NATIONAL FARM CHEMURGIC COUNCIL

BULLETIN

THE DOMESTIC PRODUCTION OF ESSENTIAL OILS FROM AROMATIC PLANTS

A Compilation of Research Papers Relating to Problems in the Agricultural Production of Essential Oils From the Cultivation of Aromatic Plants

Published by the

National Farm Chemurgic Council

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Preface

The consumption of aromatic seeds, roots and essential oils in the United States and the money spent to import them from abroad is so great as to warrant a study of the possibility of a domestic cultivation of aromatic plants and the establishment of an essential oil industry. From over one hundred known principal variations of the aromatic plants produced, only three are cultivated in this country—mint, wintergreen, and citrus; and of secondary importance—apricots, peaches and almonds.

This bulletin presents six new varieties of aromatic plants for introduction in this country and shows the quantity which the United States buys and the amount of money expended in purchasing seeds, roots, and essential oils from each from abroad. The economic possibilities for such an industry are encouraging since soil and climatic conditions in the United States admit the cultivation of aromatic plants over wide areas with a promise of high profit; no elaborate equipment is required; the standard agricultural implements are applicable to the cultivation of aromatic plants; the acre yield would give a higher profit than any other crops in the United States; an average crop of the six varieties to meet present demands comprises 100,000 acres, which is by no means an inconsiderable agricultural area, especially where the problem of overproduction in staple crops must be met; and the cost of production is cheap, in comparison with the price received for the seeds and roots and the present marketability of the oil.

The cultivation of aromatic plants and the production of essential oils in this country present an open field, and if farmers would attempt to grow these plants, there is no doubt but that they would be greatly recompensed. The authors have grown these plants experimentally and have met with much success.

In conclusion, the advantage of aromatic plants in the light of specialty crops should be stressed. Cultivation of specialty crops lends individuality to the farmer and establishes him as an initiator. Successful enterprises in business have resulted from ingenuity, not imitation. The cultivation of these plants, new to domestic agriculture, identifies itself with success.

In this bulletin the authors have presented data from the cultivation standpoint; included the various uses of the seeds, roots and oils; presented data from the engineering aspect; from quality tests, and lastly, from laboratory assayments.

The authors wish to express their indebtedness to the following members of the Seagram Research Department staff: to Miss Alexandra de

Glazoff, M.Sc., who is in charge of the Gin and Botanical Division, for the analytical work and compilation of the data; to William D. Gray, Ph.D., research mycologist, for critical reading of the botanical portions of the manuscript; to Mark T. Muller, B.Sc., for the photographic work; and to Miss Marjorie A. Metzner, M.Sc., for the preparation of the manuscript.

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The AUTHORS

Louisville, Kentucky
August, 1940

I

RAW MATERIALS FOR ESSENTIAL OILS

by HERMAN F. WILLKIE

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I.

Raw Materials For Essential Oils[1]

By HERMAN F. WILLKIE *and* PAUL J. KOLACHOV

A. INTRODUCTION

A survey of the annual consumption of raw materials for essential oil in the United States during the last five years yields no data on the domestic (U.S.) production of these commodities but gives only an indication of the consumption of these products in the import statistics. A few, no doubt, are produced here, but a glance at the volume imported and the evaluation of these commodities renders the investigation of mass domestic production worthwhile.

According to the government records from the U. S. Department of Commerce, licorice root sale to this country for 1937 and 1938 was over one and a quarter million dollars a year, representing a volume of seventy million pounds a year. From 1935 to 1938 inclusive, about twenty-five million pounds of caraway seed was imported valued at over one and a half million dollars. Essential oil from anise seed importation during this same period amounted to one million pounds costing us approximately one million dollars. Present war conditions abroad have virtually doubled, and in some cases even tripled, the price of these products.

It has been the authors' experience, moreover, that raw materials for essential oils from abroad are inferior in quality to what could be produced in this country, if given the proper attention and control. The production in Europe is not controlled, the source of supply is not identical, and many times shipments are adulterated.

In addition, the distributors of these raw materials in this country are without means of control over the products because they lack definite specifications for the individual commodities. Suitable specifications will be discussed subsequently in this article wherein are described methods by which the materials are analyzed.

In the consideration of the domestic production of these materials, another important point to be brought to the foreground is the utilization of land. Production of raw materials for essential oils would play a significant role in the agricultural problem of this country. Under the present administration the elimination of certain crops in consequence of the crop control program has left and will leave many fields idle. Sowing seeds of plants for essential oil production on this unused land not only would aid the farmers and develop a domestic market, but the products therefrom could be controlled to promote the supply of uniform, definitely specified materials.

Investigation of the growth of these commodities in Europe and the small experimentation that has been done in this country convince the

[1] From lecture delivered to the Sixth Annual Chemurgic Conference of the National Farm Chemurgic Council, Chicago, Illinois, March 29, 1940.

authors that conditions in the United States are quite satisfactory for domestic production.

The following pages are devoted to a discussion of the formation and role of essential oils in plants, descriptions of the plants presented for cultivation in this country, and their uses and importance in food and allied industries. The latest techniques in analytical methods are described in detail with illustrations of specially adapted laboratory apparatus for the benefit of food technicians.

B. ESSENTIAL OILS IN BOTANICALS

Definition and Location

The term essential oils is applied in general to fragrant compounds formed in plants. Some authors consider as essential oils only the parts of the plants recovered by steam distillation. Others include in this definition compounds extracted by solvents. In either case, essential oils are the mixture of volatile fragrant compounds belonging to various groups of organic classifications and found in the aromatic plants.

These essential oils are situated in cavities of the plants called "carriers" of essential oils. These cavities may include resinous compounds, and often the essential oils form an emulsion through a combination with intercellular substances. In recent years it has been proven that essential oils and resins are present in the cells of the parenchyma. Essential oils can be found in almost any part of a plant (stems, leaves, flowers, and fruit.) Sometimes they are confined to a specific anatomic portion of the plant; in other instances they may be distributed unequally in all parts of the plant. Usually the greatest amount of oil is contained in the reproductive organs such as the flowers and the fruit. Leaves and stems contain a lesser amount of oil than the reproductive organs while the roots usually have a low content of oil. Some plants contain oils not in a free state but in the form of glucosides or some other hetero compounds. In such instances in order to extract the oil, it is necessary to split the compound by fermentation.

