulbing and dormancy of onion plants. VIII. Relation between dormancy and the organic constituents of the bulb. *J. Jpn. Soc. Hortic. Sci.* **35,** 297.

Kato, T. (1966c). Physiological studies on bulb formation and dormancy in onion plants. IX. The relationship between bulb dormancy and the components of the juice. *J. Jpn. Soc. Hortic. Sci.* **35,** 297.

Kato, T. (1970a). Studies on the mechanism of retardation of sprouting and occurrence of functional disorders in onion bulbs applied with maleic hydrazide. *J. Jpn. Soc. Hortic. Sci.* **39,** 177.

Kato, T. (1970b). Studies on the mechanism of retardation of sprouting and the occurrence of functional disorders in onion bulbs treated with maleic hydrazide. *J. Jpn. Soc. Hortic. Sci.* **39,** 178.

Katyal, S. L. (1967). Saffron cultivates well in hill areas. *Indian Hort.* **11**(3), 5.

Kaul, G. L., and Nigam, S. S. (1966). Indian essential oil. II. Colorimetry. *Perfum. Essent. Oil Rec.* **57,** 156.

Kawakishi, S., Namiki, K., Nishimura, M., and Namiki, M. (1971). Effect of gamma irradiation on the enzyme relating to the development of characteristic odor of onion. *J. Agric. Food Chem.* **19,** 166.

Kay, D. E. (1970). The production and marketing of pepper (a review). *Trop. Sci.* **12,** 201.

Kelkar, N. C., and Sanjeeva Rao, B. (1934). Essential oil from the rhizomes of *Curcuma longa* L. *J. Indian Inst. Sci., Sect. A* **17,** 7.

Kellogg, Brochmannhanseen, and Sevendsena (1964). Gas chromatography of esters of plant acids and their identification in plant materials. *J. Pharm. Sci.* **53,** 420.

Kepka, A. K., and Sypien, M. A. (1971). The influence of some factors on the keeping quality of onions. *Acta Hortic.* **20,** 65.

Kepkowa, A. (1966). The influence of time and rate of application on the effectiveness of maleic hydrazide in the storage of onions under different conditions. *Rocz. Nauk Rolnos Ser. A* **91,** 361.

Kerzner, 1. L., and Ershova, M. A. (1961). Determination of allyl oil in mustard seeds and their byproducts. *Maslob.-Zhir. Prom-st.* **27** (11), 41.

Khalique, A., and Das, N. R. (1968). Examination of *Curcuma longa.* Part II. *Sci. Res. (Dacca)* **5,** 44.

Kharchenko, N. (1971). "Eat More Onions," Mag. Sect., Suppl. Vol. IX, No. 11748 p. A. The Daily Hitavada, Nagpur, India.

Kholodova, V. P. (1964). Composition of the proteins of poppy seeds and their relation to physiological function. *Fiziol. Rast.* **11,** 1038.

Khristova, A. (1961). Determination of pigments in red pepper. *Izv. Tsentr. Nauch- noizsled. Inst. Rastenievud.* **10,** 103.

Kihara, Y. (1937). Carbohydrates of the bulb of Allium. VIII. Distribution of scorodose. *J. Agric. Chem. Soc. (Jpn.)* **13,** 363; cf. *Chem. Abstr.* **31,** 3013 (1937).

Kihara, Y., and Inoue, T. (1962). Antioxidant activity of spice powders in foods. *Nippon Shokuhin Kogyo Gakkai-Shi* **9,** 290.

Kilgore, L. B. (1932). The emulsifying qualities of mustard. *Glass Packer* p. 621.

Kim, M. S. (1943). Effect of certain condiments on gastric secretion. *Korean Med. J.* **3,** 115.

Kimura, Y., and Yamamoto, K. (1964). Cytological effect of chemicals on tumors. XXIII. Influence of crude extracts from garlic and some related spices on MTK Sarcoma. III. *Gann* **55,** 325; cf. *Chem. Abstr.* **63** 1089d (1965).

Kinner, J. A., Kotula, A. W., and Morcuri, A. J. (1968). Microbiological examination of ingredients of Eastern type Turkey rolls. *Poult. Sci.* **47,** 1442.

Kirchner, J. G. (1967). "Thin Layer Chromatography." Wiley (Interscience), New York.

Kirchner, J. G., and Miller, J. M. (1952). Preparation of terpeneless essential oils. *Ind. Eng. Chem.* **44,** 318.

Kirchner, J. G., Miller, J. M., and Keller, G. J. (1951). Separation and identification of some terpenes by a new chromatographic technique. *Anal. Chem.* **23,** 420.

Kirk, L. D., Black, L. T., and Mustakas, G. C. (1964). Mustard seed processing: essential oil composition. *J. Am. Oil Chem. Soc.* **41,** 599.

Kirtikar, K. R., and Basu, B. D. (1933). "Indian Medicinal Plants," Vol. 2, p. 1345. L. M. Basu, Allahabad.

Kisgyorgy, Z., Horvath, M., and Bucur, M. (1962). The Capsaicin, ascorbic acid and colouring substance content of various ground pepper assortments. *Ind. Alliment., Prod. Veg.* **13,** 179.

Kjar, A., and Jart, A. (1957). Iso-thiocyanates. XXIX. Separation of volatile isothiocyanates by gas chromatography. *Acta Chem. Scand.* **11,** 1423.

Klein, E., and Rojahn, W. (1967). (+) -Verbenone and newly discovered component of Spanish rosemary oil (*Rosmarinus officinalis* L). *Dragoco Rep. (Engl. Ed.)* **14,** 75.

Kleineri, J. (1963). Vanilla and vanillin. *Rev. Int. Choc.* **18**, 246 and 323.

Klimenko, V. G. (1959). Proteins of the seeds of some representatives of the Cruciferae. *Tr. Khim. Prir. Soedin.* **2**, 25.

Klohr-Meinhardt, R. (1958a). The action of light on volatile oil synthesis. *Planta Med.* **6**, 203; *Chem. Abstr.* **52**, 20904h (1958).

Klohr-Meinhardt, R. (1958b). The synthesis of volatile oil in metroplastic grafts in umbelliferous plants. *Planta Med.* **6**, 208; Chem. Abstr. **52**, 20905a (1958).

Kneilman, M. (1970). Changes in regulations regarding essence. Gordian **70**, 206.

Knickerbocker Mills (1942). "A Short Study of Spices, 1842–1942." Knickerbocker Mills, New York.

Kobayashi, M., Lshibashi, K., Katsubara, J., and Hashimoto, K. (1967). The constituents of staranise oils and fennel oils. *J. Chem. Soc. Jpn., Ind. Chem. Sect.* **70**, 1195.

Koeppin, B. H., and Vander Spuy, J. E. (1961). Microbial hydrolyses of quercitin glycoxides from the inner scales of onion. *S. Afr. J. Agric. Sci.* **4**, 557.

Kofler, L. (1936). Increase of essential oil in caraway and fennel during storage. *Sci. Pharm.* **7**, 106.

Kofler, L., and Herensschwand, G. V. (1935). Estimation of essential oils in drugs and the oil content of peppermint, sage, fennel and caraway. *Arch. Pharm. (Weinheim, Ger.)* **273**, 388.

Kohler, W. (1957). Chemical behaviour of organic plastics towards essential oils. *Werkst. Korros.* **8**, 487.

Kohman, E. F. (1947). The chemical components of onion vapors responsible for wound healing qualities. *Science* **106**, 625.

Kohman, E. F. (1952). Onion pungency and onion flavour—their chemical determination. *Food Technol.* **6**, 288.

Kojima, M., Akawori, Y., Ichikawa, I., and Mochizuki, M. (1966). Gas chromatographic studies on the acrid components of Japanese horseradish (Wasabi) powder. *Hakko Kogaku Zasshi* **44**, 177.

Kojiya, Mizaemon Co. (1966). Nutritive garlic foodstuffs. Japanese Patent 16,104.

Kokoski, C. J., Kokoski, R. J., and Slama, F. J. (1958). Fluorescence of powdered vegetable drugs under ultraviolet radiation. *J. Am. Pharm. Assoc.* **47**, 715.

Kolousek, J., and Goulson, C. H. (1955). Plant proteins: Amino acid content of the seed protein of fenugreek and the seed protein and thallus protein (*Galanga officinalis*). *J. Sci. Food Agric.* **6**, 203.

Komissarov, V. A., and Andreeva, A. V. (1965). Changes in the carbohydrates of garlic during storage under various conditions. *Dokl. TSKhA* **115**, Part 1, 139.

Konecsni, I. (1956–1957). Studies on the microscopic determination of added food paprika in milled spice—paprika. *Kulonlenyomat Ommi* **4**, 439.

Koparieva, N. P., and Potapova, L. M. (1966). Photometric determination of essential oil in onion and garlic tissues. *Prikl. Biokhim. Mikrobiol.* **2**, 298.

Kopec, K. (1967). The activity of Redox enzymes in stored fruits and vegetables. *Acta Univ. Agric., Brno, Fac. Agron.* **15**, 447.

Kosker, O., Fellers, C. R., and Esselen, W. B., Jr. (1949). Mustard as a preservative for fruit juices. *Glass Packer* **28**, 818.

Kosker, O., Esselen, W. B., Jr., and Fellers, C. R. (1951). Effect of allyl isothiocyanate and related substances on the thermal resistance of *A. niger. S. celipipoideus* and *B. thermoacidurans*. *Food Res.* **16**, 510.

Kosuge, S., and Furuta, P. (1970). Studies on the pungent principles of Capsicum. XIV. Chemical composition of the pungent principle. *Agric. Biol. Chem.* **34**, 248.

Kosuge, S., and Inagaki, Y. (1962). Pungent principles of red pepper. VI. Determination of contents of the two pungent principles. *Nippon Nogei Kagaku Kaishi* **36**, 251; cf. *Chem. Abstr.* **57**, 15512i (1962).

Kosuge, S., Inagaki, Y., and Okumura, H. (1965). *Nippon Nogei Kagaku Kaishi* **35**, 540.

Kozma-Kovacs, E., Kevei-Pichier, E., and Lendvai, I. (1977). Determination of the capsaicin content in raw green paprika. *Acta Aliment. Acad. Sci. Hung.* **6,** 1.

Kramer, A. (1959). Glossary of some terms used in sensory panel evaluation of foods and beverages. *Food. Technol.* **13,** 733.

Krinitz, B. (1971). I. R. detection of false positives for gums in mayonnaise due to mustard flour. *J. Assoc. Offic. Anal. Chem.* **54,** 743.

Krishnamurthy, K., and Sreenivasamurthy, V. (1956). Garlic. *Bull. Cent. Food Technol. Res. Inst., Mysore* **5,** 264.

Krishnamurthy, N., and Natarajan, C. P. (1973). Colour and its changes in chilies. *Indian Food Packer* **27**(1), 39.

Krishnamurthy, N., Nambudiri, E. S., Mathew, A. G., and Lewis, Y. S. (1970). Essential oil of ginger. *Indian Perfum.* **14,** Part 1.

Krishnamurthy, N., Padmabai, R., Natarajan, C. P., and Kuppuswamy, S. (1975). Colour content of tumeric varieties and studies on processing. *J. Food Sci. Technol.* **12,** 12.

Krishnamurthy, N., Mathew, A. G., Nambudiri, E. S., Shivashankar, S., Lewis, Y. S., and Natarajan, C. P. (1976). Turmeric oleoresin. *Trop. Sci.* **18,** 37.

Krishnamurthy, N., Lewis, Y. S., and Natarajan, C. P. (1977). Ginger from hilly regions of India. *J. Food Sci. Technol.* **14,** 128.

Krishnaswamy, M. A., Patel, J. D., and Parthasarthy, N. (1971). Enumeration of microorganisms in spices and spice mixtures. *J. Food Sci. Technol.* **8,** 191.

Krishnaswamy, M. A., Patel, J. D., Parthasarathy, N., and Nair, K. K. S. (1973). Some of the types of coliforms, aerobic mesophilic spore formers, yeasts and moulds present in spices. *J. Plant. Crops* **1,** Suppl., 200.

Krishnaswamy, M. A., Patel, J. D., Nair, K. K. S., and Muthu, M. (1974a). Microbiological quality control of spices. *Proc. Symp. Dev. Prospects Spices Ind. India, 1974* p. 46.

Krishnaswamy, M. A., Patel, J. D., Nair, K. K. S., and Muthu, M. (1974b). Microbiological quality of certain spices. *Indian Spices* **11,** 6.

Krishnaswamy, M. A., Nair, K. K. S., Patel, J. D., and Parthasarathy, N. (1974c). Preliminary observations on the survival of salmonelia in curry, sambar, coriander and red-chilli powders. *J. Food Sci. Technol.* **12,** 195.

Krummel, G. (1961). Comparison of methods for determining vanillin mixed with sucrose. *Dtsch. Lebensm.-Rundsch.* **57,** 276.

Kuehn, H. (1962). The enrichment of vegetables with trace nutrients by fertilizing. *Landwirtsch. Forsch., Sonderh.* **16,** 117.

Kuehn, L., Pfeifer, S., Slavik, J., and Appelt, J. (1964). *Papaver caucasicum* alkaloids. *Naturwiissenschaften* **51,** 556.

Kuehner, R. L., ed. (1964). Recent advances in odor-theory, measurements and control. *Ann. N.Y. Acad. Sci.* **116,** 357.

Kulka, K. (1962). Chemistry of essential oils. *Perfum. Essent. Oil Rec.* **53,** 147.

Kulka, K. (1967). Aspects of functional groups and flavour. *J. Agric. Food Chem.* **15,** 48.

Kulkarni, B. M., and Pruthi, J. S. (1967). A simple and rapid test for the detection of adulteration in cardamom seeds. *Indian Food Packer* **21,** 14.

Kulkarni, V. G. (1958). Potentialities of essential oil industry in North Kanara Dist. *Proc. Symp. Essent. Oils Aromat. Chem., 1958* p. 43.

Kumar, K. R., and Anandaswamy, B. (1974). Packaging of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 000.

Kum-Tatt, L. (1961). The characterisation and determination of nitrogenous organic bases in mixtures with ammonium reineckate. *Anal. Chim. Acta* **24,** 397 (in English).

Kunz, O. (1970). Process for production of an aromatized edible oil. Swiss Patent 490,809.

Kuon, J., and Berhnard, R. A. (1963). An examination of the free amino acids of the common onion (*Allium cepa*). *J. Food Sci.* **28,** 298.

Kupiecki, F. P., and Virtanen, A. I. (1960). Cleavage of alkyl cysteine sulfoxides by an enzyme in onion. *Acta Chem. Scand.* **14,** 1913.

Kuppuswamy, S. (1974). Drying of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 29.

Kurte, D. D. (1968). Pigments of red sweet pepper. *Ber. Dtsch. Bot. Gaz.* **81,** 153.

Kurup, P. G. (1959). Observation tour report on present status of pepper cultivation in south east Asian countries, *Mimeogr. Rep.*, pp. 1–39.

Labruyere, B. L. (1966). Determination of the pungent constituents of *Piper nigrum*. *J. Agric. Food Chem.* **14,** 469.

Labruyere, B. L., Olsthoorn-De, C., and Smeenge, F. (1968). Determination of traces of volatile solvents in spices oleoresins by gas chromatographic analysis of the head-space. *Perfum. Essent. Oil Rec.* **59,** 206.

Lacroix, L. J., Chua, G. K., Levy, R., and Unrau, A. M. (1967). The application of electron capture gas chromatography in the determination of flavour components of onions. *Proc. Am. Soc. Hortic. Sci.* **91,** 410.

La Face, F. (1962). Analytical and technological research progress in the field of essential oils. *Essenze Deriv. Agrum.* **32,** 155.

Lahache, J. (1919). Antiseptic action of spices in preserved foods. *Ind. Frac. Conserve* **5,** 211; cf. *Chem. Abstr.* **15,** 2933 (1921).

Lal, G., and Sadasivan, R. S. (1944). Standardization of curry powders. *Indian J. Agric. Sci.* **14,** 32.

Lal, R. N., and Sen, T. (1971). Studies on the essential oils from the seeds of Indian varieties of *Foeniculum vulgare* Miller. *Flavour Ind.* **2,** 544.

Lamparsky, D. (1963). Studies on volatile plant substances. CLXXXII. The alcohols present in the essential oil of bay. *Helv. Chim. Acta* **46,** 185.

Landrock, A. H., and Proctor, B. E. (1951a). Measuring humidity equilibria. *Mod. Packag.* **24** (6), 123.

Landrock, A. H., and Proctor, B. E. (1951b). A new graphical method for obtaining relative humidity equilibria data with special reference to its role in food packaging studies. *Food Technol.* **5,** 332.

Landrock, L. B. (1957). A new treatment of hygroscopic equilibria; application to walnuts (*J. regia*) and other foods. *Food Res.* **22,** 608.

Lang, O. W., Farber, L., Yerman, F., and Bech, C. (1954). The determination of spoilage of proteinaceous foodstuffs, particularly fish. *Ind. Eng. Chem., Anal. Ed.* **16,** 490.

Langenau, E. E. (1959). Oleoresins for the flavour chemist. *Am. Perfum. Aromat.* **74,**

Langenau, E. E. (1968). Correlation of objective and subjective methods as applied to the perfumery and cosmetic industries. *Am. Soc. Test. Mater.* p. 71.

Lantz, E. M. (1946). Effect of canning and drying on carotene and ascorbic acid content of chili. *N.M., Agric. Exp. Stn., Bull.* **327.**

Lapworth, A., Pearson, L. K., and Royle, F. A. (1917). Pungent principle of ginger (1). Chemical characters and decomposition products of Thresh's gingerol. *J. Chem. Soc.* **111,** 777; cf. *Chem. Abstr.* **11,** 3023 (1917).

Larcher, J. (1967). The preparation of black pepper. *Agron. Trop. (Paris)* **22,** 1094.

Larry, D. (1973). GLC determination of beta-asarone component of oil of calamus in flavours and beverages. *J. Assoc. Off. Anal. Chem.* **56,** 121.

Lassanyi, S. (1965). Approximate quantitative determination of linalool in coriander oil by thin-layer chromatography. *Acta Agron. Acad. Sci. Hung.* **14,** 135; cf. *Chem. Abstr.* **64,** 9503e, (1966).

Lauber, F. U., and Hollander, F. (1950). Toxicity of the mucigogue, eugenol, administered by stomach tube to dogs. *Gastroentrology* **15,** 481; cf. *Chem. Abstr.* **15,** 9051c (1950).

Laul, M. S., Bhalerao, S. D., Rane, V. R., and Amla, B. L. (1970). Studies on the sun-drying of chilies. *Capsicum annum* Linn.) *Indian Food Packer* **24**(2), 22.

Lavington, A. L., Knowles, R. E., Edwards, R. H., and Kohler, G. O. (1974). Processing of

pimiento waste to provide a pigment source for poultry feed. *J. Sci. Food Agric.* **25,** 483.

Lawrence, B. M. (1967). A review of some of the commercial aspects of cinnamon. *Perfum. Essent. Oil Rec.* **58,** 236.

Lawrence, B. M. (1968). Thin layer chromatography. Part 1. A review of the use of thin-layer chromatography in essential oil analysis. *Perfum. Essent. Oil Rec.* **59,** 421.

Lawrence, B. M. (1969). Determination of the botanical origin of cinnamons of commerce by thin layer chromatography. *Can. Inst. Food Technol. J.* **2,** 178.

Lawrence, B. M., Hogg, J. W., and Terhune, S. J. (1969a). Essential oils and their constituents. Part II. The oil of *Alpinia officinarum,* Hance. *Perfum. Essent. Oil. Rec.* **60,** 88.

Lawrence, B. M., Hogg, J. W., and Terhune, S. J. (1969b). Artefacts found during alumina chromatography. *J. Chromatogr.* **2,** 261.

Lawrence, B. M., Hogg, J. W., and Terhune, S. J. (1971a). The essential oils and their constituents. IV. Some new constituents in trace amounts found in *Salvia officinalis* L. *Parfums, Cosmet., Savons Fr.* **1,** 256.

Lawrence, B. M., Terhune, S. J., and Hogg, J. W. (1971b). Essential oils and their constituents. VI. The so-called exotic oil of *Ocimum basilicum* L. *Flavour Ind.* **2,** 173.

Lawrence, B. M., Hogg, J. W., Terhune, S. J., and Pichitakul, N. (1972a). Essential oils and their constituents. IX. The oils of *Ocimum sanctum* and *Ocimum basilicum* from Thailand. *Flavour Ind.* **3,** 47.

Lawrence, B. M., Hogg, J. W., and Terhune, S. J. (1972b). Essential oils and their constituents X. Some new trace constituents in the oil of *Mentha piperita* L. *Flavour Ind.* **3,** 467.

Leach, A. E., and Winton, A. L. (1941). "Food Inspection and Analysis." Wiley, New York.

Lease, J. G., and Lease, E. J. (1956a). Factors effecting the retention of red colour in peppers. *Food Technol.* **10,** 368.

Lease, J. G., and Lease, E. J. (1956b). Effect of fat-soluble antioxidants on the stability of the red colour of pepper. *Food Technol.* **10,** 403.

Lease, J. G., and Lease, E. J. (1962). Effects of drying conditions on initial colour, colour retention and pungency of red peppers. *Food Technol.* **16,** 104.

Leclerc, H. (1948). Fenugreek—*Lotus corniculatus* as an anti-spasmodic. *Rev. Phytother.* **12,** 331 and 347.

Lee, G. D., Kaufman, F. 1., Harlan, J. W., and Niezabitowski, W. (1962). Application of gas chromatography to a study of nutmeg oil flavour. *Gas Chromatogr., Int. Symp. (U. S.)* 1961. **3,** 475.

Lee, L. A. (1956). Determination of free and combined formaldehyde using modified chromotropic acid procedure. Application to determination of piperine content of pepper. *Anal. Chem.* **28,** 1621.

Lee, L. A., and Ogg, C. L. (1956). Improved apparatus and method for the determination of the volatile oil content of spices. *J. Assoc. Off. Agric. Chem.* **39,** 806.

Lee, T. S. *et al.* (1970). Microbiological studies of the fermentation of red pepper paste. 1. Distribution of yeast during red pepper paste fermentation periods. *Korean J. Microbial.* **8,** 151.

Lehmann, 1., and Watts, B. M. (1951). Antioxidants in aequeous fat systems. *J. Am. Oil Chem. Soc.* **28,** 475.

Lendner, A. (1919). Detection of adulteration in black pepper. Schiweiz. *Apoth.-Ztg.* **57,** 591.

Lerke, P. A., and Farber, L. (1966). Effect of beam irradiation on the microbial content of spices and teas. *Food Technol.* **14,** 266.