Short Historical Review

It is impossible to establish with certainty when the essential oils were first extracted by man, but apparently they were known to the ancients. Indications relating to the use of calamus root and rose petal oils are found in ancient Sanskrit papers. Persian and Egyptian sources show us that extraction and distillation of aromatic compounds were known to people several thousand years before Christ. The Romans utilized various spices and aromatics brought principally from the Orient.

In the fourth century A.D. there appeared first in Arabia and later in Europe voluminous papers on methods of distillation of plants and detailed descriptions of apparatus. The oils obtained were used in medicines and perfumes. Nevertheless, the structure of these compounds was very little understood and was explained by the phlogiston theory.

In the nineteenth century the work of scientists threw more light on the entire field of the structure and properties of oils and led to additional research and discoveries. Even at the present time, the field of essential oils is far from adequately covered, and there are numerous plants which as yet have not been investigated.

Role of Essential Oils in the Plants

Up to the present time there is no universally accepted theory concerning the formation of essential oils and their role in the life of plants but there exist numerous hypotheses, some of which unfortunately are supported inadequately by experimentation. Lack of such confirmation is thought to be due to the complexity of the essential oils, which renders identification and separation difficult.

There are enumerated below a few of the existing hypotheses on the role of essential oils in plants.

1. Aromatic compounds serve as a protection against insects and fungi growth. This role appears to comprise only a small part of their importance because we know that certain plants such as *Foeniculum officinalis* (fennel), although rich in essential oil, suffer very badly from insects and fungi.
2. Essential oils afford self-preservation to plants.
3. The contents of resinous canulae in evergreen plants serve to cover wounds in the bark and cambium against moisture and contamination by fungi. This opinion enjoys wide popularity.
4. The aroma of flowers attracts insects which promote reproduction.
5. Essential oil by penetrating the intercellular spaces decreases the transpiration of the plant.
6. Essential oils increase the speed of circulation of nutritive substances in the plant which regulate the metabolism.
7. Essential oils split the glucosides; in other words, they act as enzymatic agents.
8. Essential oils are formed in the chlorophyll-bearing parts of the plant from whence they migrate during the period of reproduction toward the organs of the latter. There some of the essential oil is consumed and the remaining part returns to the leaves at the cessation of reproduction. This hypothesis indicates that the theory of essential oils being a product of elimination is not valid.
9. Aromatic compounds the same as alkaloids may play the role of hormones in plants.
10. Essential oils serve to protect the plants against the cold by evaporating and forming a protective cloud about them. Since essential oils and resins found in evergreens were in the winter on the surface of the parenchyma, and since it is known that the presence of oil on the surface of the water lowers its freezing point considerably, this conclusion was advanced recently.

Opinions concerning the role of essential oils are very numerous. This may be explained either by the dearth of knowledge relating to these compounds or by the fact that essential oils possess several specific qualities, rather than a single one.

Formation of Essential Oils in Plants

Again there are only hypotheses to explain the formation of essential oils in plants. As essential oils are formed from many different compounds, often unrelated, it is impossible to generalize on one scheme. We shall mention only the most important opinions on formation.

1. There is one theory that terpenes (principal constituents of essential oils) are formed by the addition of beta-amino normal butyric acid to l-leucine with the elimination of ammonia and water, or by the addition of alpha-amino isocaproic (hexanoic) acid to alanine (alpha-amino propionic acid) to yield cymene. Cymene plus leucine gives cinnamic acid, one of the main constituents of cinnamon bark. Cinnamic acid and cymene each isomerize to yield terpenes and, therefor, essential oils.

2. Another opinion is that terpenes are formed from carbohydrates; the first step is the formation of beta-methylcrotonic aldehyde from acetone and acetaldehyde. Two molecules of methylcrotonic aldehyde polymerize to form geraniol which in turn isomerizes very easily to terpenes.

3. Some authors consider that essential oils are formed from isoamyl alcohol, this alcohol in turn being formed from leucine. Hence terpenes are derived from proteins. Substantiating evidence is the fact that isoamyl aldehyde has been found in several essential oils such as orange, lemon, etc. In other words, the existing theories so far admit the possibilities of the formation of terpenes from both carbohydates and proteins.

It also has been proved that during the life of plants the composition and quantity of essential oils are considerably changed. This leads to the thought that essential oils may be some sort of food storage for the plants. The study of physical properties of essential oils during different periods of growth of plants disclosed at least one certainty—that essential oils play a significant role in the life of plants.

This study has aided in arriving at important conclusions by means of which standard specifications have been set up to be applied to the purchase and commercial use of plants bearing essential oils. It is now possible to predict with accuracy the amount and composition of essential oil in the plant during its different periods of growth. The value of such knowledge can be understood readily after the detailed descriptions of several important plants which follow, are read.

C. BOTANY, DESCRIPTION, CULTIVATION, PROPERTIES AND USES OF THE INDIVIDUAL SPECIES

CORIANDER — *Coriandrum sativum L.* — *Umbelliferae*

History

The exact place of the first appearance of coriander is unknown. The first reference to it is found in the early Egyptian papyri. It is mentioned in the Bible as having resemblance to manna and is also referred to in the medical books of the sixteenth century. It was introduced into America before 1670 in Massachusetts.

Regions of Cultivation

Coriander originated in southern Europe and in the eastern part of Asia Minor. It grows wild or can be cultivated. Wild coriander is especially indigenous to the southern part of Europe, Asia Minor, and south Russia. It is cultivated mainly in Russia, Moravia, England (Essex), France (Morocco), Germany (Thuringia), Hungary and Bulgaria. The best coriander is considered to be Russian, Moravian, and Hungarian. The inferior kinds are South Africa, Palestinian, and Indian.

Percent of Oil in Coriander

East India	0.15—0.20%
North Africa	0.25—0.35%
Syria	0.26—0.34%
Persia	0.25—0.35%
France (Morocco)	0.25—0.35%
Palestine	0.27—0.36%
Asia Minor	0.30—0.40%
Mexico	0.35—0.45%
Italy	0.50—0.60%
Holland	0.50—0.60%
Germany (Thuringia)	0.60—0.80%
Moravia	0.65—0.90%
Russia	0.75—1.20%
Hungary	0.50—1.00%

Botanical Description

STEMS. Coriander belongs to the plant of the family *Umbelliferae*. It is an annual plant with a round, hair-free stem, growing about two feet high, and 0.06—0.2 inch diameter, with branching stems. Before ripening, the stems of some types of coriander acquire an almost black color because of the presence of anthocyanin pigments.

ROOTS. The roots are very thin and sinuous.

LEAVES. The leaves are bipinnate, the lower ones divided into broad or wedge-shaped, deeply-cut segments, while the upper ones are divided into narrow parts and are more finely cut. The lower leaves on the stem are shiny with long petioles largest at their base. Each stem has three to five paired leaves with large indentations. The leaves have a very unpleasant odor.

FLOWERS. The flowers of coriander are formed in the shape of umbells containing three to six rays. Each ray in turn has smaller rays bearing five to thirteen flowers. The flowers are very small and uneven; the petals are white, or more often pink, sometimes yellow or violet. The marginal flowers have larger petals. The inner flowers are smaller. The petals fall off very soon and two fruits develop.

FRUIT. The fruit is commonly called seed. It has a spherical shape but with very pronounced sinuous ribs. The seed is hairless and its sum-

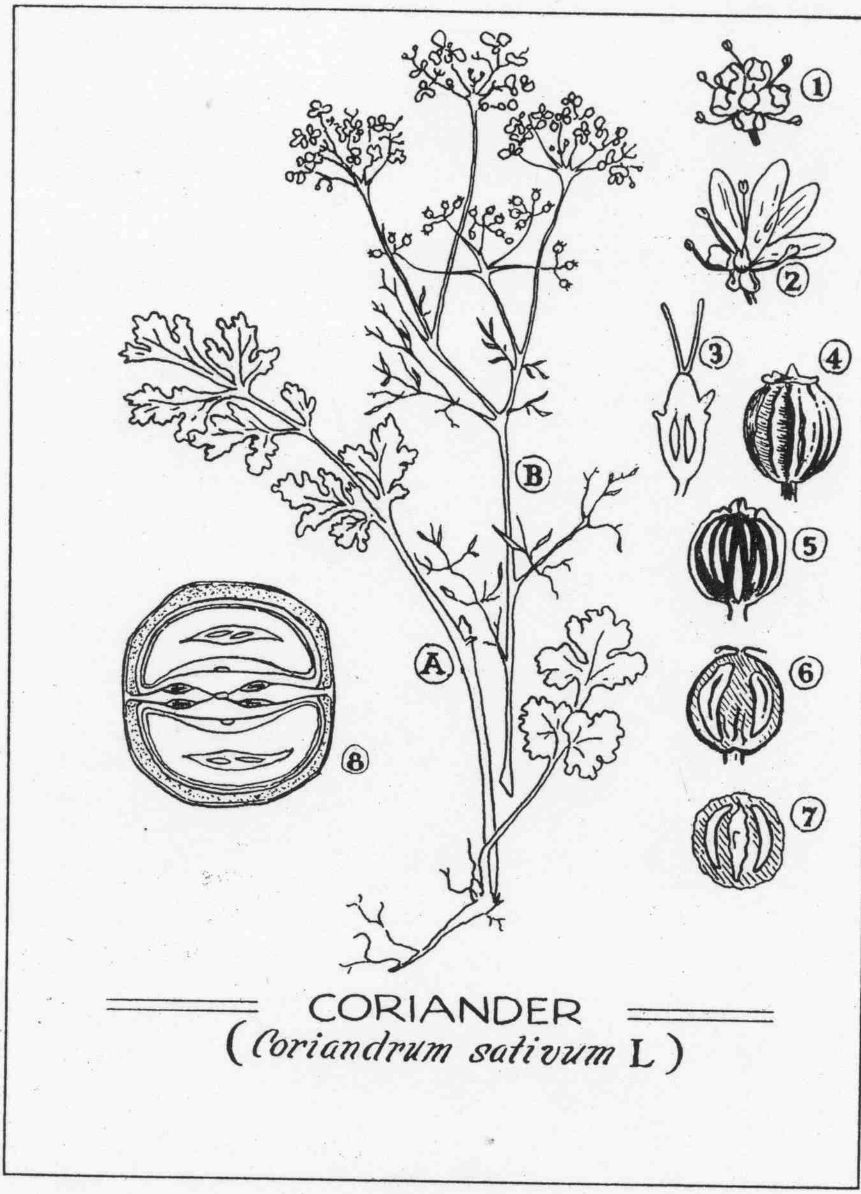


Figure 1

CORIANDER *(Coriandrum sativum L.)*

A. Leafy shoot
B. Inflorescences
1, 2. Flowers
3. Developing fruit
4, 5. Mature fruit
6. Longitudinal section of fruit
7. Transverse section of fruit
8. Transverse section of fruit (enlarged)

mit is covered with five calyx teeth and the remains of a short style. In the middle of the fruit there are two small columns tightly connected to each other in the shape of a cone. The fruit is composed of two halves or "little fruits" between which is a small cavity. The two carpels are connected to each other very tightly and can be separated only with difficulty. They are protected inside by ligneous pericarp. The unripe fruit possesses a strong disagreeable odor; for that reason it should be allowed to ripen fully before being gathered. When ripe and dry, it is sweet and fragrant.

Method of Cultivation

Coriander is a rather easily cultivated plant. It can grow in almost any soil except ground with swampy drainage. The best growth sites are considered to be either a black or rich limy soil. It requires sunny locations, as in the shade excess of humidity may affect adversely the health of the plant. Optimum growth of the plant may be accomplished either on fallow land or following perennial herbs in the practice of crop rotation. The plant does not thrive very well on soft and weeded lands. In a crop rotation, the best time for coriander is after any year-round plant, such as rye and wheat and also after such legumes as potatoes, sunflowers, etc. Very often coriander is sown together with caraway seed. The harvesting of caraway crops takes place every second year, while coriander is harvested every year, usually prior to caraway when there is a double crop. One year prior to the planting of coriander, the soil must be fertilized, but a different plant must be sown after fertilization and before the coriander crop, because increasing the fertility of the land immediately before planting a coriander crop would result in a profuse growth of leaves and stems and a delayed ripening of seed. This would not be desirable as commercially, the seed is the valuable part of the plant. Before planting, the soil is deeply plowed (4.8—7.8 inches) and harrowed early in the fall. If some undesirable herbs or weeds appear, it is necessary to harrow again. In early spring, as soon as the field dries, the soil must be harrowed again or even plowed deep (2.7—3 inches), depending on the amount of weeds present. The coriander seeds are sown in the quantity of 10 to 13 pounds per acre, if sown by rows 15—24 inches apart. Sowing by rows is the best practice because half the amount of seeds is required as compared with broadcast seeding. The former way results also in an easier elimination of weeds. Sowing is done at the end of April or at the beginning of May. The seeds used for sowing must be normally developed and have a normal moisture. Seeds are usually sown dry. Sowing is done after a slight drying in the air. Occasionally, to accelerate their sprouting, they are soaked in water for three or four days. As coriander seeds soon lose their sprouting ability, care must be taken not to use overmature seeds. The viability of seeds under normal conditions is from three to four years. However, the best age for sowing is two years.