Leterrier, F., and Viossat, B. (1967). Productions of some poppy alkaloid free radicals by a chemical means. *C. R. Hebd. Seances Acad. Sci., Ser. C* **265,** 410.

Levins, R. J., and Ottenstein, D. M. (1967). The effect of the tubing material in gas chromatography of polyols and vanillins. *J. Chromatogr.* **5,** 539.

Lewis, E. J., and Watts, B. M. (1958). Antioxidants and copper binding properties of onions. *Food Res.* **23,** 274.

Lewis, N. F. (1974). Irradiation of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 32.

Lewis, N. F., Rao, B. Y. K., Shah, A. R., Tewari, G. M., and Bandyopadhyay, C. (1977). Antibacterial activity of volatile components of onion. *J. Food Sci. Technol.* **14,** 35.

Lewis, Y. S. (1972). The importance of selecting the proper variety of a spice for oil and oleoresin extraction. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 183.

Lewis, Y. S., and Neelakantan, S. (1960). Nature of tartaric acid in tamarind. *Food Sci.* **9,** 405.

Lewis, Y. S., and Neelakantan, S. (1964). The chemistry, biochemistry and technology of tamarind. *J. Sci. Ind. Res.* **23,** 204.

Lewis, Y. S., Neelakantan, S., and Bhatia, D. S. (1961). Determination of free and combined tartaric acids in plant products. *Food Sci.* **10,** 49.

Lewis, Y. S., Nambudiri, E. S., and Philip, T. (1966). Composition of cardamom oils. *Perfum. Essent. Oil Rec.* **57,** 623.

Lewis, Y. S., Nambudiri, E. S., and Natarajan, C. P. (1967). Studies on some essential oils. *Indian Perfum.* **11,** Part I, 5.

Lewis, Y. S., Neelakantan, S., Philip, T., and Nambudiri, E. S. (1968). Production of white pepper in India. *Spices Bull.* **5**(6), 6

Lewis, Y. S., Nambudiri, E. S., Krishnamurthy, N., and Natarajan, C. P. (1969a). White pepper. *Perfum. Essent. Oil. Rec.* **60,** 53.

Lewis, Y. S., Nambudiri, E. S., and Krishnamurthy, N. (1969b). Composition of pepper oil. *Perfum. Essent. Oil Rec.* **60,** 259.

Lewis, Y. S., Nambudiri, E. S., and Krishnamurthy, N. (1969c). Essential oil of pepper. *Indian Perfum.* **13,** Part I, 22.

Lewis, Y. S., Menon, P. G. K., Natarajan, C. P., and Amla, B. L. (1970). Tamarind concentrate. *Indian Food Packer* **24**(1), 18.

Lewis, Y. S., Mathew, A. G., Nambudiri, E. S., and Krishnamurthy, N. (1972). Oleoresin ginger. *Flavour Ind.* **3,** 78.

Lewis, Y. S., Krishnamurthy, N., and Nambudiri, E. S. (1974). Spice oils and oleoresins. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 60.

Lewis, Y. S., Krishnamurthy, N., Shivashankar, S., and Natarajan, C. P. (1977). Quality studies on cinnamons marketed in India. *ISI Bull.* **29,** 377.

Lewis, Y. S., Krishnamurthy, N., Shivashankar, S., and Natarajan, C. P. (1979). Report on an unusual sample of coriander oil. *J. Indian Spices* **16**(1), 7.

Li, K. H., Bundus, R. H., and Noznick, P. P. (1967). Prevention of pink color in white onions. U.S. Patent 3,352,691 (C1.99-154).

Lindgren, D. L., and Vincent, L. E. (1966). Relative toxicity of hydrogen phosphide to various stored products insects. *J. Stored Prod. Res.* **2,** 141.

Lindgren, D. L., Vincent, L. E., and Krohne, H. E. (1954). Relative effectiveness of ten fumigants to adults of 8 species of stored products insects. *J. Econ. Entomol.* **47,** 923.

Litchfield, J. T., and Wilcoxon, F. (1949). *Acromus calamus* Linn. Antibacterial spectrum. *J. Pharmacol. Exp. Ther.* **91,** 99.

Little, Inc. A.D. (1958). "Flavour Research and Food Acceptance." Van Nostrand Reinhold, Princeton, New Jersey.

Liverseage, J.F. (1932). "Adulteration and Analysis of Foods and Drugs." Churchill, London.

Lopez, R. C., Olmedo, R. G., and Peralta, T. (1950). A study of the composition of the seed of fenugreek. *An. Bromatol.* **2,** 353.

Lossner, G. (1967). The constituents of *Marjorana hortensis* Moench. Part 2: *cis*- and *trans*-sabinene hydrate, constituents of the essential oil. *Pharmazie* **22,** 51.

Lu, A. T., and Whitaker, J. R. (1974). Some factors affecting rates of heat inactivation and reactivation of horseradish peroxidase. *J. Food Sci.* **39,** 1173.

Lukes, T. M. (1959). Pinking of onions during dehydration. *Food Technol.* **13,** 391.

Lundberg, W. O., Halvorson, H. O., and Burr, G. O. (1944). The antioxidant properties of nordihydrognaiaretic acid (N.D.G.A.). *Oil Soap (Chicago)* **21,** 33.

McCormack, T. H. (1954). Extraneous materials in spices. *J. Assoc. Off. Agric. Chem.* **37,** 157.

Mackay, D. A. M., and Hewitt, E. I. (1959). Application of flavour enzymes to processed foods. II. Comparison of the effect of flavour enzymes from mustard and cabbage upon dehydrated cabbage. *Food Res.* **24,** 253.

Mackay, D. A. M., Lang, D. H., and Berdick, M. (1961). Objective measurement of odour. Ionization detection of food volatiles. *Anal. Chem.* **33,** 1369.

McNair, J. B. (1930). Spices and condiments. *Field Mus. Nat. Hist. Publ., Bot. Ser.* **15,** 65.

Madan, C. L., Kapur, B. M., and Gupta, U. S. (1966). Saffron. *Econ. Bot.* **20,** 377.

Madhusudan, L. R. (1974). Curry powder. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 59.

Mafredini, T. A. A., Mizrahi, I., and Montes, A. (1962). Ultraviolet spectrophotometry of mint essential oils. *An Asoc. Quim. Argent* **50,** 297.

Maga, J. A. (1975). "Capsicum." Chem. Rubber Publ. Co., Cleveland, Ohio.

Mahadeviah, B., Chang, K. S., and Balasubrahmanyam, N. (1976). Packaging and storage studies on dried ground and whole chilies (*Capsicum annuum*) in flexible consumer packages. *Indian Food Packer* **33**(6), 33.

Mahendran, C. (1972). The marketing of sarawak pepper. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 223.

Mahfouz, M., and El Dakhakhny, M. (1960). The isolation of crystalline active principle from *Nigella sativa* seeds. *J. Pharm. Sci. U.A.R.* **1,** 9.

Majumdar, M. (1965). Role of spices in the export trade of India. *Spices Bull. (Semin. No.)* **4**(10), 41.

Majumder, S. K. (1962). Using ethylene dibromide with methyl bromide for grain disinfestation. *Pest Technol.* **4,** 134.

Majumder, S. K., and Muthu, M. (1964). Durofume process for pest control in stored foods. *A.S.C. J.* **17,** 22.

Majumder, S. K., and Muthu, M. (1965). An improved prophylactic process for insect control in stored foods. *Proc. Int. Congr. Food Sci. Technol., 1st, 1962* p. 507.

Majumder, S. K., Krishnamurthy, T. S., and Muthu, M. (1963). An infra-red heat disinfestation unit for foodstuffs. *Res. Ind.* **8,** 229.

Maleeny, R. T. (1961). A method of analysis of non-volatile encapsulated substances. *Food Technol.* **15,** 448.

Malla, S., Nigam, M. C., and Handa, K. L. (1962). Spectrophotometric determination of menthone and menthol in essential oil of *Mentha arvensis. Riechst., Aromen* **12,** 221.

Malter, B. T. (1952). Decolourizing and sterilizing natural spices. U.S. Patent 2,615,813.

Malysheva, A. (1965). Determination of essential oil in mustard seeds. *Essent. Oil Crops USSR* **167,** 349.

Manly, R. S., Hargreaves, G., and Pillard, R. (1959). *Arch. Oral Biol.* **1,** 145. (cited by Maruzzella, 1962).

Manning, C. E. F. (1969). The market for vanilla beans. *Trop. Prod. Inst.* [*Rep.*] *G. (G.B.)* **G42,** 1–21.

Manning, C. E. F. (1970). The market for cinnamon and cassia and their essential oils. *Trop. Prod. Inst.* [*Rep.*] *G. (G.B.)* **G44,** 1–74.

Mano Daiji (1962). The inhibitory action of some plant extracts on bacterial growth. II. Changes in the susceptibility to antibiotics of the strains of bacteria adapted by culturing with a fraction from *Allium sativum. Nippon Saikingaku Zasshi* **17,** 417.

Manolov, K. R., and Georgiev, E. V. (1967). Semi-micro method for the determination of linalool in coriander oil. *Talanta* **14,** 124.

Mantis, A. J., Karaioannoglou, Pr. G., Spanos, G. P., and Panetsos, A. G. (1978). The effect of

garlic extract on food poisoning bacteria in culture media. I. *Staphylococcus aureus*. *Lebensm.-Wiss. + Technol.* **11,** 26.

Marivala, K. V. (1974). Processing of pepper. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 17.

Marker, R. E., Wagner, R. F., Ulshafer, P. R., Wittbecker, E. L., and Goldsmith, D. P. (1947). New sources for sapogenins. *J. Am. Chem. Soc.* **69,** 2242.

Marketing Research Corporation of India (1968). "Survey of India's Export Potential of Spices," Vols. IIA and III. Market. Res. Corp. India, New Delhi.

Marketing Research Officer (Maharashtra State) (1968). Marketing, grading and quality control for export of chillies. *Indian Spices* **6**(3), 41.

Markman, A. L., and Fazylova, M. F. (1961). The polarographic behavior of vanillin. *Uzb. Khim. Zh.* **4,** 64.

Markov, S. T. (1964). A micromethod for the quantitative determination of sulphur in glucosides of garlic. *Farmatsiya (Sofia)* **14**(3), 36.

Markwell, W. A. N., and Walker, L. J. (1928). Commercial evaluation of cloves. *Perfum. Essent. Oil Rec.* **19,** 496.

Marongiu-Fortucci, P. (1972). Recent trends in the pepper economy. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 219.

Marta, S., and Halley, O. M. (1932). Note on myrosin. *J. Biol. Chem.* **96,** 443.

Martin, G. E., Dyer, R., Jansen, J., and Sahn, M. (1975). Determining the authenticity of vanilla extracts. *Food Technol.* **29,** 54.

Martinez, A. P., Lee, W. W., and Goodman, L. (1968). Fluoroethy lamines. III. Some alkylating agents derived from uracil. *J. Med. Chem.* **11,** 384.

Maruzzella, J. C. (1960). The antifungal properties of essential oil vapours. *Soap, Perfume & Cosmet.* **33,** 835.

Maruzzella, J. C. (1962). The germicidal properties of perfume oils and perfumery chemicals. *Am. Perfum.* **77,** 67.

Maruzzella, J. C., and Brenner, D. J. (1963). Anti-tumour effect of essential oils on *Crocker sarcoma* 180 in mice. *Arzneim.-Forsch.* **13,** 375.

Maruzzella, J. C., and Freundlich, M. (1959). Antimicrobial substances from seeds. *J. Am. Pharm. Assoc.* **48,** 356.

Maruzzella, J. C., and Henry, P. A. (1958). The antimicrobial activity of perfume oils. *J. Am. Pharm. Assoc., Sci. Ed.* **47,** 471.

Maruzzella, J. C., and Sicurella, N. A. (1960). Antibacterial activity of essential oil vapours. *J. Am. Pharm. Assoc., Sci. Ed.* **49,** 692.

Maruzzella, J. C., Balter, J., and Katz, A. (1959a). The action of perfume-oil vapours on fungi. *Am. Perfum. Aromat.* **74,** 21.

Maruzzella, J. C., Balter, J., and Katz, A. (1959b). Further studies on the action of perfume-oil vapours on micro-organisms. *Perfum. Essent. Oil Rec.* **50,** 955.

Maruzzella, J. C., Scrandis, D., Scrandis, J. B., and Grabon, G. (1960). Action of odoriferous organic chemicals and essential oils on wood destroying fungi. *Plant Dis. Rep.* **44,** 789.

Maruzzella, J. C., Reine, S., Solat, H., and Zeitlin, A. (1963). The action of essential oils on phytopathogenic bacteria. *Plant Dis. Rep.* **47,** 23.

Mathai, C. K. (1976). Variability in turmeric (Curcuma species) germ-plasm for essential oil and curcumin. *Qual. Plant.—Foods Plant Hum. Nutr.* **25,** 227.

Mathew, A. G., Lewis, Y. S., Jagadishan, R., Nambudiri, E. S., and Krishnamurthy, N. (1971a). Oleoresin capsicum. *Flavour Ind.* **2,** 23.

Mathew, A. G., Nambudiri, E. S., Anathakrishna, S. M., Krishnamurthy, N., and Lewis, Y. S. (1971b). An improved method for estimation of capsaicin in capsicum oleoresins. *Pab. Pract.* **20,** 856.

Mathew, A. G., Krishnamurthy, N., Nambudiri, E. S., and Lewis, Y. S. (1973). Oil of ginger. *Flavour Ind.* **4,** 226.

Mathur, P. B., Singh, K. K., and Kapoor, N. S. (1952). Review of researches on the cold storage of fruits and vegetables at the CFTRI, Mysore 1950–1952. *J. Indian Soc. Refrig. Eng.* **3,** 47.

Mathur, P. B., Date, W. B., Srivastava, H. C., and Subramanyam, H. (1958). Effects of pre-harvest foliage sprays of maleic hydrazide on the cold storage behaviour of onions. *J. Sci. Food Agric.* **9,** 312.

Mathur, V. K., Hemamalini, S., and Bhatia, B. S. (1971). Quality standards and specifications of spice mixture developed for use in dehydrated convenience foods. *Indian Food Packer* **25**(2), 24.

Mathur, V. K., Gopinathan, V. K., Bhagirathi, B., and Bhatia, B. S. (1974). Development of spice mixtures for use in dehydrated mutton curries. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 31.

Matikkala, E. J., and Virtanen, A. I. (1967). The quantitative determination of amino acids and gamma glutamyl peptides of onion. *Acta Chem. Scand.* **21,** 2891.

Mattil, K. F., and Black, H. C. (1947). A laboratory method for determining the ability of antioxidants to stabilise fats in back goods. *J. Am. Oil Chem. Soc.* **24,** 325.

Maturova, M., Moza, B. K., Sitar, J., and Santavy, F. (1962). Isolation of alkaloids from several species of section scapiflora. *Planta Med.* **10,** 345; cf. *Chem. Abstr.* **58,** 5988c (1963).

Maveety, D. J. (1938). Inhibiting rancidity in oils. U.S. Patent 2,124,706.

May, P. (1926). Determination of moisture in cloves. *Perfum. Essent. Oil Rec.* **17,** 46.

Maymone, B., and Bergonzini, E. (1964). Essential amino acid content and biological value of proteins of some forage plants of Southern Italy. *Aliment. Anim.* **8,** 9.

Mayr, G. E., and Suhr, H. (1972). Preservation and sterilization of pure and mixed spices. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London 1972* p. 201.

Mazur, S. (1964). Determination of essential oils in vanilla extracts. *Przem. Ferment.* **7**(4), 83; cf. *Chem. Abstr.* **61,** 8126c (1964).

Meerov, Y. A. S. *et al.* (1972). Method of obtaining essential oil of coriander. USSR Patent, 340,684.

Mendez, J. (1968). Separation of simple phenols by using gas-liquid chromatography. *J. Gas. Chromatogr.* **6,** 168.

Meranger, J. C., Bartlet, J. C., Radecka, C., and Nigam, J. C. (1967). Essential oils and their constituents. XXXVI. X-Ray powder diffraction data for semicarbazones of aldehydes and ketones. *Perfum. Essent. Oil Rec.* **58,** 710; cf. *Chem. Abstr.* **67,** 111323k (1967).

Messerschmidt, W. (1964). Gas and thin-layer chromatographic investigation of the volatile essential oils of several thymus species. I. Influence of various factors on the formation and variation of essential oil. *Planta Med.* **12,** 501; *Chem. Abstr.* **62,** 10289c (1965).

Messerschmidt, W. (1965). Gas and thin-layer chromatography of the essential oils of several thymus spices. II. The influence of various origins on the composition of essential oils of thyme herb and *Thymus serphyllum* herb and proposals for their chromatographic evaluation. *Planta Med.* **13,** 56; cf. *Chem. Abstr.* **62,** 10289d (1965).

Messiaen, C. M., and Pereau-Leroy, P. (1969). Improving the storage of garlic bulbs, by gamma irradiation. *C.R. Seances Acad. Agric. Fr.* **5,** 485.

Metais, M. C. (Mrs.), Hasselmann, M., and Ney, M. (1967). Characterization of vanilla aroma in foods. *Ann. Falsif. Expert. Chim.* **60,** 220.

Metlitskii, L. V., Sal'kova, E. G., Mukhine, N., Korableva, N. P., and Morozova, N. P. (1964). Use of ionizing irradiations during storage of the onion and garlic. *Biokhim. Plodov Ovoshchei* p. 163.

Michaluk, A., and Oswiecimska, M. (1961). Obtaining quercitin from onion skins. *Farm. Pol.* **17,** 258.

Miller, G. (1951). New nitrogen technique assures fine grinding in only one pass. *Food Eng.* **23,** 36.

Miller, J. M., and Kirchner, J. G. (1952). Some improvements in chromatographic techniques for terpenes. *Anal. Chem.* **24,** 1480.

Miller, J. M., and Kirchner, J. G. (1953). Chromatostrips for identifying constituents of essential oils. *Anal. Chem.* **25,** 1107.

Miller, M. T. (1967). Rapid method for separation of extraneous materials from whole and degerminated corn meal, prepared mustard, and soy flour. *J. Assoc. Off. Anal. Chem.* **50,** 505.

Miller, P. A. (1965). Thyme and its essential oil. *Seifen, Oele, Fette, Washse* **91,** 676 (in German).

Ministry of Agriculture (1966). "Onion," Adv. Leafl. 358, p. 7. Minist. Agric., London.

Misra, B. D. (1962). Studies on physico-chemical, microbiological and technological aspects of curry powder and spice mixtures. Assoc. Thesis, C.F.T.R.I. Mysore, India.

Misra, B. D., and Pruthi, J. S. (1962). Chemical composition and packaging requirements of curry powders and spice mixtures. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract p. 15.

Misra, B. D., and Pruthi, J. S. (1963a). Storage studies on curry powders: Effect of type of container, vacuumization, gas packaging and storage temperature on the quality and shelf-life of curry powder and other spice mixtures. *Spices Bull.* **2**(6), 15.

Misra, B. D., and Pruthi, J. S. (1963b). Studies on the physico-chemical evaluation of flavour and colour of curry powders and other spice mixtures. *Spices Bull.* **3,** 5 and 23.

Misra, B. D., Pruthi, J. S., and Siddappa, G. S. (1962). Chemical composition and packaging requirements of curry powders and spice mixtures. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract p. 15.

Misra, K. P., and Nigam, S. S. (1969). Chemical investigation of the oil from seeds of *Anethum sowa* DC. *Riechst., Aromen, Korperpflegem.* **19,** 189.

Misra, R. C., and Krishnarao, G. S. (1961). Oxidimetric method of evaluation of volatile oils of spices. *J. Sci. Ind. Res., Sect. D* **20,** 331.

Misra, S. C., and Chandra, G. (1973). Chemistry and manufacture of vanillin. *Indian Perfum.* **17**(1), 46.

Mitchell, J. F. (1972). The carifta marketing protocol, its creation and maintenance. *Trop. Sci.* **14,** 11.

Mitra, S. N., and Roy, S. C. (1955). Standards for black pepper. *J. Proc. Inst. Chem., Calcutta* **27,** 100.

Mitra, S. N., and Roy, S. C. (1957a). Spices, analysis of spices and detection of adulterants. *J. Inst. Chem., Calcutta* **29,** 85.

Mitra, S. N., and Roy, S. C. (1957b). Detection of small amounts of turmeric in other spices. *J. Inst. Chem., Calcutta* **29,** 155.

Mitra, S. N., Roy, S. C., and Chatterjee, R. K. (1956). Detection of coaltar dye in turmeric (*Haldi*). *J. Indian Chem. Soc., Ind. News Ed.* **19,** 155.

Mitra, S. N., Sengupta, P. N., and Roy, B. R. (1961). The detection of oil soluble coaltar dyes in chili (capsicum). *J. Proc. Inst. Chem., Calcutta* **33,** 69.

Mitra, S. N., Roy, B. R., Mathew, T. V., and Roy, A. K. (1966a). Detection and estimation of colophony resin in asafoetida. *J. Proc. Inst. Chem., Calcutta* **38,** 121.

Mitra, S. N., Roy, B. R., and Roy, A. K. (1966b). Note on the importance of starch in the analysis of black pepper. *J. Proc. Inst. Chem., Calcutta* **38,** 215.

Mitra, S. N., Sengupta, P. N., and Sen, A. R. (1970). A comparative study of starch estimation in chilies (*Mirch*). *J. Inst. Chem., Calcutta* **42,** Pt. 1, 15.

Miyoshi, H. (1969). Studies on the flavour retention of processed mustard products. *J. Food Sci. Technol.* **16,** 441.

Mohan, V. (1962). Essential oils—Raman effect. *Indian Oil Soap J.* **28,** 100.

Molnar, E. J. (1942). Cloves, oil of cloves and eugenol—their medico-dental history. *Dent. Items Interest* **64,** 521.

Moncrieff, R. N. (1951). "The Chemical Senses," 2nd ed. Leonard Hill, London.

Monro, H. A. U. (1969). Manual of fumigation for insect control *FAO Agric. Stud.* **79.**

Moorhousc, A. (1962). Distillation of cloves, etc. in Zanzibar. *Soap, Perfum. & Cosmet.* **35,** 701.

Moreau, L., and Vinet, E. (1924). Practican antiseptic value of allyl mustard oil in wine making. *Ann. Falsif. Fraudes* **17,** 477.

Morgan, E. J. (1946). Pyruvic acid in the juice of onion (*Allium cepa*). *Nature (London)* **157,** 512.