Care

When the first sprouts appear, the crop is thinned; after that, the only care necessary is to weed out once or twice more until the plant is sufficiently strong to overcome any weed growths. In order to control the weeds it is recommended that some flax, hemp or sunflower seeds be added to the coriander seeds in the ratio of 1 pound to 8-10 pounds of coriander. As it takes a long time for coriander to sprout the weeds would have time to sprout, grow up and to suppress the sprouts of coriander. The presence of additional seeds will aid in determining where the coriander seeds are planted and thus in eliminating the weeds. As soon as the first sprouts of the plant appear, it is necessary to weed at once and then soon again. About a week after the second weeding the plants are strong enough to fight the weeds by spreading their own shadows over the weeds and thus guarantee their own free growth.

Harvesting

Harvesting commences at such time as when a color change from green to light yellow occurs in the stems. At the same time the seeds harden, change color from green to grey, and acquire a pleasant aromatic odor and sweet taste. Ripening is not simultaneous for all the seeds. A portion of the ripe seeds usually falls to the ground while the green seeds are retained on the plant. The harvesters, therefore, cannot wait until all the seeds are grey. As soon as about two-thirds of them are grey it is necessary to start harvesting. To avoid the period of greatest loss, because of dropping of the seed, harvesting takes place either early in the morning (before the dew disappears) or after sunset in the evening. The crop is harvested by combine and should not take more than two or three days. The mowed plants are threshed with threshing machines. Special care in threshing is necessary because the quality of the seed is determined in the main by its integrity. After being threshed, the seeds are cleaned and dried by dry air or special equipment. If the seeds are not well dried, they absorb heat too easily and lose their light color and pleasant taste. After they are dried, they are sieved and kept in a dry place.Crop yield is from 750 pounds to 2200 pounds per acre. During storage the crop must be guarded from insects. The main enemy of coriander is *Sitodrepa panicea L.* which destroys the inside of the seed. This insect multiplies very rapidly (one female lays 60 eggs) and destroys the entire seed. *Sitodrepa panicea L.* is an oval shaped bug, covered with thick brown hair, 2-3 mm. long. In the presence of danger it feigns death. There are other enemies of the coriander: *Tyroglyplup sp.* and *Sitotroga cerealella, oliv.* and *Tinea Granella, L.* To avoid these insects the storage must be dry, well ventilated and clean. The best disinfectant to use if these insects do appear is sulphur dioxide applied over a period of 12 to 16 hours in the ratio of 150 grams of sulphur dioxide per 1 cubic meter of storage space.

Distribution of Oil

In the cavity of the seed are small canals which are the main carriers of the oil. The fruit must be broken before being subjected to distillation.

Properties of Seeds

The ripe seeds are yellowish, two to four mm. in diameter, have a sweetish taste and a very strong, characteristic odor. Unripened seeds have a very unpleasant odor. Really good seeds must be dry, ripened, light greyish-yellow (very few dark ones); they must show no contamination, and have a characteristic odor (no molded odor). The moisture content of the seed should not exceed 12 per cent, ash content not more than 7 per cent (insoluble in hydrochloric acid), acidity from 1.3 to 2.0 mg. of normal potassium hydroxide per gram, and not more than 1% of essential oil. The seed should not be unripened, because although it has more oil in that state, the oil is of the worst quality. Experimentation at Seagram's on coriander seed grown in Jefferson County, Kentucky, showed that the unripened seed had 1.17% essential oil; ripened, 1.02%; and overripe, 0.77%. After the seed ripens, the amount of oil decreases, as is the case for most plants containing essential oils. There are 120 to 140 seeds per gram of coriander.

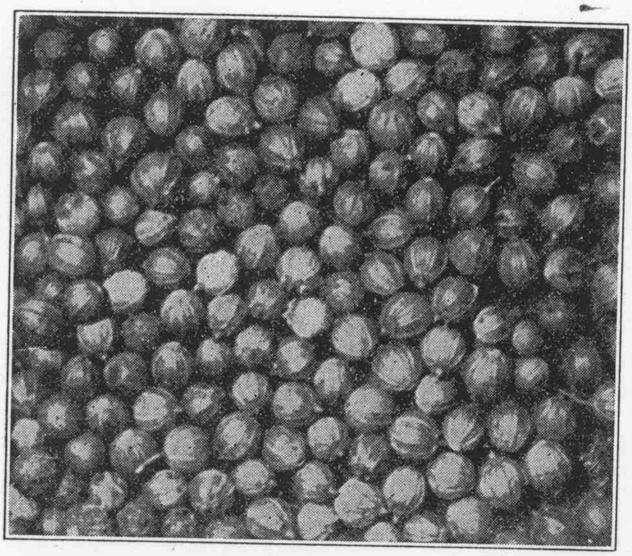


Figure 1a
CORIANDER SEED
(enlarged 2 X)

Properties of Oils

Physical Properties

Soluble in 70% alcohol 1:2 or 1:3
Sp. gr. at 15°C. 0.870 — 0.885
Opt. Rot. at 20°C. +8° to 15°
Refr. index at 20°C. 1.4569 to 1.4700

Chemical Composition

d–linalool, 60% to 80% and its acetic ester
d–alpha pinene
beta pinene
n–cymol
terpinolene
alpha and gamma terpinene
geraniol and acetic esters
1–borneol and acetic esters
n–decylic aldehyde
acetic and decylic acids
dipentene

Uses

The oil of coriander seed is used for flavoring foods such as: candies, canned foods, catsup, chili sauce, condiment sauces in general, and mayonnaise. It is used as a pickling agent; in salad dressings, soups, spice extractives, spice oils, tobacco products; for pharmaceutical purposes, as a source of linalol in perfumes, soaps; and for flavoring tobaccos, gins and cordials. In the Orient, coriander seed is used for flavoring foods, such as meats and breads, and as an ingredient of curry powder and some other condiments. After the extraction of essential oils about 20% butter fat is left in the seed which can be used in the soap and textile industry. After essential oils and butter fat have been removed the final by-product of the seed can be used as cattle feed because it contains protein to the extent of 11 to 17%.