Morozov, V. K. (1959). Fat and allyl oil contents of yellow mustard. *Maslob.-Zhir. Prom-st.* **25** (7), 15.

Mossel, D. A. A., and Van Kwik, H. J. L. (1955). A new and simple technique for the direct determination of the E. R. H. (Equilibrium Relative Humidity) of foods. *Food Res.* **20,** 415.

Mossel, D. A. A., Mossel-Hens, H. A. M., and Reith, J. F. (1951). Entrainment distillation as a reference method for the determination of water content of foods. Application of the method developed to some model substrates. *Recl. Trav. Chim. Pays-Bas* **70,** 331.

Moster, J. B., and Prater, A. N. (1952). Colour of capsicum spices. Measurements of extractable colour. *Food Technol.* **6,** 459.

Moster, J. B., and Prater, A. N. (1957a). Capsicum spices. 11. Extraction of colour. *Food Technol.* **11,** 146.

Moster, J. B., and Prater, A. N. (1957b). Colour of capsicum spices. A linear colour scale. *Food Technol.* **11,** 222.

Moster, J. B., and Prater, A. N. (1957c). Colour of capsicum spices. 1V. Oleoresin paprika. *Food Technol.* **11,** 226.

Moudgil, K. L. (1958). Standard methods of test for essential oils. *Proc. Symp. Essent. Oils Aromat. Chem, 1958* p. 18.

Moursi, M. A., and Tewfic, H. A. (1962). Nitrogen metabolism in onion bulbs during storage. *Ann. Agric. Sci.* **7,** 197.

Mouton, M. (1939). The determination of benzaldehyde and cinnamaldehyde as 2, 4, dinitro phenyl hydrazone and its possible applications to an assay of gelenie preparations of cherry laurel and cinnamon. *Bull. Sci. Pharmacol.* **46,** 148.

Moye, H. A. (1968). A comparison of phosphorimetric procedure for parathion on celery with the Averell-Norris and electron capture methods. *J. Assoc. Off. Anal. Chem.* **51,** 1260.

Mukerji, B. (1954). "Indian Pharmaceutical Codex." Counc. Sci. Ind. Res., New Delhi, India.

Mukerji, B., Zaidi, S. H., and Singh, G. B. (1961). Spices and gastric function. Part 1. Effect of *Curcuma longa* on the gastric secretion in rabbits. *J. Sci. Ind. Res., Sect. C* **20,** 25.

Mukherji, T. D., and Govind, R. (1959). A plant insecticide from *Acorus calamus*. *Indian J. Entomol.* **21,** 194.

Muller, C. J., and Jennings, W. C. (1967). Constituents of black pepper. Some sesquiterpene hydrocarbons. *J. Agric. Food Chem.* **15,** 762.

Muller, C. J., Creveling, R. K., and Jennings, W. G. (1968). Some minor sesquiterpene hydrocarbons of black pepper. *J. Agric. Food Chem.* **16,** 113.

Muller, P. A. (1963). Rosemary oil. *Seifen, Oele, Fette, Wachse* **89,** 375.

Muller, P. A., Ill (1971). Rosemary and rosemary oil. *Flavour Ind.* **2,** 633.

Munchow, P., and Pohleudekfabine, R. (1964). On thymol content of *Thymus vulgaris* L. Part 15 of the micro-analysis of essential oils. *Pharmzie* **19,** 655.

Munjal, K. K., Rai, J., and Verma, M. R. (1971). Evaluation of purity of saffron. *Res. Ind.* **16,** 294.

Murakami, T., Inugai, F., Nagasawa, M., Inatomi, H., and Mori, N. (1965). Water soluble constituents of crude drugs. 111. Free amino acids isolated from ginger rhizome. *Yakugaku Zasshi* **85,** 845.

Murav'ev, 1. A. (1946). Substituting coriander tincture for mint tincture in stomach drops. *Farmatsiya* **9,** 32.

Murphy, R. T., Gaston, L. K., and Guenther, F. A. (1965). Colorimetric analytical method for

bidrin residues in alfalfa, celery, lemon, peel, lettuce, orange peel, potatoes, string beans and tomatoes. *J. Agric. Food Chem.* **13,** 242.

Murthy, N. S. R., Prasad Rao, V. L. V., and Jayanandam, V. D. S. (1968). A note on preliminary studies on artificial drying of chili. (*C. annum*). *Andhra Agric. J.* **15,** 121.

Mustakas, G. C., Kirk, L. D., Sohns, V. E., and Griffin, E. L., Jr. (1965). Mustard seed processing: Improved methods for isolating the pungent factor and controlling protein quality. *J. Am. Oil Chem. Soc.* **42,** 33.

Muthu, M., and Majumder, S. K. (1962). A fumigation technique for the disinfestation of packaged spice products. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract p. 18.

Muthu, M., and Majumder, S. K. (1974). Insect control of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 35.

Muthu, M., Srinivasan, K. S., and Majumder, S. K. (1961). Serial fumigation of processed and packaged foods with air washed residual methyl bromide from fumigation chambers. *Food Technol.* **15,** 295.

Muthu, M., Misra, B. D., Majumder, S. K., and Pruthi, J. S. (1962). Disinfection of processed and spiced food materials by heat treatment. *Proc. Symp. Spices—Pole Natl. Econ., 1st, 1962* Abstract p. 16.

Muthu, M., Majumder, S. K., and Parpia, H. A. B. (1973a). A detector for phosphine at permissible level in air. *J. Agric. Food Chem.* **21,** 184.

Muthu, M., Narasimhan, K. S., and Majumder, S. K. (1973b). Sorption of phosphine used as a fumigant on foodstuffs. *Proc. Symp. Prog. Probl. Pestic. Residue Anal., 1971* p. 301.

Muthu, M., Krishnamurthy, T. S., and Majumder, S. K. (1974). An evaluation of impregnated respirator consister carbons for phosphine removal from air. *Pestic. Sci.* **5,** 245.

Myers (1926). *J. Pharm. Proc.* **27,** 248 (ref. Corran and Edgar, 1933).

Nabney, J., and Robinson, F. V. (1972). Constituents of pimento berry oil (*Pimenta dioica*). *Flavour Ind.* **3,** 50.

Nadkarni, A. K. (1954). "Indian Materia Medica," 3rd ed., p. 988. Popular Book Depot, Bombay.

Nagasawa, T., and Soga, O. (1961). Chemical studies on mentha. III. Odorous substances in mint plants. *Shimane Daigaku Ronshu, Shizen Kagaku* **10,** 32.

Nair, P. C. S., and Mathew, L. (1969). Vanilla. *Indian Spices* **6**(4), 2.

Nair, P. M., Thomas, P., Ussuf, K. K., Surendranathan, K. K., Limaye, S. P., Srirangarajan, A. N., and Padwal Desai, S. R. (1972). Studies on sprout inhibition of onions and potatoes and on delayed ripening of bananas and mangoes by gamma irradiation. *FAO/IAEA Int. Symp. Radiat. Preserv. Food, 1972.*

Nambudiri, E. S., Lewis, Y. S., Rajagopalan, P., and Natarajan, C. P. (1968). Production of cardamom oil by distillation. *Res. Ind.* **13,** 140.

Nambudiri, E. S., Lewis, Y. S., Krishnamurthy, N., and Mathew, A. G. (1970). Oleoresin pepper. *Flavour Ind.* **1,** 97.

Nambudiri, E. S., Mathew, A. G., Krishnamurthy, N., and Lewis, Y. S. (1975). Qualitative estimation of pungent principle in ginger. *Int. Flavour Food Additives* **6,** 135.

Nandpuri, K. S., Sooch, B. S., and Randhawa, K. S. (1969). Effect of gamma irradiation on the storage life and quality of onion bulb under ordinary storage conditions. *J. Res. Ludhiana* **6,** 755.

Nanz, R. A., and Cartwright, L. C. (1951). Comparative evaluation of spices. Flavour retention of natural spices and of spice oils and spice extratives after boiling and baking. *Food Technol.* **5,** 246.

Nanz, R. A., Tyler, G. A., and Cartwright, L. C. (1950). Comparative evaluation of spices. Flavour evaluation of natural spices and spice substitutes in representative test foods. *Food Technol.* **4,** 252.

Narasimhaiah, D. S. (1965). Improved method of conditioning and packing of chilies for export. *Spices Bull. (Semin. No.)* **4** (10), 75.

Narasimhaiah, D. S., and Rangaswamy, N. V. (1963). Quality control in black pepper, cardamom and chilies. *Spices Bull. (Annu. No.)* **2** (12), 115.

Narasimhamurthy, G., and Ranganathan, S. (1937). The nitrogen complex of Indian food stuffs. 1. Black pepper (*Piper nigrum*). *Indian J. Med. Res.* **25**, 373.

Narayana, C., Somayajulu, B. A. R., and Thirumala Rao, S. D. (1967). Recovery of fatty oil from spent seeds of ajowan. (*Trachyspermum ammi* Linn.). *Indian J. Technol.* **5**, 268.

Narayanamurthy, D., and Rao, P. R. (1961). Paper adhesive from tamarind kernel powder. *Res. Ind.* **6**, 127.

Narayanan, C. S., and Natarajan, C. P. (1977). Improvements in the flavour quality of (cardamom) oil from cultivated Malabar type cardamom grown in Karnataka State. *J. Food Sci. Technol.* **14**, 233.

Narayanan, K. M., Kapur, N. S., Bains, G. S., and Bhatia, D. S. (1957). Effect of vanillin on stability of vegetable shortening. *Food Sci.* **6**, 245.

Narayanan, K. M., Dwarakanath, C. T., Ramachandra Rao, T. N., and Johar, D. S. (1964). Studies on pepper oleoresin from pepper rejections. Part 1. Extraction of oleoresin from pepper rejections. Part II. Storage studies on oleoresin. *Spices Bull.* **3**(8), 117.

Narayanaswamy, D., Sreenathan, V. R., Miss Padmini, T., Murthy, H. B. N., and Iyengar, N. V. R. (1962). Packaging studies on ground pepper. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract p. 17.

Natarajan, C. P. (1967). Some aspects of spice research. *Indian Food Packer* **21**(3), 12.

Natarajan, C. P. (1974). Post-harvest technology of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 4.

Natarajan, C. P., and Shankaracharya, N. B. (1972). Microbial quality standards for foods. *Food Ind. J.* **5**(1), 11.

Natarajan, C. P., Kuppuswamy, S., Krishnamurthy, M. N., D'Souza, T., and Gopalan, K. K. (1967a). Preservation of green colour in cardamom. *Indian Spices* No. 1, p. 5.

Natarajan, C. P., Lewis, Y. S., Nambudiri, E. S., and Krishnamurthy, M. N. (1967). Production of white pepper, pepper oil and oleoresin. *Indian Spices* **3**, 41.

Natarajan, C. P., Kuppuswamy, S., and Krishnamurthy, M. N. (1968a). A study on the maturity, regional variations and retention of green colour of cardamom. *J. Food Sci. Technol.* **5**, 65.

Natarajan, C. P., Mahadeviah, B., and Shivashankar, S. (1968b). Chemical, technological and physiological aspects of chillies. *Indian Spices (Chilies Semin. No.)* **6**(3), 49.

Natarajan, C. P., Krishnamurthy, M. N., Padmabai, R., and Kuppuswamy, S. (1969). "Annual Report." CFTRI, Mysore, India.

Natarajan, C. P., Kuppuswamy, S., Shankaracharya, N. B., Padmabai, R., Raghavan, B., Krishnamurthy, M. N., Fazullah Khan, K., Lewis, Y. S., and Govindrajan, V. S. (1970). Product development of ginger. *Indian Spices* **7**, 8.

Natarajan, C. P., Padmabai, R., Krishnamurthy, M. N., Raghavan, B., Shankaracharya, N. B., Kuppuswamy, S., Govindarajan, V. S., and Lewis, Y. S. (1972). Chemical composition of ginger varieties and dehydration studies on ginger. *J. Food Sci. Technol.* **9**, 220.

National Agricultural Advisory Service (NAAS) of the Ministry of Agriculture, London (1970). "Dry Bulb Onions," Booklet No. 1. NAAS Hortic. Enterprises, Minist. Agric. Fish. Food Publ., U.K.

Navarao, S., Ortuno, A., and Oosta, F. (1965). Thin-layer chromatographic determination of synthetic dyes in foods. I. Fat-soluble azo dyes in paprika. *An. Bromatol.* **17**, 269.

Naves, Y. R. (1959). Analysis of thyme oil by gas chromatography. *Fr. Perfums* **2**, 23.

Naves, Y. R. (1963). Errors in the application of instrumentation to the analysis of perfumery raw materials. *J. Soc. Cosmet. Chem.* **14**, 29.

Naves, Y. R., and Tucakov, J. (1959). Presence of anetholes in Yugoslav fennel oils. *C. R. Hebd. Seances Acad. Sci.* **248**, 843.

Nawar, W. W., and Fargerson, I. S. (1962). Direct gas chromatographic analysis as an objective method of flavour measurement. *Food Technol.* **16,** 107.

Nazarov, B. B. (1959). Determination of the quality of a liquid extract of thyme. *Uch. Zap., Pyatigorsk. Farm. Inst.* **4,** 181; *Chem. Abstr.* **56,** 6104h (1962).

Neale, M. W. (1963). Spices in food products manufacture. *Canner/Packer* **32,** 41.

Neale, M. W., and Klis, J. B. (1964). A guide to spices. Natural types, processed forms and uses in food products. *Food Process., Spec. Rep.*

Nelson, E. K. (1919). The constitution of capsaicin, the pungent principle of capsicum. *J. Am. Chem. Soc.* **41,** 1115.

Nelson, E. K., and Dawson, L. E. (1923). The constitution of capsaicin, the pungent principle of capsicum. *J. Am. Chem. Soc.* **45,** 2179.

Netien, G., and Mery, L. (1962). Chromatography of safron adulterations. *Bull. Trav. Soc. Pharm. Lyon* **6,** 35.

Neuzil, E., Jensen, H., and Josselin, J. (1967). Color reaction of vanillin with millon reagent. I. Spectrophotometric study and reaction mechanism. *Ann. Pharm. Fr.* **25,** 127.

Newman, A. A. (1953). Natural and synthetic pepper flavoured substances. *Chem. Prod. Chem. News* **16,** 413.

Nichols, S. R. (1952). "Aids to the Analysis of Foods and Drugs." Basthiers, London.

Niegisch, W. D., and Stahl, W. H. (1956). The onion. Gaseous emanation products. *Food Res.* **21,** 657.

Nigam, I. C. (1966). Analysis of volatile oils. *In* "Pharmaceutical Chemistry" (L. G. Chatten, ed.), p. 439. Dekker, New York.

Nigam, I. C., and Levi, L. (1963). Sabinene hydrate and sabinene acetate. Two new constituents of American speermint oil. *J. Agric. Food. Chem.* **11,** 276.

Nigam, I. C., and Levi, L. (1964). Essential oils and their constituents. XX. Detection and estimation of menthofuranin in *Mentha arvensis* and other mint species by coupled gas-liquid and thin-layer chromatography. *J. Pharm. Sci.* **53,** 1008.

Nigam, I. C., Gupta, G. N., and Dhingra, D. R. (1960). Chemical examination of the essential oil from laurel (*Laurus nobilis*) berries. *Perfum. Essent. Oil Rec.* **51,** 351.

Nigam, I. C., Skakum, W., and Levi, L. (1963a). Determination of trace constituents of oil of ajowan. *Perfum. Essent. Oil Rec.* **54,** 25.

Nigam, I. C., Sahasrabudhe, M., and Levi, L. (1963b). Coupled gas-liquid thin-layer chromatography. Simultaneous determination of piperitone and piperiton oxide in essential oils. *J. Chem.* **41,** 1535.

Nigam, M. C., and Handa, K. L. (1964). Gas chromatographic examination of Indian pepper oil. *Indian Perfum.* **8,** Part I, 15.

Nigam, M. C., Nigam, I. C., and Levi, L. (1964). Essential oils and their constituents, XXII. Detection of new trace components in oil of ginger. *Can. J. Chem.* **42,** 2610.

Nigam, M. C., Nigam, I. C., Handa, K. L., and Levi, L. (1965a). Essential oils and their constituents. XXVIII. Examination of oil of cardamom by gas chromatography. *J. Pharm. Sci.* **54,** 799.

Nigam, M. C., Nigam, I. C., and Levi, L. (1965b). Essential oils and their constituents. XXV. Thin layer chromatography, some chemical and chemitaxonomic applications. *J. Soc. Cosmet. Chem.* **16,** 155.

Nigam, M. C., Handa, K. L., and Rao, P. R. (1970). Gas chromatography of the essential oil of sweet basil: A potential source of linalool in India. *Parfuem. Kosmet.* **51,** 151.

Nigam, S. S., and Kumari, G. L. (1963). Application of physical methods for the analysis of essential oils. II. Colorimetry. *Riechst. Aromen* **13,** 1.

Nigam, S. S., and Purohit, R. M. (1960a). Chemical examination of the essential oil derived from the seeds of *Ammomum subulatum* Roxb. *Perfum. Essent. Oil Rec.* **51,** 121.

Nigam, S. S., and Purohit, R. M. (1960b). Chromatography of the essential oil of *Ammomum subulatum* Roxb. *Perfum. Essent. Oil Rec.* **51,** 121.

Nigam, S. S., and Purohit, R. M. (1961). Chromatography of the essential oil of *Murraya koenigii, Perfum. Essent. Oil Rec.* **52,** 643.

Nigam, S. S., and Radhakrishnan, C. (1965). Chemical investigation of the essential oil obtained from the berries of *Piper longum* L. *Riochst., Aromen, Koerperpflegem.* **15,** 451.

Nigam, S. S., and Radhakrishnan, C. (1966). Preliminary investigations of a new hydrocarbon isolated from the oil of *Piper longum* L. *Riechst., Aromen, Koerperpflegem.* **16,** 1.

Nikolaev, K. (1962). Toxicological studies on *Sinapsis arvensis*. I. The amount of active principle in plant. *I-z. Vet. Nauchnoizsled. Inst. Nezarazni Boles., Zookhig., Akad. Selskostop. Nauki Bulg.* **2,** 101.

Noding, A., Stoa, K. F., and Nordal, A. (1950). Investigation of the estrogenic effect of extracts of Ceylon cinnamon. *Medd. Nor. Farm Selsk.* **12,** 68.

Nogrady, G. (1943). Determination of the spiciness of paprika by the fluoroscence titration of capsaicin. *Kiserletugyi Kozl.* **46,** 160.

Nomura, H. (1917a). Pungent principles of ginger (I). Zingiberone. *Sci. Rep. Tohoku Imp. Univ.* **6,** 41.

Nomura, H. (1917b). Pungent principles of ginger (II). Zingiberone. *J. Chem. Soc.* **111,** 769.

Nomura, H. *et al.* (1918a). Shogaol—pungent constituent of ginger. *Sci. Rep. Tohoku Imp. Univ.* **17,** 67.

Nomura, H. *et al.* (1918b). *Sci. Rep. Tohoku Imp. Univ.* **18,** 661; cf. *Proc. Imp. Acad. (Tokyo)* **5,** 159 (1927).

North, H. (1949). Colorimetric determination of capsaicin in oleoresin of capsicum. *Anal. Chem.* **21,** 934.

Novak, A. F., and Fisher, M. J. (1964). Antimycotic activity of some fatty acid derivatives. *J. Am. Oil Chem. Soc.* **41,** 503.

Okajima, M. J. (1954). Constituents of the pigmented outer skin of onion bulb. I. From the waxy matter of the skin. *Sci. Res. Inst. (Tokyo)* **48,** 281.

Oliver, R. (1972). Methods for the study of the aromatic components of vanilla extracts. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 55.

Orsi, F. (1970). Gas chromatography determination of the acetone content of paprika oleoresin. *Konzerv- Paprikaip.* **3,** 112.

Osisiogu, U. W. (1967). Essential oils of Nigeria. II. A study of the fennel oil produced at Nsukka. *Planta Med.* **15,** 30; *Chem. Abstr.* **66,** 108169 (1967).

Ozewalla, N., and Willms, M. (1962). Yield of volatile oil from cardamom seeds. *Proc. Okla. Acad. Sci.* **43,** 57.

Oztune, S. (1940). Terkiyede meyvelerin sira halinde kiy-mithlendiril. mesive hardaliye (The preservation of fruit juices by the addition of mustard). Ph.D. Dissertation, Y. Z. E., Ankara, Turkey.

Palitzsch, A. (1974). Effect of spice extracts on fat breakdown. *Fleischwirtschaft* **54,** 63.

Palotas, J., and Koneesni, 1. (1964). Applicability of the ASTA method for the investigation of paprika pigments. *Elelmiszervizsgalati Kozl.* **10,** 200.

Palotas, J., Lorant, B., and Mahilik, D. (1966). Determination of moisture in ground paprika by drying and according to the acetyl chloride method. *Nahrung* **10,** 37.

Pan, H. P. (1960). Burning-numbing taste in celery. *Proc. Fla. State Hortic. Soc.* **73,** 223.

Pandey, S. C., and Bhole Nath (1960). A new method for augmenting the allyl isothiocyanate content in the oil milling of rape and mustard seeds. *J. Oil Technol. Assoc. India* **16,** Part I, 15.

Pangborn, R. M., Jennings, W. G., and Noelting, C. E. (1970). Preliminary examination of odour quality of black pepper oil. *Flavour Ind.* **1,** 763.

Papanstassiou, Z. B., Bruni, R. J., and Edward, W. (1967). Potential carcinolytic agents. V. Enamine mustards. *J. Med. Chem.* **10,** 701.

Parczewski, A. (1962). Research on essential oils of the Polish umbelliferal plants. *Riv. Ital. Essenze, Profumi, Piante Off., Olii Veg., Saponi* **44,** 454.

Parekh, C. M., Pruthi, J. S., and Tandon, G. L. (1958). Apparent SO_2 in fresh and pickled onions. *Indian Food Packer* **12**(9), 9.

Paris, R. (1941). Identification of olive kernels. *Bull. Sci. Pharmacol.* **43,** 17; *Chem. Abstr.* **36,** 3471 (1942).

Paris, R., and Godon, M. (1961). Analysis of essential oils by paper and glass plate, chromatography. Applications to some medicinal oils. *Ann. Pharm. Fr.* **19,** 86.

Paris, R., and Godon, M. (1963). Thin-layer chromatography of essential oils. *Recherches* **13,** 48; *Chem. Abstr.* **61,** 28996 (1964).

Paris, R. R., and Rousselet, R. (1958). Characterization of dyes of vegetable origin by paper chromatography. *Ann. Pharm. Fr.* **16,** 747.

Paris, R. R., Faugeras, G., Balard, D., and Capmal, M. (1961). Estimation of morphine in papaver capsules by paper chromatography and paper electrophoresis. *Ann. Pharm. Fr.* **19,** 494.

Parker, K. D., Fontan, C. R., and Kirk, P. L. (1963). Rapid gas chromatographic method for screening of toxicological extracts for alkaloids, barbiturates, sympathominetic amines and tranquilizers. *Anal. Chem.* **35,** 356.

Parry, E. J., and Bird, E. C. J. (1925). Adulteration of saffron. *Chem. Drug.* **103,** 445. *Chem. Abstr.* **20,** 2389 (1926).

Parry, J. W. (1945). "Handbook of Spices." Chem. Publ. Co., New York.

Parry, J. W. (1953). "The Story of Spices." Chem. Publ. Co., New York.

Parry, J. W. (1962). "Spices—Their Morphology, Histology and Chemistry," 1st ed. Chem. Publ. Co., New York.

Patakova, D., and Chladek, M. (1966). A contribution to the evaluation of fruit quality in foreign varieties of coriander. *Bull. Vysk. Ustavu Zelin Olomouse* **10,** 129.

Patil, J. A. (1960). Onion storage. *Poona Agric. Coll. Mag.* **51,** 33.

Patil, J. A. (1966). Gamma irradiation for vegetable storage. A. Onion bulbs. *Coll. Agric. Parbhani Mag.* **7,** 3.

Patwardhan, M. V., and Sastry, L. V. L. (1957). Effect of asafoetida on the intestinal (wall) enzyme systems *in vitro*. *Food Sci.* **6,** 27.

Peleg, Y., and Mannheim, C. H. (1969). Caking of onion powder. *J. Food Technol.* **4,** 157.

Peleg, Y., Mannheim, C. H., and Berk, Z. (1970). Changes in quality of dehydrated kibbled onions during storage. *J. Food Sci.* **35,** 513.

Pen, R. Z. (1966). Colorimetric determination of vanillin with 2, 4 divertrophenyl hydrazine. *Tr. Sib. Tekhnol. Inst.* **38,** 151.

Pequin, L. (1961). Etiolation and oxidation activity in some Solinaceae. Catalase and peroxidase. *C. R. Hebd. Seances Acad. Sci.* **253,** 2108.

Perazza, A., and Prado, L. (1937). The action of mustard essence on wines. Its use as an antifermentative agent. *Rev. Asoc. Bioquim. Argent.* **5,** 54; *Chem. Abstr.* **32,** 6394 (1938).

Perry, T. L., Hansen, S., Hestrin, M., and MacIntyre, E. (1965). Exogenous urinary amines of plant origin. *Clin. Chim. Acta* **11,** 24.

Pertsev, I. M. (1963). Application of chromatography to the investigation of essential oil used in the pharmaceutical practice. *Farm. Zh. (Kiev)* **18,** 33; *Chem. Abstr.* **61,** 1703 (1964).

Pertsev, I. M., and Pivnenko, G. P. (1961). Chromatography of essential oils used in pharmacy. Micro-analysis of essential oils of peppermint on thin-layer adsorption plates. *Farm. Zh. (Kiev)* **16** (15), 28; *Chem. Abstr.* **55,** 13720f (1963).

Pertsev, I. M., and Pivnenko, G. P. (1962). Chromatography of essential oils used in pharmacy. III.

Micro-analysis of essential oils of coriander, levender and nutmeg by thin layer chromatography. *Farm. Zh. (Kiev)* **17,** 35.

Peryam, D. R. (1958). Sensory difference tests. *Food Technol.* **12,** 231.

Petkov, V. (1966). Pharmacological and clinical studies of garlic. *Dtsch. Apoth.-Ztg.* **106,** 1861.

Petkov, V., Penova, M., Paparkova, K., and Jekov, S. (1969). Screening studies on the antimicrobial action of plants growing in Bulgaria. Antibacterial activity of spices. **16**(2), 11.

Petrusenko, R. I. (1967). The treatment of arsenical periodontites. *Stomatologiya (Moscow)* **46**(2), 32.

Peyron, L. (1961). Precursors of certain sulfur compounds encountered with essential oils. *Bull. Soc. Fr. Physiol. Veg.* **7,** 46.

Peyron, L., Benezet, L., De Dortan, D., and Garnero, J. (1969). On the presence of 1,8-terpin in star-anise and sweet fennel oils. *Bull. Soc. Chim. Fr.* **1,** 339.

Pfiefer, I. S. (1962). The opium poppy, medicinal plant for more than two thousand years. *Pharmazie* **17,** 467.

Pfiefer, S., and Banerjee, S. K. (1965). Red coloured alkaloids of the genus papaver. IV. Papaverrubins. *Arch. Pharm. (Weinheim, Ger.)* **298,** 385; cf. *Chem. Abstr.* **62,** 14990d (1965).

Pfiefer, S., and Heydenreich, K. (1961). The alkaloids of germinating poppy plants. *Naturwissenschaften* **48,** 222.

Phillip, T. (1969). Packaging of spices in flexible films. *Indian Spices* **6**(1), 15.

Phillip, T., and Francis, F. J. (1971a). Oxidation of capsanthin. *J. Food Sci.* **36,** 96.

Phillip, T., and Francis, F. J. (1971b). Isolation and chemical properties of capsanthin and derivatives. *J. Food Sci.* **36,** 823.

Phillip, T., Nawar, W. W., and Francis, F. J. (1971). The nature of fatty acids and capsanthin esters in paprika. *J. Food Sci.* **36,** 98.

Phokas, G. (1966). Thin-layer chromatography of the essential oils of *Melisa officinalis* and some of its adulterants. *Pharm. Delt., Epistem. Ekdosis* **5,** 9; *Chem. Abstr.* **65,** 3643 (1960).

Piedelievre, R., and Derobert, L. (1942). *Ann. Med. Leg.* **22,** 82; *Chem. Zentralbl.* **11,** 439 (1943); *Chem. Abstr.* **38,** 5305 (1944).

Piedelievre, R., and Derobert, L. (1943). *Chem. Zentralbl.* **11,** 439; *Chem. Abstr.* **38,** 5305 (1944).

Pillai, K. S. (1973). The clove. *Arecanut Spices Bull.* **4**(3), 9

Pillai, S. P., Muthu, M., Sharangapani, M. V., Majumder, S. K., and Amla, B. L. (1970). Development of a gas-proof strip pack of ethylene dibromide. *Inst. Pest Control* **12,** 23.

Pithawala, H. R., and Sreenivasan, A. (1950). Indian Patent 40,835.

Platenius, H. A. (1935). A method for estimating the volatile sulfur content and pungency of onion. *J. Agric. Res.* **51,** 847.

Platenius, H. A., and Knott, J. E. (1941). Factors affecting onion pungency. *J. Agric. Res.* **62,** 37.

Pleat, G. B., Harley, J. H., and Wiberley, S. E. (1952). Use of infrared spectra in the qualitative and quantitative determination of alkaloids. *J. Am. Pharm. Assoc.* **40,** 107.

Pogany, D., Bell, C. L., and Kirch, E. R. (1968). Composition of oil of sweet basil (*Ocimum basilicum* L.), obtained from plants grown at different temperatures. *Perfum. Essent. Oil Rec.* **59,** 858.

Pohja, M. S. (1957). Microbial counts of solid and liquid spices. *Fleischwirtschaft* **9,** 547.

Pohle, W. D., and Gregory, R. L. (1960). Colour of capsicum species. *Food Technol.* **14,** 245.

Polyakov, A., and Falina, L. (1962). Spectrophotometric determination of citral in oxidized coriander oil. *Russ. Uch. Lavadenie, Pischeraya Tekhn.* **6,** 139.

Pomerantz, R., Goldblith, S. A., and Procter, B. E. (1957). Rapid spectrophotometric technique for evaluation of vanilla extracts. *J. Agric. Food Chem.* **5,** 292.

Pomini, A. (1962). The Juniper. *Minerva Farm.* **11,** 207.

Pomini, A. (1963). Caraway. *Minerva Farm.* **12,** 61.

Porcaro, P. J., and Johnston, V. D. (1961). Determination of menthols by gas chromatography. *Anal. Chem.* **33,** 1748.

Porsch, F., Farnow, H., and Winkler, H. (1965). Star anise oil. *Dragoco Rep. (Engl. Ed.)* **12,** 119.

Portales, J. V., and Honrubia, P. G. (1967). Extraction of pepper oil. Spanish Patent 321,890.

Poulose, T. T. (1967). Credit and marketing facilities for pepper. *Indian Spices* **4,** 36.

Powers, E. M., Lawyer, R., and Masuoka, Y. (1975). Microbiology of processed spices. *J. Milk Food Technol.* **38,** 683.

Powers, J. J., and Shinholser, K. (1979). pH and buffering capacity of canned green peppercorn. *J. Food Sci.* **44**(6).

Powers, J. J., Prat, D. E., Downing, D. L., and Powers, 1. T. (1961). Effect of acid level, calcium salts, monosodium glutamate, and sugar on canned pimientos. *Food Technol.* **15,** 67.

Powers, J. J., Morse, R. E., Sane, R. H., and Mills, W. C. (1950). Acidification and calcium firming of canned pimientos. *Food Technol.* **4,** 485.

Prakash, V., and Natarajan, C. P. (1974). Studies on curry leaf (*Murraya Koenigii* L.). *J. Food Sci. Technol.* **11,** 284.

Prasad, D. N., Bhattacharya, S. K., and Das, P. K. (1966). A study of anti-inflammatory activity of some indigenous drugs in albino rats. *Indian J. Med. Res.* **57,** 582.

Prasad, H. H., and Joshi, N. V. (1929). The preserving value of spices used in pickling raw fruits in India. *Agric. J. India* **24,** 402.

Pravatoroff, N. (1967). Ginger—the properties and chemistry of some natural spicy compounds. *Manuf. Chem. & Aerosol News* **38,** 40.

Pravatoroff, N. (1972). Some details of the distillation of spice oils. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 173.

Preuss, R. (1964). Photochemical mechanism of aging of the caraway oil components. *Dtsch. Apoth.-Ztg.* **104,** 1797; *Chem. Abstr.* **62,** 10287f (1965).

Processed Foods Export Promotion Council, New Delhi (1974). Export of processed foods during 1973–74. *Profodcil Bull.* **9,** 30.

Proctor, B. E., and Goldblith, S. A. (1951). Food processing with ionizing radiations. *Food Technol.* **5,** 376.

Proctor, B. E., Goldblith, S. A., and Fram, H. (1950). Effect of super-voltage cathode rays on bacterial flora of spices and other dry food materials. *Food Res.* **15,** 490.

Pruidze, V. (1965). Physical and chemical properties of dry leaves of *Laurus nobilis*. *Konservn. Ovoshchesush. Prom.* **20,** 16.

Pruidze, V. G. (1968). Changes in the dry bay leaf during storage. *Subtrop. Kul't.* **2,** 134.

Pruidze, V. G. *et al.* (1971). Essential oil changes during storage of bay leaves. *Tr. Gruz., Nauchno-Issled. Inst. Pishch. Prom-Sti.* **5,** 151.

Pruthi, J. S. (1957). Spin-pasteurizer—its principle and applications. *Food Sci.* **6,** 179.

Pruthi, J. S. (1964). Chemistry, microbiology and technology of curry powders. *Spices Bull.***3**(6), 7

Pruthi, J. S. (1966). Review on the chemistry and quality evaluation of spices. Part I. Curry powders. *Agmark Sci. Bull.* **1,** 1.

Pruthi, J. S. (1968a). Reviews on the chemistry and quality evaluation of spices. Part II. Black pepper. *Indian Spices* **5,** 11.

Pruthi, J. S. (1968b). Technical report on the meetings of the ISO/TC-34 Technical committee and sub-committee SC-7 on spices and condiments, held in London (UK) from 29th October '68 to 1st Nov. '68. *Cent. Agmark Lab. Nagpur (India), Tech. Commun.* No. CAL/16/68.

Pruthi, J. S. (1969a). International collaborative studies on pungency (capsaicin) and colour (capsanthin content) of Hungarian ground red paprika. *Cent. Agmark Lab. Nagpur (India), Tech. Commun.* No. CAL/30/69, ISO/TC-34/SC-7-DOC.

Pruthi, J. S. (1969b). Analytical techniques for the determination of piperine/pungent principles in black and white pepper (*Piper nigrum* L.). *Indian Spices* **7**(2), 14.

Pruthi, J. S. (1969c). *Ibid. Indian Spices* **7**(3), 5.

Pruthi, J. S. (1969d). *Ibid. Indian Spices* **7**(4), 13.

Pruthi, J. S. (1970a). "Analytical Techniques for the Determination of Piperine/Pungent Principles in Black and White Pepper (*Piper nigrum* L)," ISO (Int. Organ. Stand.) Doc. No. 290E, pp. 1–35. Int. Organ. Stand., Hungary.

Pruthi, J. S. (1970b). "International Collaborative Studies on the Standardization of Methods for the Estimation of Piperine in Black and White Pepper," 1st rep, ISO/TC 34-SC 7, Doc. No. 219E, p. 1. Int. Organ. Stand., Hungary.

Pruthi, J. S. (1970c). "International Collaborative Work on the Determination of Capsanthin and Capsaicin in Ground Paprika," ISO/TC 34-SC 7, Doc. No. 227. Int. Organ. Stand., Hungary.

Pruthi, J. S. (1970d). Packaging of spices and condiments. *J. Packag. (India)* **3**(1), 11.

Pruthi, J. S. (1972). Marketing and standardization of oil-seeds. *Indian Oil Soap J.* **37**, 288a.

Pruthi, J. S. (1974). Quality control and standardization of spices. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 49.

Pruthi, J. S. (1976). "Spices and Condiments," 1st ed. Natl. Book Trust, India, New Delhi.

Pruthi, J. S. (1979a). Cinnamon—its taxonomy, agronomy, processing and quality control. *Arecanut Spices Bull.* (communicated).

Pruthi, J. S. (1979b). Spice essential oils—their production, packaging, storage and uses. Paper presented at the *International Seminar on Essential Oils,* International Trade Centre, Geneva and Chemixcil (India), Bangalore, Oct. 29–Nov. 2, 1979.

Pruthi, J. S. (1980a). Quality control, packaging and storage of ginger. *Proc. Natl. Seminar on Ginger and Turmeric,* Calicut, Kerala, India, April 8–9, 1980.

Pruthi, J. S. (1980b). Quality control, packaging and storage of turmeric. *Proc. Natl. Seminar on Ginger and Turmeric:* Calicut, Kerala, India, April 8–9, 1980.

Pruthi, J. S. (1980c). Spice essential oils—Their use in perfumery and cosmetics. *Pafai J.* (Bombay), **2**(2).

Pruthi, J. S., and Kulkarni, B. M. (1969). A simple technique for the rapid and easy detection of papaya seeds in black pepper berries. *Indian Food Packer* **23**(1), 51.

Pruthi, J. S., and Lakshmishankar. (1968a). Rapid determination of moisture in important spices and condiments - calibration of Marconi moisture meter. *Indian Spices* **5**(4), 20.

Pruthi, J. S., and Lakshmishankar. (1968b). Rapid determination of moisture in chilies. *Indian Spices* **6**(3), 56.

Pruthi, J. S., and Lal, G. (1960). Studies on equilibrium relative humidity (ERH) for onion powder. *Curr. Sci.* **29**, 19.

Pruthi, J. S., and Misra, B. D. (1963a). Packaging requirements of Indian curry powders and other spice mixtures. *Spices Bull.* **2**(6), 5.

Pruthi, J. S., and Misra, B. D. (1963b). Quality standards for curry powders and other spice mixtures. *Spices Bull. (Annu. No.)* **2**(12), 97.

Pruthi, J. S., and Misra, B. D. (1963c). Physico-chemical and micro-biological changes in curry powders during drying, milling and mixing operations. *Spices Bull.* **3**(3–5), 8.

Pruthi, J. S., and Satyavati, K. K. (1980). A study of factors governing the recovery and quality of pectin from nutmeg waste (husk). *Indian Food Packer* (submitted for publication).

Pruthi, J. S., and Srivas, S. R. (1962). An integrated process for the recovery of volatile oil, oleoresin and starch from ginger. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract, p. 11.

Pruthi, J. S., and Subrahmanyan, V. (1962). A simple process for the manufacture of garlic powder. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract, p. 9.

Pruthi, J. S., Lal, G., and Subrahmanyan, V. (1958). Improvements in or relating to the manufacture of garlic powder. CSIR (India) Patent 65,138.

Pruthi, J. S., Lal, G., and Subrahmanyan, V. (1959a). Chemistry and technology of garlic powder. *Food Sci.* **8**, 429.

Pruthi, J. S., Singh, L. J., and Lal, G. (1959b). Determination of the critical temperature of dehydration of garlic. *Food Sci.* **8**, 436.

Pruthi, J. S., Singh, L. J., and Lal, G. (1959c). Some technological aspects of dehydration of garlic—a study of some factors affecting the quality of garlic powder during dehydration. *Food Sci.* **8**, 441.

Pruthi, J. S., Singh, L. J., Kalbagh, S. S., and Lal, G. (1959d). Effect of different methods of dehydration on the quality of garlic powder. *Food Sci.* **8**, 444.

Pruthi, J. S., Singh, L. J., Ramu, S. D. V., and Lal, G. (1959e). Pilot-plant studies on the manufacture of garlic powder. *Food Sci.* **8**, 448.

Pruthi, J. S., Singh, L. J., Indiramma, K., Sankaran, A. N., and Lal, G. (1959f). Effect on nitrogen packaging and storage temperature on the quality of garlic powder. *Food Sci.* **8**, 461.

Pruthi, J. S., Ramanathan, P. K., and Lal, G. (1959g). Improvements in or relating to spin-pasteurizer for canned acid foods. Indian Patent 69,697.

Pruthi, J. S., Singh, L. J., and Lal, G. (1959h). The equilibrium relative humidity of garlic powder. *J. Sci. Food Agric.* **7**, 359.

Pruthi, J. S., Singh, L. J., and Pruthi, J. S. (1959i). Thermal stability of allinase and enzymatic regeneration of flavour in odorless garlic powder. *Curr. Sci.* **28**, 403.

Pruthi, J. S., Lal, G., and Subrahmanyan, V. (1960a). Utilization of ginger (*Zingiber officinale*). I.C.A.R. souvenir. *All-Indian Spices Cashewnut Exhibition, 3rd., 1960,* p. 17.

Pruthi, J. S., Singh, L. J., and Lal, G. (1960b). Non-enzymatic browning in garlic powder during storage. *Food Sci.* **9**, 243.

Pruthi, J. S., Ramu, S.D.V., and Jayaraman, A. (1962). Studies on the chemical composition and sorption isotherms of spices and spice products. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract, p. 10.

Pruthi, J. S., Bhat, A. V., and Varkey, A. G. (1974a). "Cinnamon Leaf Oil," Annu. Rep. CFTRI Exp. Stn., Trichur, Kerala, India.

Pruthi, J. S., Satyavathi, V. K., Varkey, A. G., Bhat, A. V., and Gopalakrishnan, M. (1974b). "Processing of Tender Green Pepper (*Piper nigrum*) and Formulation of Quality Standards Therefor," Annu. Rep., p. 5. CFTRI Exp. Stn., Trichur, Kerala, India.

Pruthi, J. S., Bhat, A. V., and Varkey, A. G. (1974c). "Determination of the Optimum Conditions and Methods of Harvesting, Curing and Drying of Cinnamon and Utilization of Cinnamon Waste," Annu. Rep., p. 11. CFTRI Exp. Stn., Trichur, Kerala, India.

Pruthi, J. S., Varkey, A. G., and Bhat, A. V. (1975). "Post-harvest Technology of Cinnamon and Utilization of Cinnamon Waste," Annu. Rep. CFTRI, Mysore, India.

Pruthi, J. S., Satyavati, K. K., Gopalakrishnan, M., Bhat, A. V., and Varkey, A. G. (1976a). Thermal Processing (canning) of green pepper (*P. nigrum* L.) and development of quality standards therefor. *Food Technol.* **30**, 356 (abstr.).

Pruthi, J. S., Bhat, A. V., Satyavati, K. K., Varkey, A. G., and Gopalakrishnan, M. (1976b). Preservation of fresh green pepper by canning, bottling and other methods of preservation. *Proc. Int. Semin. Pepper, Spices Export Promotion Counc. India, Ernakulam* p. 15.

Pruthi, J. S., Bhat, A. V., Satyavati, K. K., and Gopalkrishnan, M. (1978a). Studies on the chemical composition and utilization of mace, nutmeg and nutmeg waste. *Indian Arecanut, Spices, Cocoa J.* (submitted for publication).

Pruthi, J. S., Varkey, A. G., Satyavati, K. K., Bhat, A. V., and Gopalakrishnan, M. (1978b). Discolouration in canned and bottled green pepper. *Indian Food Packer* (submitted for publication).

Pruthi, J. S., Varkey, A. G., and Bhat, A. V. (1978c). Studies on post-harvest technology of cinnamon and utilization of cinnamon waste. (Leaves and twigs.) *Indian Arecanut, Spices, Cocoa J.* **1**, 87.

Putkaradze, S. A., and Taudgiridz, E. G. N. (1970). The essential oil content of bay leaf oil. *Subtr. Kul't.* **4**, 79 (in Russian); Hortic. Abstr. **40**, 6973 (1970).

Rabinowitsch, C. (1950). Experimental studies on the influence of the spices on the formation of the gastric juice. *Ges. Physiol. Pathol. Stoffwechselskr* [N.S.] **2**, 822.

Rader, B. R. (1966). Moisture in dried vegetables and spices. *J. Assoc. Off. Anal. Chem.* **49**, 726.

Raghavachari, P. S. (1963). Marketing of cardamoms. *Spices Bull. (Annu. No.)* **3**(12), 39.

Raghavan, B., Abraham, K. O., Shankaranarayana, M. L., Sastri, L. V. L., and Natarajan, C. P. (1974). Asafoetida. II. Chemical composition and properties. *Flavour Ind.* **5**, 179.

Raghubanshi, C. S. (1971). Marketing of hill capsicum in Saproon valley (Himachal Pradesh). *J. Agric. Mark.* **13**(4), 6.

Raghunandan Rao, R., Srinivasa Rao, S., and Venketaraman, P. R. (1946). Investigations on plant antibiotics. Part I. Studies on allicin—the antibacterial principle of *Allium sativum* (garlic). *J. Sci. Ind. Res., Sect. B* **5**, 31.

Raina, B. L., Teotia, M. S., and Pruthi, J. S. (1978). Technological assessment of ginger grown in Himachal Pradesh. *Proc. 1st Annu. Conv. Food Sci. Technol.* p. 47.