Experimental cultivation of coriander seed was carried out in Jefferson County, Kentucky, by the authors. The land used had previously been in legume cultivation. The soil was plowed and harrowed 5 to 6 inches the first part of April and original Russian coriander seeds were planted 1.25 inches deep on April 22. They were sowed in rows 12 inches apart. The seed had a sprouting ability of 92%. On May 8 the first sprouting was noticed. The plants were weeded on May 15 and June 5. Blossoms appeared on June 20. The flowering was complete on July 10. The first green seed was harvested on July 20, properly ripened seed was collected on July 25, and the last portion was left to over-ripen and was collected on July 29. Seeds are properly ripened when two-thirds have changed from green to grey.

One hundred stems were measured and an average taken. The average length was 17.25 inches (shortest was 14.00 inches and longest, 21.50 inches). The average length of the roots was 2.92 inches (shortest was 1.75 inches and longest, 4.75 inches.) The yield obtained was 1100 pounds of seed per acre. The straw yield was about the same. The three grades of seeds were analyzed and the chemical analyses are reported next. Figure 1b shows coriander plants in blossom.

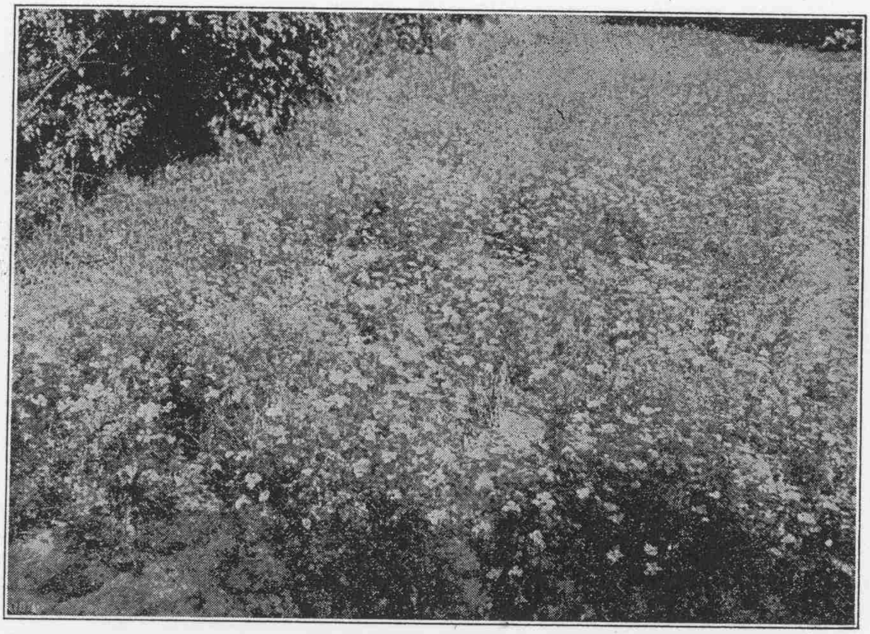


Figure 1b

The cultivation of coriander has also been done experimentally in Hancock County, Ohio, by Mr. J. J. Mergenthaler. Moroccan, Hungarian and Russian seeds were planted May 26, 1940 and the crop photographed on July 17, which is reproduced in Figure 1c. The Moroccan in the foreground of the picture grew to 3 inches in height; the Hungarian in the center to 4 inches; and the Russian in the background to 12 inches. The cultivation consisted of ordinary sowing in rows and the plants were

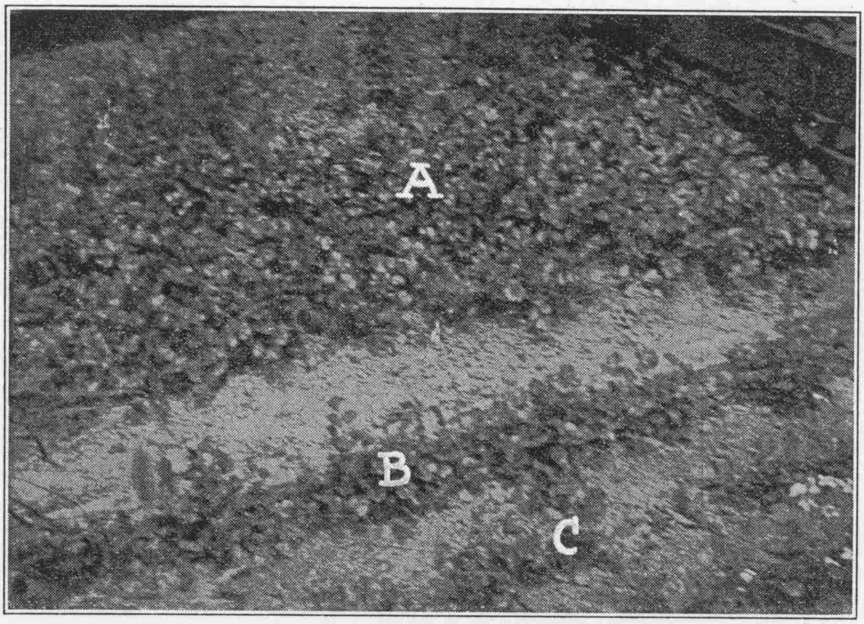


Figure 1c

weeded along with a truck garden near by. The Russian coriander was by far the best.

Successful experimentation with coriander has been carried out at Pewee Valley, Kentucky, by Mr. Clayton Stoess of Crestwood, Kentucky. This coriander is shown in Figure 1d.