Rainova, T. L., and Penova, M. (1967). Uterine effect of parsley (*Petroselinum sativum*). II and IV. *Farmatsiya (Sofia)* **17**(4), 39.

Rajapakse-Arambewela, L. S., and Wijesekera, R. O. B. (1979). GLC study of the essential oil of wild cardamom oil of Sri Lanka. *J. Sci. Food. Agri.* **30**, 521.

Rajlakshmi, R., Nanavati, K., and Gumasth, A. A. (1964). Effect of cooking procedures on the free and total niacin content of certain foods. *J. Natl. Diet. (Coimbatore, India)* **1**, 276.

Rakova, N. V. (1961). Change of the assay of the essential oil upon storage of coriander seeds. *Spirt. Prom-st.* **27**, 13; cf. *Chem. Abstr.* **55**, 26375e (1961).

Rakuzin, M. A., and Starobina, A. (1924). Fat content of the fruits of the most important Umbelliferae. *Landwirtsch. Vers.-Stn.* **103**, 103.

Ramachandra Rao, T. N., Dwarkanath, C. T., and Johar, D. S. (1959). Peppersal—a new flavouring agent from waste pepper. Indian Patent 67,931.

Ramachandra Rao, T. N., Dwarkanath, C. T., and Johar, D. S. (1960). Analysis of piperine by spectrophotometric methods. Part 1. *J. Proc. Inst. Chem., Calcutta* **32**, Part III, 125.

Ramalanjaona, G. I., and Jourdan, E. (1968). The essential oils of Madagascar. *Proc. Int. Conf. Essent Oils, Trop Prod. Inst. London, 1967* p. 21.

Ramanathan, P. K., and Srinivasa Rao, P. N. (1974). Equipment needs of spice industry. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 82.

Ramanujam, S., and Tewari, V. P. (1968). Production of chilies in various states of India and their commercial classification. *Indian Spices (Chilies Semin. No.)* **6**(3), 13.

Ramaswamy, T. S., and Banerjee, B. N. (1948). Vegetable dyes as antioxidant for vegetable oils. *Ann. Biochem. Exp. Med.* **8**, 55.

Ramprasad, C., and Sirsi, M. (1956). Studies on Indian medicinal plants. *Curcuma longa* Linn—effect of curcumin and the essential oil of *C. longa* on bile secretion. *J. Sci. Ind. Res., Sect. C* **15**, 262.

Randhawa, K. S., and Nandpuri, K. S. (1966). Effect of plant growth regulators on sprouting of onions under ordinary storage conditions. *J. Agron.* **11**, 238.

Rao, B. A. S., Misra, B. D., and Pruthi, J. S. (1962). Microbiological examination of curry powders and spice mixtures. *Proc. Symp. Spices.—Role Natl. Econ., 1st, 1962* Abstract, p. 3.

Rao, B. G. V. N., and Rao, P. S. (1971). The efficiency of essential oils against pathogenic fungi. *Riechst. Aromen, Koerperpflegm.* **21**, 241.

Rao, B. S., Sudborough, J. J., and Watson, H. E. (1925). Notes on some Indian essential oils. *J. Indian Inst. Sci., Sect. A.* **8**, 143.

Rao, P. S. (1948). Tamarind seed jellose (pectin)—its jellying properties. *J. Sci. Ind. Res., Sect. B*, **7**, 89.

Rao, P. S. (1949). Jellies and related products from tamarind seed kernels. *J. Sci. Ind. Res., Sect. A* **8**, 354.

Rao, P. S. (1959). Tamarind. *In* "Industrial Gums" (R. L. Whistler, ed.), p. 461. Academic Press, New York.

Rauscher, H., Mayr, G. D., and Kaemmerer, H. (1957). Ethylene oxide for cold sterilization. *Food Manuf.* **32**, 169.

Rauscher, K., and Korn, O. (1961). Water determination in food products by distillation procedures. *Nahrung* **5**, 656.

Razdan, T. A., and Kaul, G. L. (1973). Qualitative separation of Indian essential oil components. (Oil of caraway) by chromatography. 1. *Indian Perfum.* **17**, 56.

Reddy, H. S. (1965). Some aspects of marketing of cardamom. *Spices Bull. (Semin. No.)* **4**(10), 37.

Redgrove, H. S. (1930). Some notes on ginger. *Pharm. J.* **125**, 54.

Redshaw, E. S., Hougen, F. W., and Baker, R. J. (1971). A distillation technique for isolation of volatile materials for gas chromatographic analysis and its application to coriander seed (*Coriandrum sativum*). *J. Agric. Food Chem.* **19**, 1264.

Reisch, J., Schratz, E., and Qadry, S. M. J. S. (1966). The composition of the essential oil of *Coriandrum sativum* in both the fruit and living plant. III. Oil composition during ontogenesis. *Planta Med.* **14**, 436; *Chem. Abstr.* **66**, 44263e (1967).

Reitsema, R. H. (1954). Characterisation of essential oils. Mentha genus oils. *J. Am. Pharm. Assoc.* **43**, 414.

Reitsema, R. H. (1958). Some new constituents of mint oil. *J. Am. Pharm. Assoc., Sci.* Ed. **47**, 265.

Reitsema, R. H., Cramer, F. J., and Fass, W. E. (1957). Chromatographic measurements of variations in essential oils within a single plant. *J. Agric. Food Chem.* **5**, 779.

Reitsema, R. H., Cramer, H. T., Scully, N. J., and Chorney, W. (1961). Essential oils synthesis in mint. *J. Pharm. Sci.* **50**, 18.

Renis, H. E., and Henze, R. E. (1958). Cystein derivatives in mature onion bulbs. *Food Res.* **23**, 345.

Revankar, C. D., and Sen. D. P. (1974). Antioxidant effect of a spice mixture on sardine oil. *J. Food Sci. Technol.* **11**, 31.

Rhyu, H. Y. (1978). Some comments on the estimation of sensory threshold by Kaerber and Scoville methods. *J. Food Sci.* **43**(5), 1632.

Rhyu, H. Y. (1979a). Gas chromatographic characterization of sages of various geographic origins. *J. Food Sci.* **44**, 758.

Rhyu, H. Y. (1979b). Gas chromatographic characterization of oregano and other selected spices of the Labiate family. *J. Food Sci.* **44**, 1373.

Richard, H. M., Russell, G. F., and Jennings, W. G. (1971). The volatile components of black pepper varieties. *Chromatogr. Sci.* **9**, 560.

Richard, W. K. (1921). Ground cinnamon adulterated with others including a contribution to the ash and sand content of insect powder. *Z. Unters. Nahr.-Genussm. Gebrauchsgegenstaende* **41**, 78.

Richardson, K. C. (1966). Effect of dehydration temperature on the quality of dried ginger. *Food Technol.* **18**, 93.

Richter, G. (1965). Quick quality evaluation of commercial cinnamon oils, flavours, and cinnamon bark drugs by paper chromatography. *Riechst., Aromen, Koerperpflegem.* **15**, 379.

Ripa, A. (1966). Quantitative and qualitative composition of proteins and amino acids in some leguminous plants. *Tautsaimn. Derigie Augi* **3**, 127; cf. *Chem. Abstr.* **62**, 5577g (1966).

Rippetoe and Wise (1912). *J. Am. Pharm. Assoc.* **1**, 1273 (ref. Corrans and Edgar, 1933).

Rivas, S. (1932). Semi-micro method for the determination of cinnamaldehyde in cinnamon bark. *Bol. Farm. Mil.* **10**, 18.

Roaf, A., and Brickey, P. M., Jr. (1968). Method for the extraction of extreneous materials from ground cinnamon. *J. Assoc. Off. Anal. Chem.* **51**, 518.

Roberts, J. S., and Sharts, C. M. (1962). *Org. React.* **12**, 1.

Roberts. L. A. (1968). Gas chromatographic determination of methylene chloride, ethylene dichloride and trichloroethylene residues in spice oleoresins. *J. Assoc. Off. Anal. Chem.* **51**, 825.

Robinson, F. V. (1974). Quality control of spices during harvesting and processing. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 40.

Robinson, R. F., Overbeck, R. C., and Porter, F. E. (1957). X-ray sterilization of spices. *Coffee Tea Ind. Flavor Field* **77**, 61.

Rodriguez, D. W. (1969). "Pimento—A Short Economic History," Commodity Bull. No. 3, p. 16. Minist. Agric. & Fish., Jamaica.

Rodriguez, D. W. (1971). "Ginger—A Short Economic History," Commidity Bull. No. 4. Agric. Planning Unit, Minist. Agric. & Fish., Jamaica.

Rogers, J. A. (1966). Advances in flavour and oleoresin chemistry (a chapter cited in "Flavor Chemistry" by Gold (1966). *Adv. Chem. Ser.* **56**, 203.

Roos, L., and Hughes, E. (1924). Allyl mustard oil in enology. *Ann. Falsif. Fraudes* **413** (cf. Corran and Edgar, 1933).

Rosebrook, D. D. (1971). Collaborative study of a method for extractable colour in paprika and paprika oleoresin. *J. Assoc. Off. Anal. Chem.* **54**, 37.

Rosebrook, D. D., and Barney, J. E., II (1968). Investigation of the determination of volatile isothiocyanates in mustard seeds and flour. *J. Assoc. Off. Anal. Chem.* **51**, 633.

Rosebrook, D. D., Bolze, G. C., and Barney, J. E., II (1968a). Improved method for determination of extractable colour in capsicum spices. *J. Assoc. Off. Anal. Chem.* **51**, 637.

Rosebrook, D. D., Bolze, C. C., and Barney, J. E., II (1968b). An improved method for determining steam volatile oil in cassia. *J. Assoc. Off. Anal. Chem.* **51**, 644.

Rosengarten, F., Jr. (1969). "The Book of Spices." Livingston Publ. Co., Wynnewood, Pennsylvania.

Rosenthaler, L. (1926). Behaviour of Chinese and Ceylon cinnamon toward iodine. *Pharm. Acta Helv.* **1**, 190.

Rothbacher, H. (1965). Cineole and limonene components of Romanian peppermint oil. *Farmacia (Bucharest)* **13**, 475; *Chem. Abstr.* **64**, 3279b (1966).

Rothbacher, H., Crisan, C., and Bedo, E. (1964). Thin-layer chromatography, the separation of the main constituents of Romanian peppermint oil. *Farmacia (Bucharest)* **12**, 733; *Chem. Abstr.* **62**, 10288c (1965).

Rounanizadesh, N. (1969). Preservation of vegetables by irradiation in Iran. *Food Irrad.* **10**, 8.

Rovesti, P. (1971). Ecological influences on composition of essential oils. Chemotexonomic varieties of wild thymes in Italy. *Parfums, Cosmet., Savons Fr.* **1**, 139.

Rowan, P. A. (1942). The spices of The Netherlands East Indies. *Meded. Kolon, Inst. Amst.* **58**, 85.

Roy, B. R. (1959). Detection of artificial fat-soluble coloring substances in turmeric and chilli powders. *J. Sci. Ind. Res., Sect. C* **18**, 18.

Ruber, C. N. (1913). *Ber. Dtsch. Chem. Ges.* **46**, 335 (cited by Labruyere, 1966).

Rusev, D., and Bacvarov, S. (1970). The possibility of prolonged onion seed storage. *Gradinar Lozar. Nauka* **7**, 45.

Russell, G. F., and Else, J. (1973). Volatile compositional differences between cultivars of black pepper (*Piper nigrum*). *J. Assoc. Off. Anal. Chem.* **56**, 344.

Russell, G. F., and Else, J. (1975). Volatile compositional differences between cultivars of black pepper (*Piper nigrum*). *Indian Spices* **12**(1), 4.

Russell, G. F., and Jennings, W. G. (1970). Occurrence of *cis* and *trans* sabinene hydrate in oil of black pepper. Synthesis by oxy-mercuration demercuration procedure. *J. Agric. Food Chem.* **18**, 733.

Russell, G. F., and Olson, K. V. (1972). The volatile constituents of oil of thyme. *J. Food Sci.* **37**, 405.

Russell, G. F., Murray, W. J., Muller, C. J., and Jennings, W. G. (1968). Bergamotenes in oil of black pepper. *J. Agric. Food Chem.* **16**, 1047.

Sabel, W., and Warren, J. D. F. (1972). Theory and practice of oleoresin extraction. *Proc. Int. Conf. Spices, Trop. Prod. Inst., London, 1972* p. 189.

Sacchetta, R. A. (1960). Paper chromatography of red paprika (*Capsicum frutescens*) Powders. *Rev. Asoc. Bioquim. Argent.* **25**, 187.

Sadtler, S. S., Lathrop, E. C., and Mitchel, C. A., eds. (1948). "Allen's Commercial Organic Analysis," 5th ed. McGraw-Hill (Biakiston), New York.

Sagarin, E. (1945). "The Science and Art of Perfumery." McGraw-Hill, New York.

Saghir, A. R., Mann, L. K., Bernhard, R. A., and Jacobsen, J. V. (1964). Determination of aliphatic mono and disulphides in Allium by gas chromatography and their distribution in the common food spices. *Proc. Am. Soc. Hortic. Sci.* **84**, 386.

Saha, J. C., and Kasinathan, S. (1961a). Ecbolic properties of Indian medicinal plants. 1. *Indian J. Med. Res.* **49**, 130.

Saha, J. C., and Kasinathan, S. (1961b). Ecbolic properties of Indian medicinal plants. II. *Indian J. Med. Res.* **49**, 1094.

Sahasrabudhe, M. R., and Bhatia, D. S. (1953). A note on antioxidant properties of spices. *Sci. Cult.* **18**, 384.

Sair, L., and Klee, L. (1967). Debittering of turmeric. U.S. Patent 3,340,250; *Chem. Abstr.* **67**, 107522g (1967).

Sakurai, H., and Umeda, M. (1961). Analysis of opium alkaloids. XlV. Determination of main alkaloids in opium. *Yakugaku Zasshi* **81**, 1217; *Chem. Abstr.* **56**, 4869c (1962).

Saleh, M. R. I., and Mohamed, V. A. (1965). Phytochemical investigation of *Rosemarinum officinalis* grown in the United Arab Republic. 1. Examination of the volatile oil. *J. Pharm. Sci.* U.A.R. **6**, 137; *Chem. Abstr.* **67**, 84753s (1967).

Salem, S. A. (1974). Effect of gamma radiation on the storage of onions used in dehydration industry. *J. Sci. Food Agric.* **25**, 257.

Salzer, U. J. (1975). Fatty acid composition of the lipids of some spices. *Fette, Seifen, Anstrichm.* **77**, 446.

Sammy, G. M., and Nawar, W. W. (1968). Identification of the major components of nutmeg oil by gas chromatography and mass spectrometry. *Chem. Ind. (London)* p. 1279.

Sanchez-Palomera, E. (1951). The action of spices on the acid gastric secretion on the appetite and on caloric intake. *Gastroenterology* **18**, 269.

Sancho, J. (1962). Method for the determination of colorimetric purity of paprika extracts. *Coffee Tea Ind. Flavor Field* **86**(8), 27.

Sancho, J., and Navarro, F. (1962a). Physico-chemical studies of species of capsicum. I. Absorption spectra of the carotenoids in the ultraviolet. *An. R. Soc. Esp. Fis. Quim., Ser. B* **58**, 565.

Sancho, J., and Navarro, F. (1962b). Colour in capsicum species. 1. Variation of colour in paprika and its extracts. *An. Bromatol.* **14**, 25.

Sandermann, W. (1942). Caraway and its volatile constituents. *Chem. Zentralbl.* **1**, 2072.

Sane, R. H., Powers, J. J., Morse, R. E., and Mills, W. C. (1950). The pH and total acidity of raw and canned pimientos. *Food Technol.* **4**, 279.

Sanford, K. J., and Heinz, D. E. (1971). Effects of storage on volatile composition of nutmeg. *Phytochemistry* **10**, 1245.

Sangameswaran, S. V., Asha, C. B., and Geetha, R. (1974). "A Select Bibliography on Spices 1966–72," CFTRI Bibliogr. Ser. No. 11. CFTRI, Mysore, India.

Santa Maria, J., and Ruiz-de-Assin, O. (1961). Methods of analysis of Spanish red pepper—their application and the standardization of the commercial products and the control of fabrication. *Bol. Inst. Nac. Invest. Agron. (Spain)* [N.S.] **21**, 169.

Saraf, A., and Niga, S. A. R. (1964). Estrogenic activity of pomegranate seed oil. *J. Endocrinol.* **29**, 91.

Sastry, L. V. L. (1957). Estimation of organic sulphur in asafoetida. *J. Sci. Ind. Res., Sect. B* **16**, 508.

Sastry. L. V. L., Srinivasan, M., and Subrahmanyan, V. (1955a). Saffron (*Crocus sativus*). *J. Sci. Ind. Res., Sect. A* **14**, 178.

Sastry, L. V. L., Srinivasan, M., and Subrahmanyan, V. (1955b). Asafoetida. Part II. Assessment of quality. *J. Sci. Ind. Res., Sect. A* **14**, 585.

Satoh, Z. (1952). Effect of Allium diets on bacterial synthesis of thamine in the intestinal tract. 1. Test on dietary intake of Allium plants. *Vitamins* **5**, 184 (cited by Krishnamurthy and Sreenivasamurthy, 1956).

Savur, G. R. (1955). Utilization of tamarind seed polyose in food industries. *Indian Food Packer* **9**(3), 31.

Sawicki, E., Hauser, T. R., and McPherson, S. (1962). Spectrophotometric determination of formaldehyde and formaldehyde-releasing compounds with chromatropic acid, 6-amino-1-naphthol-3-sulfonic acid (Jacid) and 6-amino-1-naphthol-3-sulfonic acid (Phenyl J. acid). *Anal. Chem.* **34**, 1460.

Saxena, S. N., Baser, B. L., and Durge, A. S. (1964). Turmeric procedure for routine analysis of boron in soil and water. *J. Indian Soc. Soil Sci.* **12**, 325.

Scarpa, A. (1950). Experiments on the anthelmintic power of helba fenugreek. *Fitoterapia* **21**, 143.

Schenk, G. (1954). Experiments on the standardization of capsicum fruit and tincture of capsicum. *Dtsch. Apoth.-Ztg.* **94**, 970; *Anal. Abstr.* **2**, 453 (1955).

Schenk, G. (1957). Methods of evaluation of *Fractus capsaici* (Capsicum fruit). *Farmacognosia* **17**, 3.

Schenk, G., Froemming, K. H., and Kluge, H. J. (1962). The enzymic and nonenzymic oxidation of therapeutically active plant substances. XXIV. Morphine losses in poppy capsules (*Papaver somniferum*). *Pharm. Z. V. Apoth.-Ztg.* **107**, 1777; cf. *Chem. Abstr.* **61**, 5449f (1964).

Scher, A., and Ivanov, S. A. (1976). Natural anti-oxidants in fennel oil. *Fette Seifen* **78**, 224.

Schillinger, A., and Zimmermann, G. (1965). Stability of various vitamins in dried potatoes and vegetables. 1. Vitamins in stored dried potatoes and vegetables. *Dtsch. Lebensm.-Rundsch.* **61**, 45.

Schipper, P. A. (1968a). A preliminary experiment on weight loss in onions. *N.Z. Comm. Gr.* **23**(10), 23.

Schipper, P. A. (1968b). Shelf-life of vegetables. *N. Z. Comm. Gr.* **23**(10), 31.

Schmal, F. H. (1940). Ceylon artificial cinnamon. *Vorratspflegelebensmittelforsch.* **3**, 58.

Schneider, A. (1921). "Micro Analysis of Powdered Vegetable Drugs." McGraw-Hill (Blakiston), New York.

Schneider, M. A., DeLuca, V., Jr., and Gray, S. J. (1956). The effect of spice ingestion upon the stomach. *Am. J. Gastroenterol.* **26**, 722.

Schnekenburger, J. (1964). New determination of morphine in opium. *Planta Med.* **12**, 321.

Schonberg, F. (1952). The significance of bacteria containing spices in the spoilage of sausage products and canned meats and reduction in no. of bacteria in spices of means by ultraviolet sterilization lamps. *Fleischwirtschaff.* **4**, 132; *Brt. Food Sci. Abstr.* **25**, 2357 (1953).

Schonberg, F. (1955). The importance of natural spices free from bacteria or with a low bacterial content in the production of unobjectionable raw sausage. *Fleischwirtschaft* **7**, 243.

Schramm, R. (1961). Paper chromatography of organic acids in storage roots of some umbelliferae. *Acta Soc. Bot. Pol.* **30**, 285.

Schratz, E., and Qadry, S. M. J. S. (1966). The composition of the essential oil of *Coriandrum sativum* in both the fruit and living plant. II. Identification and synthesis of tridecen-2-al-1. *Planta Med.* **14**, 326; cf. *Chem. Abstr.* **65**, 13530h (1966).

Schratz, E., and Qedan, S. (1965). Composition of the essential oil of *Thymus serpyllum* complex, 1. Detection of the oil components by thin-layer-chromatography. *Pharmazie* **20**, 710; *Chem. Abstr.* **64**, 6398g (1966).

Schroder, H. (1959). The influence of nitrogen, phosphoric acid and potassium on the yield and essential oil content of marjoram (*Majorana hortensis* Moench). *Pharmazie* **14**, 329.

Schroder, U. (1969). Investigations on the daily rhythm of essential oils. 2nd Report. Daily variations in the content of essential oil in *Majorana hortensis*. *Pharmazie* **24**, 421.

Schulte, K. E., and Krueger, H. M. (1955). Colorimetric determination of capsaicin in drugs. *Z. Anal. Chem.* **147**, 266.

Schulte, K. E., and Krueger, H. M. (1957). The determination of capsaicin in drugs and similar medicinal preparations. *Arch. Pharm. (Belrin)* **27**, 202.

Schultz, J. E. R., and Hansen, H. (1971). Investigations on the variations in content of volatile oils in Danish grown caraway. *Tidsskr. Planteav* **75**, 377.

Schultz, O. E., and Mohramann, H. L. (1965a). Analysis of constituents of garlic (*Allium sativum*). I. Thin-layer chromatography of garlic oil. *Pharmazie* **20**, 379.

Schultz, O. E., and Mohrmann, H. L. (1965b). Analysis of constitutents of garlic, *Allium* sativum. II. Gas chromatography of garlic oil. *Pharmazie* **20**, 441; cf. *Chem. Abstr.* **63**, 8116f (1965).

Schuphan, W. (1937). Die bestimmug Schwefelhaltiger atherischer ole in genusen. *Forschungsdienst* **4**, 127.