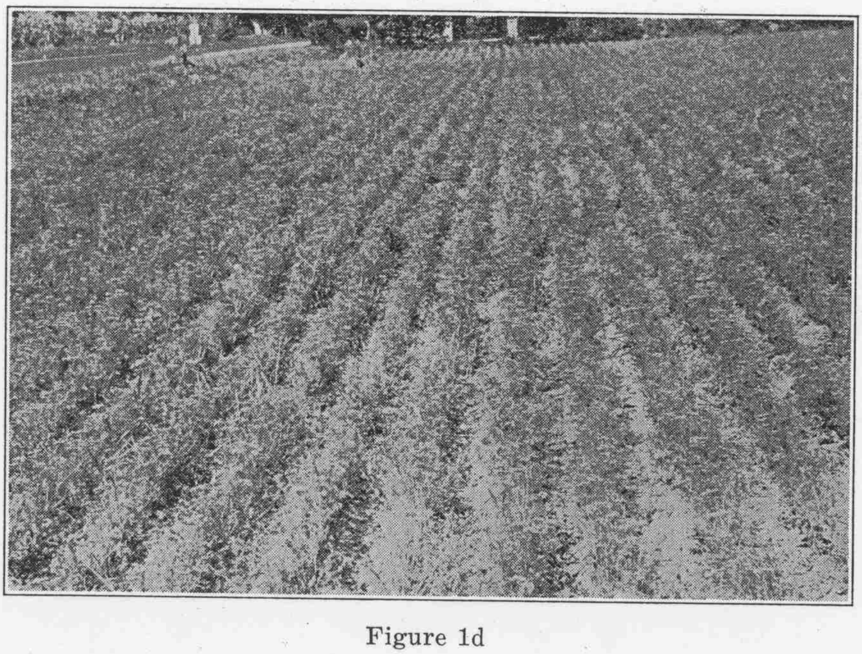


Figure 1d

Following are the chemical analyses on six different types of coriander seed as performed in the Seagram Research Department. The constant variation of the essential oil content from coriander of different origins

CORIANDER SEEDS ASSAYMENT

TABLE I

Experiment No.	491 (1) A.deG.	491 (2) A.deG.	491 (3) A.deG.	491 (4) A.deG.	491 (5) A.deG.	491 (6) A.deG.
Company						
Age	1940 crop	1939 crop	1939 crop	1940 crop	1940 crop	1940 crop
Amount represented by lot						
Country	Morocco	Hungary	Russia	U.S.A. Kentucky unripe	U.S.A. Kentucky ripe	U.S.A. Kentucky overripe
Moisture by desiccator	2.709%	2.788%	2.748%	3.202%	2.967%	2.810%
Moisture by oven	6.042%	6.510%	6.388%	7.662%	6.900%	6.690%
Total moisture	8.751%	9.298%	9.137%	10.864%	9.867%	9.500%
Moisture by Dean-Stark	....	....	....	....	....	....
Acidity as mg. KOH per gram	2.2%	1.75	1.33	2.45	2.14	2.00
Total ether extr. matter	0.2845 gms.	0.3145 gms.	0.3300 gms.	0.2430 gms.	0.2445 gms.	0.2465 gms.
Total volatile matter	0.0048 gms.	0.0170 gms.	0.0194 gms.	0.0221 gms.	0.0193 gms.	0.0146 gms.
Total non-volatile matter	0.2797 gms.	0.2975 gms.	0.3206 gms.	0.2209 gms.	0.2252 gms.	0.2329 gms.
% essential oil (dry basis)	0.25%	0.89%	0.98%	1.17%	1.02%	0.77%
% oil by Clevenger (dry basis)	0.25%	0.90%	1.00%	1.20%	1.0%	0.80%
Physical properties (Solub., Sp. Gr., Opt. Rot., Refr. Index)	1.4626	1.4640	1.4637	1.4635	1.4630	1.4645
Number per gram	67 seeds	143 seeds	116 seeds	157 seeds	161 seeds	152 seeds
% Contamination	....	....	....	....	....	....

is shown. There is also shown that definite times for harvesting the seeds will secure the best quality of essential oil. For a more complete picture of the quality of essential oils, the chemical analyses should be combined with a quality study, such as odor tests. A section following will discuss quality tests on these essential oils.

CARAWAY — *Carum carvi L.* — *Umbelliferae*

History

Caraway was found in the debris of Swiss Lake dwellings, and because of this origin, the plant has been considered a native of Europe. In the twelfth and thirteenth centuries it was grown in Morocco by the Arabs. From Asia, it was spread by Phoenician commerce to Western Europe. Distilled oil of caraway is first mentioned in the price ordinance of Berlin for 1574.

Regions of Cultivation

The plant is now widely distributed, and is found growing wild in Northern and middle Europe, Russia, Persia, Tibet, South Africa, Australia, and America. However, it is cultivated mostly in Europe, Holland, Scandinavia, Prussia, and Asia. The best caraway is grown in Holland.

Botanical Description

STEMS. Caraway is a biannual umbelliferae about two to three feet high.

ROOTS. The roots are small and sinuous.

LEAVES. The leaves are long, bipinnate, and subdivided into numerous segments, narrow-pointed and of a dark color.

FLOWERS. The flowers grow in compound umbels bearing small white flowers.

FRUIT. The fruit, commonly called seed, is laterally compressed and ovate. The seeds are naked, brown, striated; they have a hot, acrid, but pleasant taste. The seeds abound in essential oil containing gummy and resinous parts; they are situated in spindle-shaped cavities.

Method of Cultivation

The method of cultivation is very similar to that used for coriander. Caraway grows best on damp, flat ground. Exhausted or weeded lands are not suitable for caraway, just as in the case of coriander. The soil must be well prepared prior to seeding. It is usually sown after perennial herbs in the crop rotation system. The soil must be replowed after the last crop and then deeply replowed again in September. Early spring is the best planting time for caraway; however, it is also possible to sow in the early fall. In this case the caraway will have time to acquire the necessary strength before winter. Quite frequently this botanical is planted with coriander. The coriander matures earlier as it is an annual plant, and it is harvested before the caraway produces a flowering stem. The seeds of caraway are sown in drills 7 inches to 8 inches deep. The distance between drills must be 15 to 24 inches. Should this distance be increased, the quan-

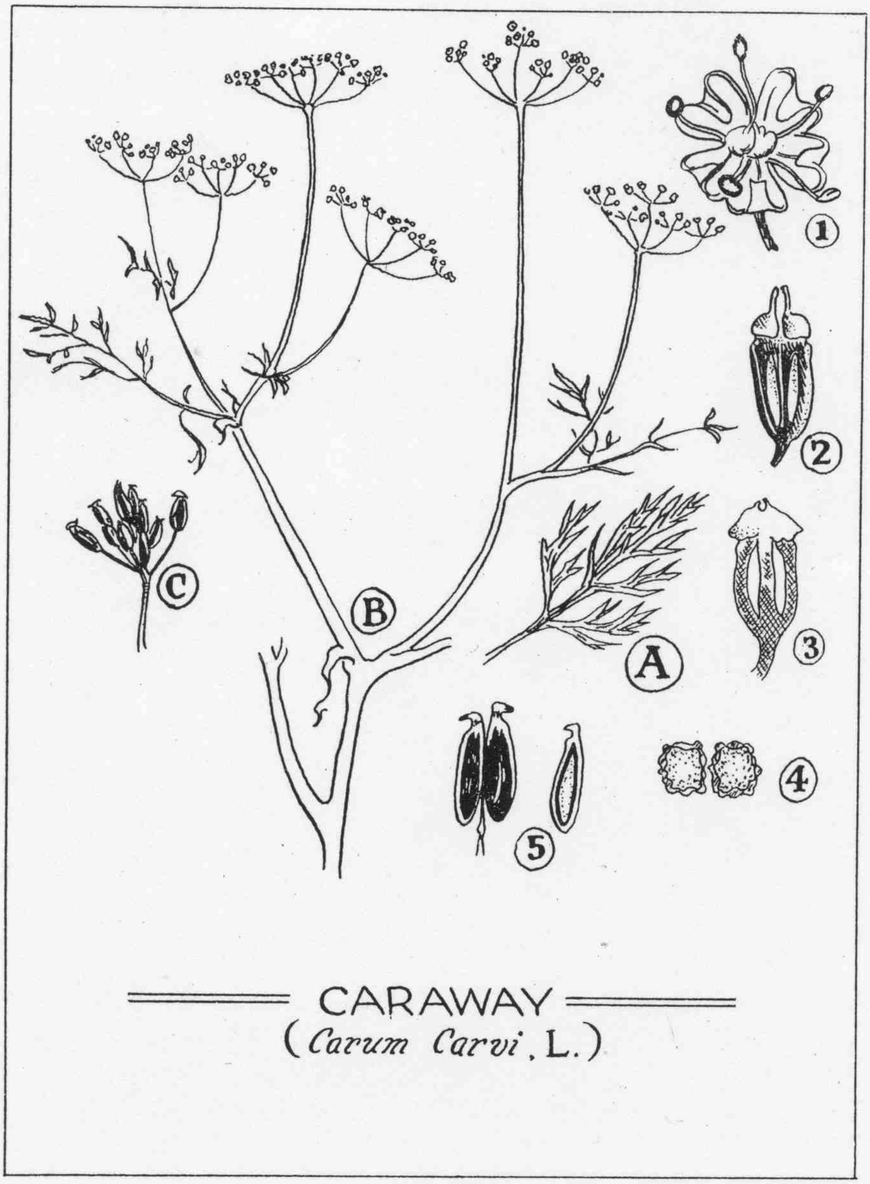


Figure 2

CARAWAY *(Carum Carvi L.)*

A. Leaf
B. Shoot bearing inflorescences
C. Fruit cluster
1. Flower
2. Immature fruit
3. Longitudinal section of fruit
4. Cross section of fruit
5. Mature fruit

tity of oil produced by the plant will be diminished. It requires 10.5 to 13 pounds of seed per acre for sowing. In the fall the seeds ripen and soon afterwards the new planting is done. The sprouts must be thinned out in the same manner as for coriander. Weeding must be practiced continually to insure a healthy crop. Cold exercises no effect on the caraway crop, whereas dryness does affect it to a slight degree.

Harvesting

Caraway is harvested prior to complete ripening (for the same reason as in the case of coriander) in the latter part of July, and in some instances, in August. It is cut before dawn in order to keep the natural flavor of the seed in the crop. Crop yield is from 900 pounds to 1100 pounds per acre.

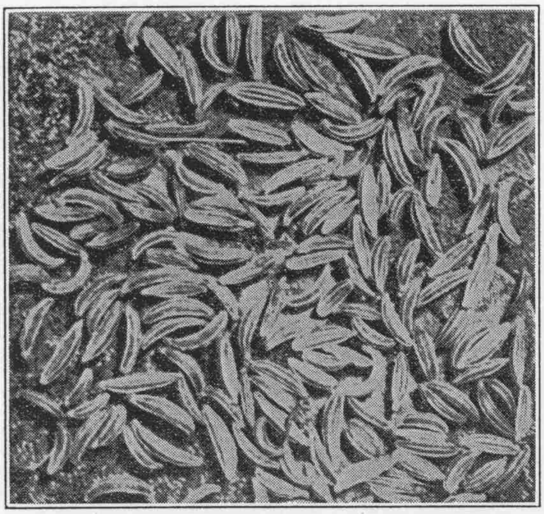


Figure 2a
CARAWAY SEED
(enlarged 2 X)

Distribution of Oil

Most of the oil is in the seeds. Its quantity increases with the ripening of the seed.

Properties of Seeds

The seeds must be well dried, of a brownish-grey color, aromatic, and without foreign matter. Moisture content should not exceed 10.0 per cent, ash content should be not over 9.0 per cent, oil content from 3.0 to 7.0 per cent. Wild caraway sometimes has 9.0 per cent oil. Acidity can be from 4.0 to 4.5 mg. of normal potassium hydroxide per gram. There are from 380 to 400 seeds per gram of caraway.

Properties of Oil

Physical Properties

Soluble in 90% alcohol 1:1
Sp. Gr. at 15°C. 0.907 to 0.920
Opt. Rot. at 20°C. +70° to +82°
Refr. index at 20°C.. 1.4885 to 1.4890

Chemical Composition

carovone 50% to 60%
d-limonene (or carvone)
carveol
dihydrocarveol

In unripe fruit the quantity of limonene is predominant; as the fruit ripens, limonene oxidizes into carvone.

Uses

Caraway is used in the food industry in the making of bread, cheese, salads, sauces, soups, and candy. In medicine the tincture of caraway oil is used as a stomachic and carminative. It also is employed in the manufacture of perfumes, soaps, liqueurs and gin. Besides the essential oil the caraway seed contains about 15% butter fat. The extraction of this fat leaves behind a valuable cattle feed and straw.