Schut, W. (1925). Determination of essential oils in spices. *Chem. Weekbl.* **22**, 344.

Schwartzman, G. (1955). A method for the recovery of extraneous material in ground cinnamon. *J. Assoc. Off. Agric. Chem.* **38**, 781.

Schwien, W. G., and Miller, B. J. (1967). Detection and identification of dehydrated red beets in capsicum spices. *J. Assoc. Off. Anal. Chem.* **50**, 523.

Schwimmer, S. (1967). Development of bitter substance in onion juice. *Food Technol.* **221**, 292.

Schwimmer, S., and Guadagni, D. G. (1962). Relation between olfactory threshold concentration and pyruvic acid content of onion juice. *J. Food Sci.* **27**, 94.

Schwimmer, S., and Guadagni, D. G. (1967). Kinetics of the enzymatic development of pyruvic acid and odor in frozen onions treated with cysteine C-S lyase. *Food Sci.* **33**, 193.

Schwimmer, S., and Mazelis, M. (1963). Characterisation of allinase of *Allium cepa* (onion). *Arch. Biochem. Biophys.* **100**, 66.

Schwimmer, S., and Weston, W. J. (1961). Enzyme development of pyruvic acid in onion as a measure of pungency. *J. Agric. Food Chem.* **9**, 301.

Schwimmer, S., Carson, J. F., Makower, R. U., Mazelis, M., and Wong, F. F. (1960). Demonstration of allinase in proteinase preparation from onion. *Experientia* **16**, 449.

Schwimmer, S., Venstrom, D. W., and Guadigni, D. G. (1964). Relation between pyruvate content and odor strength of reconstituted onion powder. *Food Technol.* **18**, 121.

Scott, P. M., and Kennedy, B. P. (1973). Analysis and survey of ground black, white and capsicum peppers for aflatoxins. *J. Assoc. Off. Anal. Chem.* **56**, 1452.

Scoville, W. L. (1912). Note on capsicums. *J. Am. Pharm. Assoc., Pharm.* Ed. **1**, 453.

Seher, A. (1962). Chromatographic analysis procedures in research and production control. *J. Soc. Cosmet. Chem.* **13**, 385.

Seideman, J. (1962a). Starch and *Piper longum* in ground pepper. *Q. J. Crude Drug Res.* **2**, 245.

Seideman, J. (1962b). Microscopic studies of the bark of cassia. *Z. Lebensm.-Unters.-Forsch.* **116**, 24; *Chem. Abstr.* **56**, 10651c (1962).

Semmler, F. W. (1892). Die athersche ol der kuchenzweibel. (*Allium cepa* L.) *Arch. Pharm. (Weinheim, Ger.)* **230**, 443.

Sen, A. R., and Roy, B. R. (1975). Cinnamon and Cassia—their standards and chemical characterisation. *ISI Bull.* **27**, 200.

Sen, A. R., Sardar, P. K., Sil S., and Mathew, T. V. (1973a). Importance of volatile oil and cold water extract estimation in analysis of cumin (*jira*). *J. Food Sci. Technol.* **10**, 187.

Sen, A. R., Sengupta, P., Ghosedastidar, N., and Mathew, T. V. (1973b). Chromatographic detection of small amounts of mineral oil in chillies. *Res. Ind.* **18**, 97.

Sen, A. R., Sengupta, P., Sarkar, P. K., and Roy, B. R. (1977). Thin-layer chromatographic detection of *Amomum subulatum* in *Elletaria cardamomum*. *J. Assoc. Off. Anal. Chem.* **60**, 307.

Sen, S. K., and Rao, C. V. N. (1966). Pectic substances in onion (*Allium cepa*). *Indian J. Appl. Chem.* **29**, 127.

Sen Gupta, P., Sen, A. R., Bose, A., and Mathew, T. V. (1973). Detection of coumarin in vanilla flavoured food by TLC. *Res. Ind.* **18**, 96.

Seniko, I. V., and Reznikov, V. M. (1967). Micromethod for the simultaneous determination of vanillin and vanillic acid in alkaline mitrobenzene oxidation products. *Zh. Prikl. Khim.* **40**, 1879.

Serna, A. (1962). Colour in capsicum species. II. Method of colour determination. *An. Bromatol.* **14**, 35.

Sethi, S., Nigam, M. C., and Rao, P. R. (1965). Studies on chemical and economic utilization of *Anethum sowa*. *Indian Perfum.* **9**, Part A, 17.

Sethi, S. C., and Aggarwal, J. S. (1950). Stabilization of edible fats by spices and condiments or spices. *Nature (London)* **166**, 518.

Sethi, S. C., and Aggarwal, J. S. (1952). Stabilization of edible fats by spices and condiments. *J. Sci. Ind. Res., Sect. B* **11**, 468.

Sethi, S. C., and Aggarwal, J. S. (1956). Stabilization of edible fats by spices. *J. Sci. Ind. Res., Sect. B* **15**, 34.

Sethi, S. C., and Aggarwal, J. S. (1957). Stabilization of edible fats by spices. *J. Sci. Ind. Res., Sect. A* **16**, 181.

Sethi, V., Anand, J. C., Netrapal, and Bhagchandani, P. M. (1973). Quality screening of white onion cultivars for use in dehydration. *Indian Food Packer* **22**(5), 5.

Shalaby, G. I. (1966). Genetic and environmental factors affecting internal doubling in onions, *Allium cepa* L. *Diss. Abstr. B* **27**, 1040.

Shankaracharya, N. B., and Natarajan, C. P. (1971a). Leafy spices—chemical composition and uses. *Indian Food Packer* **25**(2), 29.

Shankaracharya, N. B., and Natarajan, C. P. (1971b). Cardamom—chemistry, technology and uses. *Indian Food Packer* **25**(5), 28.

Shankaracharya, N. B., and Natarajan, C. P. (1971c). Coriander—chemistry, technology and uses. *Indian Spices* **8**(2), 4.

Shankaracharya, N. B., and Natarajan, C. P. (1971d). Chemical composition and uses of cumin. *Indian Food Packer* **25**(6), 22.

Shankaracharya, N. B., and Natarajan, C. P. (1972). Fenugreek—chemical composition and uses. *Indian Spices* **9**(1).

Shankaracharya, N. B., and Natarajan, C. P. (1973a). Turmeric—chemistry, technology and uses. *Indian Spices* **10**(3), 8.

Shankaracharya, N. B., and Natarajan, C. P. (1973b). Vanilla—chemistry, technology and uses. *Indian Food Packer* **26**(3), 29.

Shankaracharya, N. B., and Natarajan, C. P. (1973c). Problems in post-harvest technology of spices. *Indian Food Packer* **26**(6), 14.

Shankaracharya, N. B., and Natarajan, C. P. (1977). Role of spices in health. *Arogya (J. Health Sci.)* **3**, 99.

Shankaracharya, N. B., Anandaraman, and Natarajan, C. P. (1973). Chemical composition of raw and rosted fenugreek seeds. *J. Food Sci. Technol.* **10**, 179.

Shankaranarayana, M. L., Nagalakshmi, S., and Natarajan, C. P. (1970). Colorimetric method of determination of pungent constituents of pepper. *Flavour Ind.* **1**, 173.

Shankaranarayana, M. L., Abraham, K. O., Raghavan, B., and Natarajan, C. P. (1974a). Analytical methodology in pesticide research. *Indian Food Packer* **28**(1), 65.

Shankaranarayana, M. L., Raghavan, B., Abraham, K. O., and Natarajan, C. P. (1974b). Volatile sulfur compounds in food flavors. *Crit. Rev. Food Technol.* **4**, 395.

Shannon, S., Yamaguchi, M., and Howard, F. D. (1967a). Reactions involved in formation of a pink pigment in onion purees. *J. Agric. Food Chem.* **15**, 417.

Shannon, S., Yamaguchi, M., and Howard, F. D. (1967b). Precursors involved in the formation of pink pigment in onion purees. *J. Agric. Food Chem.* **15**, 423.

Sharaf, A., and Goma, N. (1965). Phytoestrogens and their antagonisms to progesterone and testosterone. *J. Endocrinol.* **31**, 289; cf. *Chem. Abstr.* **63**, 9208b (1963).

Sharma, A., Tewari, G. M., Shrikhande, A. J., Padwal-Desai, S. R., and Bandyopadhayay, C. (1979). Inhibition of aflatoxin producing fungi by onion extracts. *J. Food Sci.* **44**, 1545.

Sharma, J. N., and Seshadri, T. R. (1955). Survey of anthocayanins from Indian sources. Part II. *J.*

Sharp, L. K. (1951). The assay of vanillin. *Analyst (London)* **76**, 215.

Sharpnell, G. S. (1967). Technological development of ginger industry in Australia. *Food Technol.* **19**, 604.

Shellard, E. J. (1967). Oil of coriander—comparison of oils of coriander according to Polish Pharmacopoelia III (1954) and British Pharmacopoeia 1963. *Acta Pol. Pharm.* **24**, 183.

Sheneman, J. M. (1973). Survey of aerobic thermophilic bacteria in dehydrated onion products. *J. Food Sci.* **38**, 203.

Sherwood, P. W. (1961). Vanillin. *Perfum. Essent. Oil Rec.* **52**, 577.

Shifrina, Kh. B. (1961). The biochemistry of onion. *Biokhim. Ovoshchn. Kul't.* p. 328.

Shimazu, F., and Sterling, C. (1967). Changes in crystallinity of cellulose during dehydration. *J. Food Sci.* **32**, 273.

Shivrina, A. N. (1962). Biochemistry of garlic. *Biokhim. Ovoshchn. Kult.* p. 378; review from *Zh. Khim. Biol. Khim. Abstr.* **135**, 667 (1962).

Shulgin, A. T. (1963). Composition of the myristicin fraction from oil of nutmeg. *Nature (London)* **197**, 379.

Shulgin, A. T., and Kerlinger, H. O. (1964). Isolation of methoxyeugenol and transisoelemicin from oil of nutmeg. *Naturwissenschafften* **51**, 360.

Shuster, H. V., and Lockhart, E. E. (1954). Development and application of objective methods for quality evaluation of spices. I. Color grading of capsicums. *Food Res.* **19**, 472.

Sica, S. J., and Nanz, R. A. (1950). Identification of insect fragments in spices and other food materials utilizing a differential stain. *J. Assoc. Off. Agric. Chem.* **33**, 895.

Siddappa, G. S., and Bhatia, B. S. (1955). Canning of chilies. *Bull. Cent. Food Technol. Res. Inst., Mysore* **4**, 9.

Sidhu, G. S., and Bhramaramba, A. (1963). Chromatographic studies on Indian cinnamon leaf oil. *Perfum. Essent. Oil Rec.* **54**, 732.

Sigmund, S., and Guadagni, D. G. (1968). Kinetics of the enzymic development of pyruvic acid and odor in frozen onions treated with cysteine C-S lyase. *J. Food Sci.* **33**, 193.

Silliker, J. H. (1963). Total count as indexes of food quality. *In* "Microbiological Quality of Foods." (L. W. Slanetz, A. R. Gaufin, and Z. J. Ordal, eds.), p. 102. Academic Press, New York.

Sinah, G. S., and Baslas, K. K. (1968). Studies of essential oils from odoriferous plants of Kumaon. Essential oil from *Artemisia vulgaris* Linn. *Perfum. Essent. Oil Rec.* **59**, 866.

Sinah, G. S., and Baslas, K. K. (1971). Essential oil from leaves of *Artemisia vestita* Wall. *Flavour Ind.* **2**, 539.

Sindall, H. E. (1920). Report on spices. *J. Assoc. Off. Agric. Chem.* **3**, 427.

Singh, H. S., Bhagat, S. D., Mathur, R. K., Gopinath, K. W., and Ganguly, D. (1971). Cultivation of basil (*Ocimum basilicum* L.) at Jorhat, Assam and the chemical composition of its oil. *Flavour Ind.* **2**, 481.

Singh, K., and Kumar, S. (1969). Morphological and biochemical changes in stored onions as influenced by nitrogen and phosphorus fertilization. *Plant Sci.* **1**, 181.

Singh, K. P., and Gupta, G. N. (1964). A critical study of the methods used for the estimation of carvone and dihydrocarvone in the essential oil of dill. *Indian Perfum.* **8**, Part 1, 25.

Singh, L. J., Pruthi, J. S., Sreenivasamurthy, V., and Lal, G. (1959a). Effect of regional variability on the quality of garlic powder. *Food Sci.* **8**, 431.

Singh, L. J., Pruthi, J. S., Sreenivasamurthy, V., Swaminathan, M., and Subrahmanyan, V. (1959b). Effect of type of packaging and storage temperature on allyl sulphide, total sulphur, antibacterial activity and volatile reducing substances in garlic powder. *Food Sci.* **8**, 453.

Singh, L. J., Pruthi, J. S., Sankaran, A. N., Indirama, K., and Lal, G. (1959c). Effect of type of packaging and storage temperature on flavour and colour of garlic powder. *Food Sci.* **8**, 457.

Singh, R. S. (1969). Content and characteristics of essential oil of *Anethum graveolens*. L. raised on alkali soil. *Plant Sci.* **1**, 173.

Singh, R. S., and Ojah, T. P. (1974). Equilibrium moisture content of groundnut and chilies. *J. Sci. Food Agric.* **25**, 451.

Singh, R. S., and Singh, C. P. (1970). Studies on some agronomic characters in the growth, yield and essential oil content of dill. *Indian Oil Soap J.* **36**, 55.

Singh, S., and Bhatia, I. S. (1972). Carbohydrates in the bulbs of *Allium cepa* Linn. I. Detection, isolation and analysis of the first members of the series. *J. Res. Ludhiana* **9**, 327.

Sinnadurai, S., and Amuti, S. K. (1971). Dormancy of shallots in Ghana. *Exp. Agric.* **7**, 17.

Sinsheimer, J. E. (1959). An intensity response method for the measurement of flavour intensity. *Food Res.* **24**, 445.

Siregar, J. A., Steinberg, M. P., Wei, L. S., and Nelson, A. I. (1967). End of storage conditions on standard plate counts of raw onions. *J. Food Sci.* **32**, 349.

Sirsi, M. (1962). The pharmacology of zerumbone, a sesquiterpene ketone isolated from *Zingiber zerumbet*. *Indian J. Pharm.* **24**, 83.

Skovronskii, V. A. (1952). The effect of caraway, anise and sweet fennel on urine elimination. *Sb. Nauchn. Tr., L'vov. Gos. Vet.-Zootekh. Inst.* **6**, 275.

Smith, D. M. (1965). Comparison of methods for the determination of vanillin in vanilla extract. *J. Assoc. Off. Agric. Chem.* **48**, 509.

Smith, D. M., and Levi, L. (1961). Treatment of compositional data for the characterisation of essential oils. Determination of geographical origin of peppermint oils by gas chromatograph analysis. *J. Agric. Food Chem.* **9**, 230.

Smith, D. M., Skakum, W., and Levi, L. (1963). Determination of botanical and geographical origin of spearmint oils by gas chromatographic and ultraviolet analysis. *J. Agric. Food Chem.* **11**, 268.

Smith, E. R., Samuel, A., and Mitchell, L. C. (1926). Detection of added peppershells in pepper. *J. Assoc. Off. Agric. Chem.* **9**, 233; *Chem. Abstr.* **20**, 31985 (1926).

Smith, H. W. (1940). Treated spices reduce spoilage. *Food Ind.* **12**(11), 50.

Sokolinikov, N. P. (1958). "Tekhnologiya Efiromaslitchnogo Proizvodstva," p. 25. God. Nauch-Tekh. Isda. Mini. Prom. Prodovst. Tovarov, Moscow.

Soldatenkov, T. A., Mazurava, T. A., and Panteleev, A. N. (1960). Organic acids in onion and spinach. *Tr. Petergof. Biol. Inst., Leningr. Gos. Univ.* **18**, 55; cf. *Chem. Abstr.* **55**, 10739b (1961).

Song, C. S., Kimys, L. D. J., and Nam, C. C. (1963). A blood anticoagulant substance from garlic. II. Chemical analysis and studies on the biochemical and pharmocological effects. *Yonsei Med. J.* **4**, 21.

Sonnenblick, B. P., Hollander, F., and Sober, H. A. (1950). Variations in cell types and cell density of gastric secretions following eugenol stimulation. *Gastroenterology* **16**, 211; cf. *Chem. Abstr.* **44**, 10935h (1950).

Spanyar, P., and Blazovich, M. (1969). A thin-layer chromatographic method for the determination of capsaicin in paprika. *Analyst (London)* **94**, 1084.

Spanyar, P., Kevel, J., and Kevel, J. (1956a). Determination of capsaicin content. *Elelmez. Ipar* **10**, 52.

Spanyar, P., Kevel, J., and Kiszel, J. (1956b). Determination of capsaicin by polarometric titration. *Elelmiszervizsgalati Kozl.* **11**, 257.

Spanyar, P., Kevel, E., and Kiszel, M. (1957). Determination of capsaicin content. *Sep. Acta Chim. Acad. Sci. Hung.* **11**, 137.

Spanyar, P., Levei, E., and Kiszel, M. (1958). Quick method for the determination of capsaicin in raw paprika. *Konzerv-, HUS- Hutoipari Kutato Intezet, Kozl.* **I-II**, 34.

Spanyar, P., Kevel, J., and Kiszel, J. (1959). A fuszrpaprika Kapsazaicint art almanak meghatarozasara szolgalo visagalati modszerek osszehasonlitasa. *Konzerv.- paprikaip.* **7**, 311.

Spare, C. G., and Virtanen, A. L. (1963). On the lachrymatory factor in onion (*Allium cepa*). Vapours and its precursor. *Acta Chem. Scand.* **17**, 641.

Spaziano, V. T., Shah, H. C., Chou, T. C., Price, C. C., and Lin, H. H. (1968). The synthesis of some 9 p- (bis-2-cholorsethylamino) phemyl - 6 - substituted purines as potential cytotoxic agents. *J. Chem. Soc.* p. 915.

Spices Export Promotion Council, Cochin, India (1968a). Chilies in competing countries. *Spices Bull (Chilies Semin. No.)* **6**, 62.

Spices Export Promotion Council, Cochin, India (1968b). Chilies markets abroad. *Spices Bull. (Chilies Semin. No.)* **6**(3), 65.

Spices Export Promotion Council, Cochin, India (1979). Export of Spices from India. *Indian Spices* **16**, 9.

Spinks, M. (1969). Islands of cloves. *Perfum. Essent. Oil. Rec.* **60**, 322.

Sreenathan, V. R., Narasimhan, K. S., Majumder, S. K., and Iyengar, N. V. R. (1962). Some aspects of packaging of spices. *Proc. Symp. Spices—Role Natl. Econ., Ist, 1962* Abstract, p. 18.

Sreenivasamurthy, V., and Krishnamurthy, K. (1959). Place of spices and aromatics in Indian dietry. *Food Sci.* **8**, 284.

Sreenivasamurthy, V., and Sastry, L. V. L. (1958). Some observations on the effect of asafoetida oil on intestinal microflora. *Proc. Symp. Antibiot., 1958* p. 237.

Sreenivasamurthy, V., Sreekantiah, K. R., and Johar, D. S. (1960). Effect of garlic on the growth of yeasts and moulds. *J. Sci. Ind. Res., Sect. C* **19**, 61.

Sreenivasamurthy, V., Sreekantiah, K. R., and Johar, D. S. (1961). Studies on the stability of allicin and alliin present in garlic. *J. Sci. Ind. Res., Sect. C* **20**, 292.

Sreenivasamurthy, V., Sreekantiah, K. R., and Johar, D. S. (1962a). Therapeutic usefulness of garlic. Its use in the treatment of rheumatoid arthritis. *Natl. Med. J.* Jan.-Sept. Issue, p. 8.

Sreenivasamurthy, V., Sreekantiah, K. R., Jayaraj, A. P., and Johar, D. S. (1962b). A preliminary report on the treatment of acute *Lepromatous neurites* with garlic. *Leprosy in India* **34**, 171.

Sreenivasa Rao, Y. V., and Sreenivasaya, M. (1932). 111. Studies in the proteins of Indian foodstuffs. Part 1V. The proteins of fenugreek. *J. Indian Inst. Sci. Sect. A* **15**, 122.

Sreenivasa Rao, Y. V., Sastri, B. N., and Narayanan, M. (1933). Studies on the proteins of Indian foodstuffs. Part V. The alcohol-soluble protein of fenugreek. *J. Indian Inst. Sci. Sect. A* **16**, 85.

Srinivasan, K. R. (1953). Chromatographic study of the curcumnoids in *Curcuma longa*. *J. Pharm. Pharmacol.* **5**, 408.

Srinivasan, M. (1972). Effect of curcumin on blood sugar as seen in a diabetic subject. *Indian J. Med. Sci.* **26**, 269.

Srinivasan, M., and Bhatia, I. S. (1954). Glucofructosan from *Polianthus tuberosa* Linn. and garlic (*Allium sativum* Linn.), *Curr. Sci.* **23**, 192.

Srinivasan, M., Bhatia, I. S., and Satyanarayan, M. N. (1953). Carbohydrates of garlic (*Allium sativum* L.) and onion (*A. cepa* L). *Curr. Sci.* **22**, 208.

Srinivasan, S., Ramchandrarao, T. N., and Sastry, L. V. L. (1959). Some indegeneous spices and taste adjuncts. *Food Sci.* **8**, 289.

Sriraman, K. (1970). Marketing of chillies in Tamil Nadu. *J. Agric. Market.* **12**(4), 6.

Srivas, S. R., Pruthi, J. S., and Siddappa, G. S. (1962). Determination of the optimal harvest time for ginger. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962,* Abstract, p. 4.

Srivas, S. R., Pruthi, J. S., and Siddappa, G. S. (1963a). Nitrogenous substances in ginger (Zingiber officinale, Roscoe). *Spices Bull.* **2**(12), 61.

Srivas, S. R., Patwardhan, S. R., and Siddappa, G. S. (1963b). Chemistry and technology of some major spices of India—pepper, cardamom, ginger and turmeric—a review. *Spices Bull. (Annu. No.)* **2**, 77.

Srivastava, H. C., and Mathur, P. B. (1956). Studies on the smoke-curing of garlic. *Bull. Cent. Food Technol. Res. Inst., Mysore* **5**, 286.

Srivastava, H. C., Narasimhan, P., Moorthy, N. V. N., and Dalal, V. B. (1962). Some aspects of storage and transportation of spices. *Proc. Symp. Spices—Role Natl. Econ. 1st, 1962* Abstract, p. 6.

Stahl, E. (1962). "Thin-Layer Chromatography. A Laboratory Handbook". Springer-Verlag, Berlin and New York.

Stahl, E., and Jork, H. (1964). Chemical breeds of medicinal plants. I. Cultivation varieties of European parsley. *Arch. Pharm. (Weinheim, Ger.)* **297**, 273; *Chem. Abstr.* **61**, 2901c (1964).

Stahl, E., Bollinger, H. B., and Lehnert, L. (1963). *In* "Carotine and Carotenoids" (K. Lang, ed.), p. 129. Steinkopff, Darmstadt.

Stahl, W. H. (1965). Critical review of methods for analysis of oleoresins. *J. Assoc. Off. Anal. Chem.* **48**, 515.

Stahl, W. H. (1972). Oleoresin quality analysis: Facts or fancy? *Trop. Sci.* **14**, 335.

Stahl, W. H., Voelker, W. A., and Sullivan, J. H. (1960). Analysis of vanilla extract. I. Organic acid determination. II. Preparation and reproduction of two dimential fluoresceuce chromatograms. *J. Assoc. Anal. Chem.* **43**, 601 and 606.

Stahl, W. H., Sullivan, J. H., and Voelkar, W. A. (1961). Analysis of vanilla extracts. III. Use of paper chromatography in the analysis of pure and adulterated vanilla extracts. *J. Assoc. Off. Agric. Chem.* **44**, 549.

Stahl, W. H., Voelker, W. A., and Sullivan, J. H. (1962). Analysis of vanilla extracts. IV. Amino acid determination. *J. Assoc. Off. Agric. Chem.* **45**, 108.

Stahl, W. H., Skarzynski, J. N., and Voelker, W. A. (1967). Determination of geographical origin of spices I. Cassia and cinnamon by T.L.C. *J. Assoc. Off. Anal. Chem.* **50**, 852.

Stahl, W. H., Skarzynski, J. N., and Voelker, W. A. (1969a). Differentiation of certain types of cassias and cinnamon by measurement of mucilaginous character. *J. Assoc. Off. Anal. Chem.* **52**, 741.

Stahl, W. H., Skarzynski, J. N., and Voelker, W. A. (1969b). Differentiation of geographic origin of spices. II. Oregano by gas chromatography and thin-layer chromatography. *J. Assoc. Off. Anal. Chem.* **52**, 1184.

Stall, S., and Prat, Y. (1960). Degradation of vanillin, ethyl vanillin and p-hydroxybenzaldehyde by autooxidation. *Ann. Falsif. Expert. Chim.* **53**, 316.

Staniforth, V. (1972). Spices or oleoresins. A choice. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 193.

Stanley, R. (1929a). Some notes on cinnamon. *Pharm. J.* **123**, 168.
Stanley, R. (1929b). Some notes on cloves. *Pharm. J.* **123**, 368.
Stanley, W. L. (1963). Recent developments in coumarin chemistry. *In* "Aspects Plant Phenolic Chemistry," 3rd ed., p. 79.
Starke, H., and Herrmann, K. (1976a). Flavonols and flavones of vegetables. VI. On the behaviour of the flavonols in the onion. *Z. Lebensm.-Unters. -Forsch.* **161**, 25.
Starke, H., and Herrmann, K. (1976b). Flavonols and flavones of vegetables. VII. Flavonols of leek, chive and garlic. *Z. Lebensm.-Unters. -Forsch.* **161**, 137.
Starr, H., Kilgemagi, U., and Terriere, L. C. (1963). Insecticide residues in peppermint and their distillation with peppermint oil. *J. Agric. Food Chem.* **11**, 482.
State Marketing Officer, Madras (1968). Present system of marketing, grading and quality control for export of chilies. *Indian Spices* **6**, 35.
Staudinger, I. H., and Muller, F. (1923). Relation between chemical constitution and pepper taste. II. Aliphatic aromatic acid piperides. *Ber. Dtsch. Chem. Ges. B* **56**, 711.
Steindelowna, H. (1957). Quantitative estimation of linalool on coriander oil. *Biul. Inst. Rosl. Lecz.* **3**, 139.
Stelzer, H. (1963). Identification of synthetic coloring in paprika. *Nutr., Bromatol., Toxicol.* **2**, 177.
Steponaviciene, V. (1964). Vitamin C content in herbs cultivated in Lithuanian SSR. *Liet. TSR Aukst. Mokyku Mokslo Darb., Med.* **6**, 215.
Sterrett, F. S. (1962a). The nature of essential oils. 1. Production of essential oil. *J. Chem. Educ.* **39**, 203.
Sterrett, F. S. (1962b). Essential oils. II. Chemical constituents, analysis. *J. Chem. Educ.* **39**, 246; cf. *Chem. Abstr.* **56**, 14411g (1962).
Stobart, T. (1970). "Herbs, Spices and Flavouring." International Wire & Food Publishing Co., London.
Stoll, A., and Seebeck, E. (1947). Alliin, the pure mother substance of garlic oil. *Experientia* **3**, 114.
Stoll, A., and Seebeck, E. (1948). Allium compounds. 1. Alliin, the true mother compound of garlic oil. *Helv. Chim. Acta* **31**, 189; *Chem. Abstr.* **42**, 4136 (1948).
Stoll, A., and Seebeck, E. (1949a). Uber den enzymatischem abbau des Alliins und die Eigenschaften der Allinase. *Helv. Chim. Acta* **31**, 189.
Stoll, A., and Seebeck, E. (1949b). Allium compounds. II. Enzymatic degradation of alliin and the properties of allinase. *Helv. Chim. Acta* **32**, 197; *Chem. Abstr.* **43**, 3482d (1949).
Stoll, A., and Seebeck, E. (1949c). Allium compounds. III. Specificity of allinase and synthesis of compounds related to alliin. *Helv. Chim. Acta* **32**, 866; cf. *Chem. Abstr.* **43**, 3482d (1949).
Stoll, A., and Seebeck, E. (1951). Chemical investigation on alliin, the specific principles of garlic. *Adv. Enzymol.* **11**, 377.
Stoll, S., and Barnier, G. (1964). Study and characterisation of hydroxymethyl furfural in vanilla sugars and identification of the components of vanillin and ethyl vanillin by paper and thin-layer chromatography. *Ann. Falsif. Expert. Chim.* **57**, 131.
Stone, H., Pangborn, R. M., and Ough, C. S. (1965). Techniques for sensory evaluation of food odors. *Adv. Food Res.* **14**, 1.
Strauss, D. (1967). Investigation of nutmegs and nutmeg powder. *Dtsch. Lebensm.-Rundsch.* **63**, 239.
Strong, D. H., Canada, J. C., and Griffiths, B. B. (1963). Incidence of *Clostridium perfringens* in American foods. *J. Appl. Microbiol.* **11**, 42.
Su, H. C. F. (1977). Insecticidal properties of black pepper to rice weavils and cowpea weavils. *J. Econ. Entomol.* **70**, 18.
SubbaRao, M. S., Soumithri, T. C., Johar, D. C., and Subrahmanyan, V. (1963). Studies on brine pickles. Part III. Preservative emulsion for pickles. *Food Sci.* **12**, 381.
Subrahmanyan, T. V. (1942). A new indigenous ensecticide. *Indian J. Entomol.* **4**, 238.

Subrahmanyan, V., and Srinivasan, M. (1955). Asafoetida—its origin, nature and place in human dietary and medicine. *Bull. Cent. Food Technol. Res. Inst., Mysore* **5**, 27.

Subrahmanyan, V., Sastry, L. V. L., and Srinivasan, M. (1954). Asafoetida. I. *J. Sci. Indian Res., Sect. A* **13**, 382.

Subrahmanyan, V., Krishnamurthy, K., Sreenivasamurthy, V., and Swaminathan, M. (1957a). Effect of garlic in the diet on the intestinal microflora of rats. *J. Sci. Ind. Res., Sect. C* **16**, 173.

Subrahmanyan, V., Sreenivasamurthy, V., Krishnamurthy, K., and Swaminathan, M. (1957b). Studies on the antibacterial activity of spices. *J. Sci. Ind. Res., Sect. C* **16**, 240.

Subrahmanyan, V., Krishnamurthy, K., Sreenivasamurthy, V., and Swaminathan, M. (1958). The effect of garlic on certain intestinal bacteria. *Food Sci.* **7**, 223.

Subramanyam, H., D'Souza, S., and Srivastava, H. C. (1962). Storage behaviour of ginger. *Proc. Symp. Spices—Role Natl. Econ., 1st, 1962* Abstract, p. 5.

Sugii, M., Nagasawa, S., and Suzuki, T. (1963). Biosynthesis of S. methyl-l-cysteine and S-methyl-l-cysteine sulfoxide from methionine in garlic. *Chem. Pharm. Bull.* **11**, 135.

Sugii, M., Suzuki, T., Kakimoto, T., and Kato, J. (1964). Sulphur containing amino acids and related compounds in garlic. Assimilation of sulphate - 35 S in garlic. *Bull. Inst. Chem. Res., Kyoto Univ.* **42**, 246.

Suhara, T. (1962). Detection of thio-thiamine in onions and cabbage. *J. Vitaminol.* **8**, 37; cf. *Chem. Abstr.* **58**, 751c (1963).

Suhonen, 1. (1970). On the storage life of the leek. *Acta Agric. Scand.* **20**, 25.

Sundt, E., and Saccardl, A. (1962). Thin layer chromatography and paper chromatography of vanilla flavouring compounds. *Food Technol.* **16**, 89.

Sun Spice Co. Ltd. (1970). Cayenne pepper extract. Japanese Patent 40, 264.

Surinder Kaur and Vijayaraghavan, P. K. (1961). Leaf-proteins in nutrition. *Curr. Sci.* **30**, 298.

Suzuki, J. I., Tausig, F., and Moore, R. E. (1957). Some observations on red pepper. New method for the determination of pungency in red pepper. *Food Technol.* **11**, 100; cf. *Chem. Abstr.* **51**, 9968g (1957).

Suzuki, J. 1., Dainius, B., and Kilbuck, J. H. (1973). A modified method for aflatoxin determination in spices. *J. Food Sci.* **38**, 949.

Suzuki-Shinobu Sohonsha Co. Ltd. (1963). Cosmetic aerosol preparations for the skin. French Patent 1,314,357.

Svendsen, A. B. (1960). Morphine determination in opium. *Pharmazie* **15**, 550.

Sviderskaja, Z. 1., and Govsina, O. T. (1968). Dynamics of losses of essential oil in harvested clary sage flowers. *Tr. Vses. Nauchno-Issled. Inst. Efirnomaslichen Kul't.* **1**, 104.

Svishchuck, A. A., Makhnovskii, M. K., Visotskii, M. M., and Overchuk, O. D. (1968). Obtaining water soluble chlorophyllins. *Farm. Zh. (Kiev)* **23**, 8.

Swarup, J., and Mathur, R. S. (1972). Seed microflora of some umbelliferous spices. *Indian Phytopathol.* **25**, 125.

Swedish Institute for Food Preservation Research (1968). Semsory evaluation of foods-principles and methods. *Proc. Int. Symp. Swed. Inst. Food Preserv. Res., Sweden, 1963* p. 1.

Synodinos, E., Kotakis, E., and Kotakis, G. (1964). Recent analytical method for vanilla extracts. *Riv. Ital Sostanze Grasse* **41**, 150.

Szabo, P. (1968). Experiments to reduce microbiological contemination of paprika. *Konzerv-Paprikaip.* **4**, 128.

Szabo, P. (1969). Manufacture of paprika oleoresin. *Konzerv- Paprikaip.* **5**, 7.

Szabo, P. (1970). Production of paprika oleoresin. *Am. Perfum. Cosmet.* **85**, 39.

Szirmai, E. (1961). The action of paprika and pepper on blood clotting factors *in vitro*. *Acta Med. Okayama* **15**, 91.

Tada, M., and Shiroishi, M. (1969). The effect of gamma irradiation and storage on the content of sugar in the onion bulb. *Sci. Rep. Fac. Agric., Okayama Univ.* **33**, 71.

Taiyo Chemical Industry Co. (1971). Curry powder. Japanese Patent 121,940.

Takagi, N. (1962). Sulphur containing amino acids from onions. Japanese Patent 12,280; *Chem. Abstr.* **58**, 13731e.

Talwar, Y. P., Nigam, M. C., and Handa, K. L. (1963). The essential oil from cohobation waters of *Anethum graveolens* L. *Indian Oil Soap J.* **28**, 274.

Tanaka, S., and Ono, Y. (1965). Determination of safrole in red camphor oils by infra red spectra. *Bunseki Kagaku* **14**, 711; cf. *Chem. Abstr.* **63**, 11244 (1965).

Tanasenko, F. S. (1962). Content of essential oil in the split fruits of coriander. *Sb. Nauchno-Issled. Rab. Maslichn.-Efiromaslichn. Kul't.* p. 156.

Tanasenko, F. S., and Sobol'eva, E. G. (1961). Effect of the conditions of the storage of coriander on the content of linalool (coriandrol) in the volatile oils. *Maslob.-Zhir. Prom-st.* **27**, 24.

Tandon, G. L., and Lal, G. (1960). Chemical composition of mustard (mustard flour or ground mustard) and mustard compounds. *Indian Food Packer* **14**(8), 15.

Tandon, G. L., and Siddapa, G. S. (1963). Mustard (mustard flour) and mustard compounds. *Indian Oilseeds J.* **7**, 55.

Tandon, G. L., Dravid, S. V., and Siddappa, G. S. (1964). Oleoresin of capsicum (red chilies). Some technological and chemical aspects. *J. Food Sci.* **29**, 1.

Tassan, C. G., and Russell, G. F. (1975). Chemical and sensory studies on cumin. *J. Food Sci.* **40**, 1185.

Tatar, J. (1964). Essential oil content of the condiment paprika. *Herba Hung.* **3**, 455; cf. *Chem. Abstr.* **65**, 15146g (1966).

Tausig, F., Seizuki, J. I., and Morse, R. F. (1956). Observations on black pepper. I. Analysis of bite principles. *Food Technol.* **10**, 151.

Teisseire, P. (1962). Application of gas-chromatography to perfumery. *Recherches* **12**, 54.

Teisseire, P. (1966). Contribution to the knowledge of essential oil of noble laurel leaf. *Recherches* **15**, 85.

Tekeli, S. T. (1943). "Zirast Sanatlari (Food Technology)." Y.Z.E. Maasi, Ankara, Turkey.

Tempel, K. H. (1962). Effect of garlic on experimental cholesterol atherosclerosis in rabbits. *Med. Ernechr.* **3**, 197.

Thaler, H. (1959). Ethyl vanillin—a natural ingredient of vanilla. *Dtsch. Lebensm.-Rundsch.* **55**, 172.

Tharanathan, R. N., and Anjaneyulu, Y. V. (1974). Polysaccharides from the seed mucilage of *Ocimumm basilicum* Linn. *Indian J. Chem.* **12**, 1164.

Theodose, R. (1972). Traditional methods of vanilla preparation, improvement of these techniques at the Antalaha station (Madagascar). *Proc. Int. Conf. Spices, Trop. Prod. Inst. London 1972* p. 71.

Thiems, H., and Tam, N. T. (1968). A method for spectrophotometric determination of methyl chavicol in estragon oils. *Pharmazie* **23**, 339.

Thomas, A. I., and John, J. M. (1966). Before cardamom goes to the market. *Indian Farming* **16**, 51.

Thomas, M. H. (1958). The stability of vitamin fortified dehydrated soups. *U.S. Dep. Comm., Off. Tech. Serv., PB Rep.* **144881.**

Thomas, P. M. (1965). Warehousing of spices in India. *Spices Bull. (Semin. No.)* **4**(10), 61.

Thomas, P. M. (1968). Scientific warehousing of chillies. *Indian Spices (Chilies Semin. No.)* **6**(3), 59.

Thompson, A. R., Booth, R. H., and Proctor, F. J. (1977). Onion storage in tropics. *Trop. Sci.* **14**, 19.

Thompson, C. J. S. (1927). "The Mystery and Lure of Perfumes." Lippincott, Philadelphia, Pennsylvania.

Thompson, R. H. (1966). A review of the properties and usage of methyl bromide as a fumigant. *J. Stored Prod. Res.* **1**, 353.

Thornburg, W. (1971). Pesticide residues. *Anal. Chem.* **43**, 145R.

Thorpe, E. (1945–1956). "Thorpe's Dictionary of Applied Chemistry," 4th ed., Vols. I–XII. Longmans, Green, New York.

Thrasher, J. J., and Brickey, P. M., Jr. (1968). Rapid method for the extraction of light filth from paprika. *J. Assoc. Off. Anal. Chem.* **51**, 525.

Thresh, J. C. (1876). *Pharm. J. Trans.* **7**, 21 (cited by Rogers, 1966).

Thresh, J. C. (1878). *Pharm. J. Trans.* **8**, 187 (cited by Rogers, 1966).

Thresh, J. C. (1882). *Pharm. J.* **12**, 721 (cited by Guenther, 1948–1952).

Thung, S. B. (1964). Comparative moisture determination in dried vegetables by drying after lyophilization or by the Karl Fischer method. *J. Sci. Food Agric.* **15**, 236.

Tice, L. F. (1932). A simplified and more efficient method for the extraction of capsaicin together with colorimetric method for its determination in capsicum fruit and oleoresin. *Am. J. Pharm.* **105**, 320.

Tiechert, K., Mutschler, E., and Rochelmeyer, H. Z. (1961). *Anal. Chem.* **181**, 325 (cited by Spanyar and Blazovich, 1969).

Tihanyi, S. G., and Zienty, F. B. (1970). Comparison of stability data obtained from accelerated storage and oxygen bomb tests on essential oils. *Am. Soft Drink J.* **124**, 73.

Tilgner, D. J. (1962a). Dilution tests for odour and flavour analysis. *Food Technol.* **16**, 26.

Tilgner, D. J. (1962b). Anchored sensory evaluation tests—a status report. *Food Technol.* **16**, 47.

Tilgner, D. J., and Barylko-Pikielna, N. (1957). "Analiza Organoloptyozna Zyconosci." Warszair.

Tilgner, D. J., and Barylko-Pikielna, N. (1959). Taste, acidity and method of threshold assessment. *Acta Phys. Pol.* **10**, 733.

Todd, P. (1964). Vegetable base food colouring. U.S. Patent 3,162,358 (Cl. 99–148).

Todd, P. H., Jr. (1958). Spices. Detection of foreign pungent compounds in oleoresin capsicum, ground capsicum and chili. *Food Technol.* **12**, 468.

Todd, P. H., Jr. (1960). Estimation of residual solvents in spice oleoresins. *Food Technol.* **14**, 301.

Todd, P. H., Jr., Bensinger, M. G., and Biftu, T. (1977). Determination of pungency due to capsicum by gas liquid chromatography. *J. Food Sci.* **42**, 660.

Tomita, M., Kozuka, M., Naka Gawa, and Mitsunori (1963). Alkaloids of bay leaves (*Laurus nobilis*). *Yakugaku Zasshi* **83**, 763.

Tookey, H. L., Ernst, A. J., Lohmar, R. L., and Wolff, 1. A. (1961). Evaluation of seeds galactomannan from legumes as paper sizes. *Tappi* **44**, 910; *Chem. Abstr.* **56**, 15709c (1962).

Torok, G., and Farkas, J. (1961). Assays into the cell count reducing effect of ionising radiation in ground paprika. *Keki Kozl.* **3**, 1.

Torricelli, A. (1937). Loss of essential oils in drugs kept in ordinary packages. *Mitt. Geb. Lebensmittelunters. Hyg.* **28**, 117.

Trabaud, L. (1958). Oil of lovage. *Soap, Perfum. & Cosmet.* **31**, 59.

Trojanek, J., Kavka, F., Vit, J., and Cekan, Z. (1965). Isolation of mosphine from opium poppy capsules. I. Investigation on the extraction of morphine from opium poppy capsules. *Pharmazie* **20**, 172.

Tropical Products Institute, London (1964). "Memorandum on Ginger," Rep. No. G.8. Trop. Prod. Inst., London.

Tropical Products Institute, London (1968). "Essential Oil Production in Developing Countries," Rep., pp. 1–164. Trop. Prod. Inst., London.

Truitt, F. B., Jr., Calaway, E., III, Brande, M. C., and Krantz, J. C. (1961). Pharmacology of myristicin. Psychopharmacology of nutmeg. *J. Neuropsychiatry* **2**, 205.

Tsatsaronic, G. C., and Kehayoglou, A. H. (1964). Colorimetric examination of green pepper. *Chem. Chron. A* **29**, 273 (in Greek).

Tsuno, S. (1958a). XVI. Allinase in Allium plants. *Vitamins* **14**, 659.
Tsuno, S. (1958b). XIX. Thiamine content of Allium plants. *Vitamins* **14**, 676.
Tsuno, S., Murakami, E., Tazoe, K., and Kikumoto, S. (1960). Influence of garlic and related sulfur compounds upon mouse liver rhodanese. *Vitamins* **20**, 112.
Tucakov, J. (1964a). Influence of exogenous factors on the yield and the quality of *Thymus vulgaris* L. Oil. *Fr. Perfum* **7**, 277.
Tucakov, J. (1964b). The influence of ecological factors on the yield and the quality of the oil of *Thymus vulgaris* L. *Riv. Ital. Essenze, Profumi, Piante Off., Aromi, Saponi, Cosmet.* **46**, 376.
Turhan, B. (1950). A new adulterant of saffron. *Pharm. Acta Helv.* **25**, 337.
Tyihak, E., and Vagujfalvi, D. (1962). Thin layer chromatography of essential oils. I. *Herba Hung.* **1**, 95; *Chem. Abstr.* **62**, 3879 (1965).
U.S. Department of Agriculture (1921). Bureau of Chemical Service and Regulatory Announcement, No. 26, p. 14; *Analyst* **46**, 144; *Chem. Abstr.* **15**, 2693 (1921).
U.S. Department of Agriculture (1970). "Foreign Agriculture Circular," Trop. Prod. No. FTEA, 1–70, p. 1. U.S.D.A., Washington, D.C.
U.S. Food and Drug Administration (1957). Pesticide chemicals tolerance for residues of ethylene oxide. *Fed. Regist.* **22**, 93.
Vagujfalvi, D., and Tyihak, E. (1963). Reagents of thin-layer chromatography of essential oils. *Herba Hung.* **2**, 361; *Chem. Abstr.* **63**, 6781b (1965).
Vajdi, M., and Peperira, R. R. (1973). Comparative effects of ethylene oxide gamma irradiation and microwave treatments on selected spices. *J. Food Sci.* **38**, 893.
Van Blaricum, L. O., and Martin, J. A. (1945). Effect of pre-drying treatments and varying storage conditions upon retention of colour by cayenne pepper. *S.C., Exp. Stn., Annu. Rep.* **57**, 39.
Van Blaricum, L. O., and Martin, J. A. (1951). Retarding the loss of red colour in cayene pepper with oil soluble anti-oxidants. *Food Technol.* **5**, 337.
van Os, F. H. L. (1950). Coriander seeds. *Pharm. Weekbl.* **85**, 732.
van Os, F. H. L. (1965). The content of essential oils in plant drugs. *Pharm. Weekbl.* **100**, 377.
van Oss, J. F., and van Oss, C. J. (1956). "Warenkennis on Technologie," 6th ed., p. 39.
van Pinxteren, J. A. C., and Verloop, M. E. (1962). Oxidimetric determination of morphine. *Pharm. Weekbl.* **97**, 1.
Varga, L. (1938). Change of stomach acidity under various stimuli. *Orv. Hetil.* **80**, 702.
Variati, G. L., and Rovesti, P. (1960). Aroma-cosmetic action of the essential oil of rosemary. *Riv. Ital. Essenze, Profumi, Piante Off., Olii Veg., Saponi* **42**, 536.
Varo, P. T., and Heinz, D. E. (1970a). Volatile components of cumin seed oil. *J. Agric. Food Chem.* **18**, 234.
Varo, P. T.; and Heinz, D. E. (1970b). Identification and characterization of 1,4-p-methadien-7-al isolated from cumin seeds. *J. Agric. Food Chem.* **18**, 239.
Varshney, I. P., and Sharma, S. C. (1966). Saponins and sapogenins. XXXII. Trigonella. *J. Indian Chem. Soc.* **43**, 564; cf. *Chem. Abstr.* **64**, 7044b (1966).
Vashista, V. N., Nigam, M. C., and Handa, K. L. (1963). The essential oil of *Majorana hortensis*. *Riechst. Aromen* **13**, 61.
Vaughn, R. H. (1951). The microbiology of dehydrated vegetables. *Food Res.* **16**, 429.
Vaughn, R. H. (1970). Incidence of various groups of bacteria in dehydrated onions and garlic. *Food Technol.* **24**, 83.
Veek, M. E., and Russell, G. P. (1973). Chemical and sensory properties of pimento oil. *J. Food Sci.* **38**, 1028.
Venketachalam, K., and Gray, R. N. (1974). Marketing of spice and spice products. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 73.
Verbeek, G. (1970). Leeks can be kept healthy. *Tuinbouwberichten* **34**, 26.
Vereshchagin, A. G. (1962). Composition of poppy seed oil. *Biokhimiya* **27**, 866.

Verma, S. M. (1960). Carvone analysis by ultra-violet absorption in Indian dill oil (*Anethum Sowa*). *Curr. Sci.* **29**, 345.

Videki, L., and Videki, 1. (1961). Determination of pigment content of paprika by spectrophotometric (ASTA) method. *Elelmiszervizsgalati Kozl.* **7**, 103.

Vidyarthi, N. L., and Dasa Rao, C. J. (1939). Fatty acids and glycerides of the fat from the seeds of *Garcinia indica (Kokum Butter)*. *J. Indian Chem. Soc.* **16**, 437.

Vijayan, P. K. (1974). Solar energy for cardamom curing. *Proc. Symp. Dev. Prospects Spice Ind. India, 1974* p. 37.

Vik, J. (1972). The effects of plant spacing plastic tunnels and row distance on total yield, size, keeping quality and skin colour of direct seeded onions in experiments at Landvik during 1967–69. *Meld. Nor. Landbrukshoegsk.* **51**, 1.

Vilkki, P. (1954). Chromatographic studies on the formation of pyruvic acid in onion juice. *Suom. Kemistil. B* **27**, 21.

Villalobos, P. A., and Sagues, G. L. (1967). Prolonging the storage of Valenciana onions by applying a sprouting inhibitor. *Invest. Prog. Agric.* **1**, 33.

Villalobos, P. A., Kocher, G. F., and Ramirez, U. M. (1966). The inhibition of sprouting in Valencia onions during storage by maleic hydrazide sprays. *Agric. Tec. (Santiago)* **26**, 69.

Villanua, L., Nunez, M., Portoles, A., and Fernandez, P. M. J. (1954). Analysis de condiments (Analysis of condiments). *An Bromatol.* **6**, 399.

Viraktamath, C. S. (1964). Studies on the packaging of spices—dry chilies. (*Capsicum annuun*). *Indian Food Packer* **18**(4), 4.

Viraktamath, C. S., Iyengar, N. V. R., and Sreenivasan, A. (1964). Packaging and storage of dry green cardamom. *Res. Ind.* **9**, 33.

Virmani, O. P., and Datta, S. C. (1968). Oil of spearmint. *Perfum. Essent. Oil. Rec.* **59**, 351.

Virmani, O. P., and Datta, S. C. (1970a). Oil of *Mentha piperita* (oil of peppermint). *Flavour Ind.* **1**, 59.

Virmani, O. P., and Datta, S. C. (1970b). Prospects for production of essential oil of *Mentha spicata* (spearmint) at Lucknow. *Flavour Ind.* **1**, 549.

Virmani, O. P., and Datta, S. C. (1970c). Essential oil of *Anethum graveolens*. *Flavour Ind.* **1**, 856.

Virmani, O. P., Gulati, B. C., and Datta, S. C. (1968). Present production of some of the important essential oils in India. Part IV. *Perfum. Essent. Oil Rec.* **59**, 702.

Virtanen, A. 1., and Matikkala, E. J. (1959). Isolation of S. methyl and S. propyl cystiene sulfoxide from onion and the antibiotic activity of crushed onion. *Acta Chem. Scand.* **13**, 1898; *Chem. Abstr.* **56**, 13250e (1962).

Virtanen, A. I., and Matikkala, E. J. (1960). Neua-glutanylpeptide inder ziviebel (*Allium cepa. Hoppe-Seyler's Z. Physiol. Chem.* **322**, 8.

Virtanen, A. I., and Mattila, I. (1961a) γ-1-Glutanyl-s-allyl-1-cystein in garlic. *Suom. Kemistil. B* **34**(3), 44.

Virtanen, A. 1., and Mattila, 1. (1961b). γ-1-Glutamyl peptides in garlic (*Allium sativum*). *Soum. Kemestil. B.* **34**, 73.

Virtanen, A. 1., and Spare, C. G. (1962). 2-methyl-2-pentenal, a compound in onion vapours. *Soum. Kemistil. B* **35**, 28 (cited by Spare and Virtanen, 1963).

Viseglio-Tomasset, L. (1966). Gas chromatographic examination of some essential oils of *Acrorus calamus*. *Ind. Agrar.* **4**, 153; *Anal. Abstr.* **14**, No. 5051 (1967).

Viswanath, B., and Ayyar, C. V. R. (1934). Report on the examination of samples of Indian and foreign coriander. *Agric. Live-Stock India* **4**, 583.

Viswanathan, T. K. (1967). Grading of black pepper at the level of farmers through the growers co-operatives. *Indian Spices* **5**(3), 27.

Voelker, W. A., Skarzynski, J. N., and Stanl, W. H. (1967). Determination of geographical origin

of spices. 1. Cassia and cinnamon by thin layer chromatography. *J. Assoc. Off. Anal. Chem.* **50**, 852.

von Fodor, K. (1930). Capsaicin reaction and evaluation of paprika. *Kiserletugyi Kozl.* **33**, 155.

von Fodor, K. (1931). New reaction for capsaicin. *Z. Unters. Lebensm.* **61**, 94.

von Jaminet, L. (1965). Ginger and ginger oil. *Seifen, Oele, Fette, Wachse* **91**, 823.

von Jaminet, L. (1968). Ginger plants and their essential oils. *Seifen, Oele, Fette, Wachse* **94**, 209.

von Rudloff, E., and Hefendehl, F. W. (1966). Gas liquid chromatography of terpenes. XV. The volatile oil of *Mentha arvensis* Var. *Glabrata Ray. Can. J. Chem.* **44**, 2015.

Vorontsova, R. V., and Zabolotskaya, A. S. (1970). Effect of pesticides on essential oil content of bay leaves. *Khim. Sel'sk. Khoz.* **8**, 510.

Voskerusa, J. (1960). Morphine content of opium poppy capsules. *Pharmazie* **15**, 552.

Wachsmuth, H., and Lenaers, R. (1946). Determination of cinnamaldehyde and other essential principles. *J. Pharm Belg.* [N.S.] **1**, 65.

Wagenaar, M. (1928). The detection of rice flour in other flours and in spices. *Pharm Weekbl.* **65**, 246.

Waldi, D. (1958). "Chromatography of the Alkaloids." E. Merck A. G., Darmstadt (cited by Spanyar and Blazowich, 1969).

Walker, M. C., Morell, S., and Foster, H. H. (1937). Toxicity of mustard oils and related sulphur compounds to certain fungi. *Am. J. Bot.* **24**, 536.

Wallace, A. R., Porte, A. L., and Hodges, R. (1963). Lignans from *Myristica otoba.* The structure of hydroxyotobain and isootobain. *J. Chem. Soc.* p. 1445.

Walter, R. H. (1972). β-Caryophyllene in native clove bud oil. *Phytochemistry* **11**, 405.

Waly, Y. A., and Hassan, Y. M. (1965). Qualitative chromatographic survey of the sugars prevailing in some horticultural crops. *Proc. Am. Soc. Hortic. Sci.* **87**, 264.

Wands, S. P. (1972). The American Spice Market 1972. *Proc. Int. Conf. Spices, Trop. Prod. Inst. London, 1972* p. 233.

Wannamaker, A. K. (1966). Effects of sulfur dynamizations on onions. *J. Am. Inst. Homeopathy* **59**, 287.

Ward, J. F. (1961). "Pimento," Bull. p. 1. Minist. Agric. and Lands, Jamaica.

Warmbrod, F., and Fry, L. J. (1966). Coliform and total bacterial counts in spices, seasonings and condiments. *J. Assoc. Off. Anal. Chem.* **49**, 678.

Watanabe, E. (1953). *Vitamins* **6**, 121 (cited by Krishnamurthy and Sreenivasamurthy, 1956).

Watanabe, H., and Goto, M. (1954). Minute constituents of crude drugs. 1. Biotin. *J. Pharm. Soc. Jpn.* **74**, 157.

Watanabe, H., Iwata, S., and Otani, Y. (1963). Active principles in garlic. *Osaka Shiritsu Daigaku Kaseigakuba Kiyo* **11**, Part 1, 1.

Wayse, S. B. (1967). Effect of N, P and K on yield, keeping quality of onion bulbs (*A. cepa*). *Indian J. Agron.* **12**, 379.

Webb, A. H., and Tanner, F. W. (1945). Effect of spices and flavouring materials on growth of yeast. *Food Res.* **10**, 273.

Weil, A. T. (1965). Nutmeg as a narcotic. *Econ. Bot.* **19**, 194.

Weisenfelder, A. E., Huffman, V. L., Villalon, B., and Burns, E. E. (1978). Quality and processing attributes of selected Jalapeno pepper cultivars. *J. Food Sci.* **43**, 885.

Wellendorf, M. (1966). Gas-chromatography investigation of cardamom types. *Dan. Tidsskr. Farm.* **40**, 156.

Wendt, A. (1963). Historical AOAC data on four FEMA (Flavouring Extract Manufacturers' Association) samples of vanilla extracts. *J. Assoc. Off. Agric. Chem.* **46**, 337.

Wenkam, N. S., Miller, C. D., and Kanehiro, Y. (1961). Sodium content of Hawaii-grown fruits and vegetables in relation to environment. *J. Food Sci.* **26**, 31.

Wesley, F., Rourke, B., and Darbshire, O. (1965). The formation of persistant toxic chlorohydrins in foodstuffs by fumigation with ethylene oxide and with prophylene oxide. *J. Food Sci.* **30**, 1037.

Westaway, H., and Williams, J. F. (1959). Oil of nutmeg. *J. Appl. Chem.* 9, 440.

Wheeler, O. H., Nieto, M. A., De Storer, C. B., Antunano, N. C., and Medina, V. Y. (1961). Analysis of some aldehydes, and ketones in essential oils by gas chromatography. *Rev. Colomb. Quim. P. R.* **18**, 31.

White, R. T. (1957). Studies on the storage and shipment of whole black pepper grown in the orient. *J. Econ. Entomol.* **50**, 423.

Wick, E. L., and Cairncross, S. E. (1956). A chemical and flavour study of off-flavour due to artificial pepper. *Food Technol.* **10**, 423.

Wijesekera, R. O. B., and Fonseka, K. H. (1974). Essential Oils II. Infra-red spectroscopy in the analysis of the volatile oil of cinnamon. *J. Natl. Sci. Coun.*, Sri Lanka, **2**(1), 35.

Wijesekera, R. O. B., and Jayawardena, A. L. (1972). Recent developments in the production of spices and their essential oils in ceylon. *Proc. Int. Conf. Spices, Trop. Prod. Inst., London, 1972* p. I59.

Wijesekera, R. O. B., Senanayake, U. M., and Jayawardena, A. L. (1972). Methods for the quality control of spice oleoresins. 1. The oleoresin of pepper. *Flavour Ind.* **3**, 33.

Wijesekera, R. O. B., Jayawardena, A. L., and Rajapakse, L. S. (1974). Volatile constituents of leaf, stem and root oils of cinnamon (*C. zeylanicum*). *J. Sci. Food Agric.* **25**, 1211.

Wilhelm, P. (1921). The presence of magnesium salt in cloves and its detection. *Z. Unters. Nahr.-Genussm. Gebrauchsgegenstaende* **42**, 246.

Willis, J. C. (1951). "A Dictionary of the Flowering Plants and Ferns," 6th rev. ed. Cambridge Univ. Press, London and New York.

Wilson, C. W. (1965). Separation of volatile flavours from celery. *Proc. Fla. State Hortic. Soc.* **78**, 249.

Wilson, C. W., III (1969a). Terpene and sesquiterpenes hydrocarbon in the essential oil from fresh celery. *J. Food Sci.* **34**, 521.

Wilson, C. W., III (1969b). Identification and quantitative estimation of alcohol in celery essential oil. *J. Food Sci.* **34**, 535.

Wilson, C. W., III (1970). Relative recovery and identification of carbonyl compounds from celery essential oil. *J. Food Sci.* **35**, 766.

Wilson, C. W., III *et al.* (1969). Relationship of methods for recovering essential oil from fresh celery on the chemical composition and flavour. *Proc. Fla State Hortic. Sci.* **82**, 187.

Wink, F. A. (1946). Determining the moisture equilibrium curves of hygroscopic materials. *Ind. Eng. Chem., Anal. Ed.* **18**, 251.

Wink, F. A. (1947). Moisture equilibrium. *Mod. Packag.* **20**, 135.

Winkler, W., and Lunau, E. (1959). The differentiation of essential oils of *Curcuma xanthorhiza* and *Curcuma longa* by thin-layer chromatography. *Pharm. Ztg.* **104**, 1407; *Chem. Abstr.* **54**, 15847e (1960).

Winterton, D., and Richardson, K. (1965). Constituents of Queensland-grown ginger. *Queensl. J. Agric. Anim. Sci.* **22**, 205.

Winton, A. L., and Winton, K. B. (1916). "Microscopy of Vegetable Foods." Wiley, New York.

Winton, A. L., and Winton, K. B. (1939). "Structure and Composition of Foods," Vol. IV. Wiley, New York.

Winton, A. L., and Winton, K. B. (1945). "Analysis of Foods." Wiley, New York.

Wistreich, H. E., and Schafer, W. E. (1962). Freeze-grinding ups products quality. *Food Eng.* **34**(5), 62.

Woggon, H., and Kohler, U. (1960). Paper chromato-oscillopolarographic determination of vanillin and ethylvanillin in foods. *Ernaehrungsforschung* **5**, 402.

Wolter, J. (1964). Differences in the occurrence of free amino acids in some diploids and polyploid crop plants. *Ann. Univ. Mariae Curie-Sklodowska, Sect. E* **19**, 347.

Woodman, A. C. (1941). "Food Analysis. Typical Methods and the Interpretation of Results," 4th ed. McGraw-Hill, New York.

Woodward, E. R. (1949). Sterilisation of spices. U.S. Patent 2,482,958.

Worseck, M. (1958). Preservation of foodstuffs by irradiation. *Berl. Muench. Tieraetztl. Wochenschr.* Wsghr. 11.

Worseck, M. (1959). *Z. Lebensm.-Unters. -Forsch.* **109**, 366.

Worthington, J. T. (1961). Moisture content and texture of dried chili peppers stored in polyethelene-lined burlap bags. *U.S., Agric. Market. Serv.* **AMS-444**, 1.

Woyke, H., and Manczak, M. (1965). Number of seed stalks and seed yield of several newly bred and local onion varieties as affected by the temperature at which the bulbs had been stored. *Biul. Warzywniczy* **8**, 239.

Wright, W. J., Bice, C. W., and Fogelberg, J. M. (1954). The effect of spices on yeast fermentation. *Cereal Chem.* **31**, 100.

Wrolstad, R. E. (1965). The terpene hydrocarbons of *Piper nigrum*. *Diss. Abstr.* **26,** 1334 (Univ. Microfilms, Ann. Arbor, Michigan, Order No. 65-2947).

Wrolstad, R. E., and Jennings, W. G. (1962). Volatile constituents of black-pepper. II. Effects of light and heat. *Food Technol.* **16**, 107.

Wrolstad, R. E., and Jennings, W. G. (1964). Sources of monoterpene hydrocarbons. *J. Agric. Food Chem.* **12**, 507.

Wrolstad, R. E., and Jennings, W. G. (1965). Volatile constituents of black pepper. III. Monoterpene hydrocarbon fraction. *J. Food Sci.* **30**(2), 274.

Wulff, H. D., and Stahl, E. (1960). Chemical races of *Acorus calamus*. *Naturwissenschaften* **47**, 114.

Wurziger, J. (1968). Nutmegs and nutmeg powder. *Gordian* **68**, 441.

Yamada Seiyaku, K. K. (1973). Garlic powder. Japanese Patent 734,737.

Yamamoto, K. (1960). Experimental study on ginger ingestion. Dermatological study on spice ingestion. *Igaku Kenkyu* **30**, 207.

Yamanishi, T., Sato, T., and Omura, A. (1964). Linalool epoxides in the essential oils from freshly picked tea leaves and black tea. *Agric. Biol. Chem.* **28**, 653.

Yellearacamino, A. (1971). Rosemary essence in Spain and in Mediterranian countries. *Ion (Madrid)* **31**, 217.

Yesair, J., and Williams, O. B. (1942). Spice contamination and its control. *Food Res.* **7**, 118.

Yin, Y., Zarglami, N., and Heinz, D. E. (1970). Effects of pH and temperature on volatile constituents of caraway. *J. Food. Sci.* **35**, 531.

Yoshimura, M. (1958). The nutritional value of plants. X. Studies to prove existence of allicin and its homologs. *Vitamins* **14**, 627.

Yoshimura, M., and Arai, T. (1958). XIV. Doubts of theory of intestinal thiamine synthesis. *Vitamins* **14**, 650.

Yoshimura, M., Tsuno, S., and Murakami, F. (1958). XV. Aliin homologs in Allium plants. *Vitamins* **14**, 654.

Yoshio Hirose, Shigehiro, Shokoeto *et al.* (1962). The utilisation of plant products. II. (7–15). *Kumamoto Pharm. Bull.* **5**, 36.

Yurugi, S. (1954). *J. Pharm. Soc. Jpn.* **74**, 519 (cited by Krishnamurthy and Sreenavasamurthy, 1956).

Zacsko-Szasz, M., and Szasz, G. (1965). Thin layer chromatographic studies on aniseed oil. *Fette, Seifen, Anstrichm.* **67**, 332.

Zacsko-Szasz, M., and Szasz, G. (1966). Thin layer chromatography of aniseed oil. *Herba Hung.* **5**, 91.

Zaidi, S. H., and Mukerjee, B. (1958). Experimental peptic ulceration. Part I. The significance of mucous barrier. *Indian J. Med. Res.* **46**, 27.

Zaidi, S. H., Singh, G. B., and Khanna, N. M. (1958). Experimental peptic ulceration. IV. Effect of mucous stimulants on histanine induced ulceration. *Indian J. Med. Res.* **46**, 732.

Zanina, C., Posso, A. D., and Dasi, A. (1961). Principal components of oil from dwarf pine needles. *Boll. Chim. Farm.* **100**, 83; *Chem. Abstr.* **55**, 20335g (1961).

Zawahry, M. R. (1964). Isolation of a new hypotensive fraction from *Nigella sativa* seed. *Kingr. Pharm. Wise Vorlr Originalwitt* **23**, 193.

Ziegler, J. A. (1969). (Griffith Laboratories Ltd.) Liquid pepper. Canadian Patent 807,614.

Zielinska-Sowicka, R., and Szepczynska, K. (1965). Investigation of essential oils by thin layer chromatography. *Ann. Acad. Med. Lodz.* **6**, 35.

Zinchentko, L. M. (1959). The pigment composition of onion-skin and the biological activity of individual components. *Uch. Zap., Pyatigorsk. Farm. Inst.* **3**, 68.

Zitko, V. (1957). Determination of capsaicin, I. Reaction with ρ-diazobenzene sulfonic acid. *Chem. Zvesti* **11**, 590; *Chem. Abstr.* **52**, 7947d (1958).

Zitko, V., and Durigova, Z. (1961). Biochemistry in the production of spice paprika. III. Phenolic products. *Chem. Zvesti* **14**, 450.

Zitko, V., and Ondreicka, R. (1960). Biochemistry in the production of spice paprika. IV. Various kinds of natural wilting. *Chem. Zvesti* **14**, 535.

Zofia, E. K. (1948). *Satigmata croci*-Saffron-history and recent advances of biological science. *Ann. Univ. Mariae Curie-Sklodowska, Sect. E* **3**, 123.

Zolotovich, G., Nachev, Kh., and Silyanovska, K. (1967). Cytotoxic effect of carbonyl compounds isolated from essential oils. *C.R. Acad. Bulg. Sci.* **20**, 1213.

Zoschke, M. (1962). Genetically caused variability in the alkaloid content of *Papaver somniferum*. *Angew. Bot.* **36**, 185.

Zsadon, B. (1966). Preparation of narcotoline and morphine from poppy capsules. *Herba Hung.* **5**, 205.

Zwergal, A. (1952). Active principles of *Allium sativum*. *Pharmazie* **7**, 245; *Chem. Abstr.* **47**, 3224c (1953).

INDEX

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G

N

O

P

Q

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ISBN 0-12-016454-7