ANISE — *Pimpinella anisum L.* — *Umbelliferae*

History

Aniseed, as it is popularly called, is the dried ripe fruit of anise, indigenous to Egypt and to several of the Mediterranean islands. The ancients valued this plant highly for its flavor and its real, and supposedly

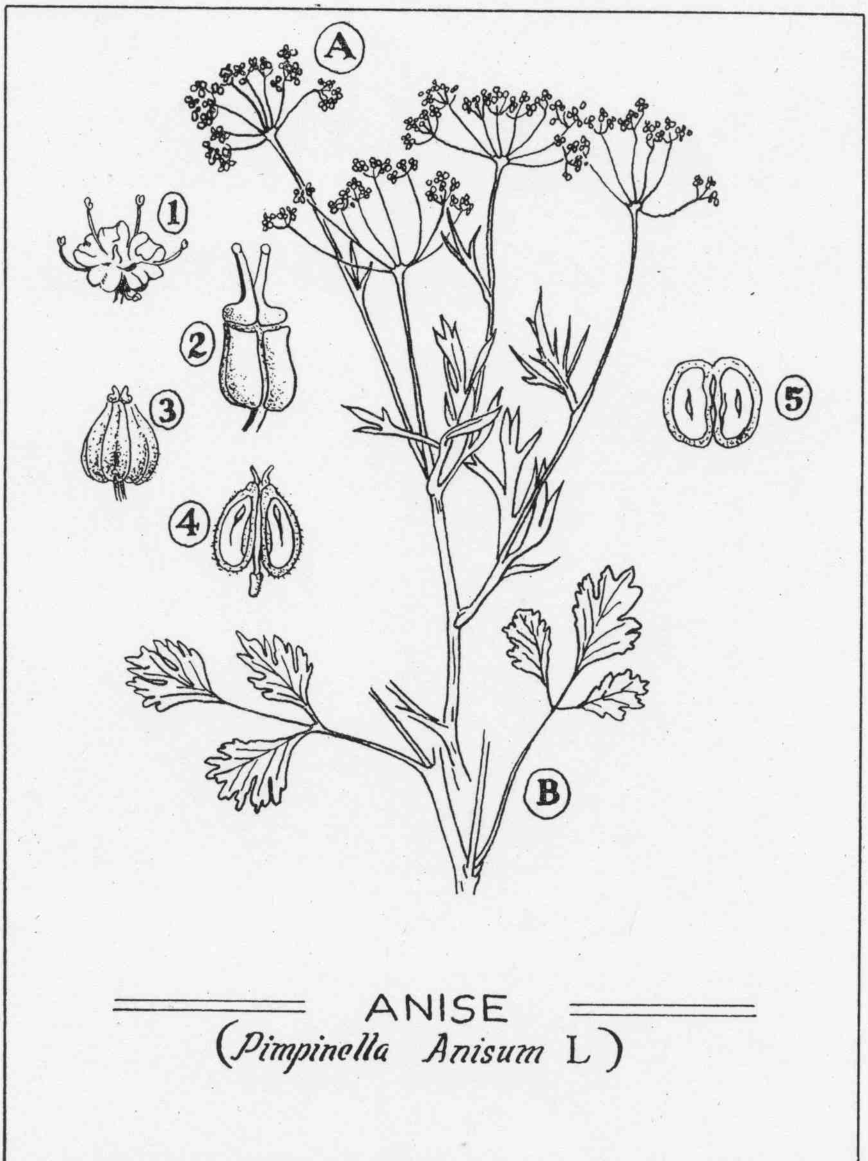


Figure 3

ANISE *(Pimpinella Anisum L.)*

A. Inflorescence
B. Leafy shoot
1. Flower
2. Immature fruit
3. Mature fruit
4. Longitudinal section of fruit
5. Cross-section of fruit

real, medicinal values. Pliny mentioned anise from Crete and referred to its effect as an appetizer. Aniseed has been a common constituent of cough mixtures for several centuries. The plant is known to have been cultivated in Spain as early as the twelfth century and in England at the beginning of the sixteenth century.

Regions of Cultivation

The plant is cultivated in southern Russia, Spain, France, Germany, Bulgaria, Turkey and other parts of Europe, and also in Syria, Tunis, India and Chile. The best anise comes from Russia, Italy and Spain. The per cent of essential oil content in aniseed from different countries is shown below:

Chile	1.9–2.6%
Syria	2.0–6.0%
Bulgaria	2.4%
Mexico	2.0–2.2%
Macedonia	2.2%
Russia	2.5–3.2%
Asia Minor	2.75%
Spain	3.0%
West Prussia	3.4%
Italy (Bologna)	3.5%
Germany	4.0–4.2%

Botanical Description

STEMS. Anise is an annual plant belonging to the natural order of Umbelliferae. It attains a height of about one foot or more under cultivation. The stems are round, hairy and resemble grass.

LEAVES. The leaves are of different shapes. The lower ones have thick stems, are few in number, triangular in shape, and deeply and irregularly toothed. The middle leaves likewise have thick stems and three large teeth, whereas the upper leaves do not have any stem, are narrow and long, and are composed of from three to five segments.

FLOWERS. Anise has yellowish-white flowers in large, loose, compound umbels. The flowers are very small, with hanging bells composed of five small petals. They form flat umbels with six to ten branches or rays. At the end of each ray there are smaller umbellets, each containing four to nine flowers. The flowers later form the fruit.

FRUIT. The fruit, commonly called seed, has two united mericarps with the fruit stalks attached together. The fruit is ovoid in shape, tapering to a point, and varying in length from 2 mm. to 5 mm. At the anterior end of the seed is found a small, disc-like cover containing bent stalks which are in the form of a hook. The ripe fruit is of a greenish-grey color and possesses barbels. It has a sweetish taste and a characteristically pungent odor.

Method of Cultivation

Anise thrives best in a rich, black limy with humus soil, free from weeds, in a rather warm dry climate. Because the vegetation period is long (130-140 days) anise is sown earlier than all other plants. This is also the reason for its cultivation in warm climates. Its most usual method of growth is directly from the seed. Seeding may be accomplished by broadcast or by rows. Sowing should be done the second year after fertilization, following either wheat or rye. During the fall, the soil is further improved by deep furrowing. Sowing is done in the spring, as early as possible after a second shallow ploughing (5 to 7 inches) and harrowing. The usual distance between rows is from 15 to 24 inches. The amount sown depends on the purity of seed, usually being from 10.5 to 12.5 pounds of seed per acre. During the summer at least two weedings are necessary to eliminate the competitive and undesirable weeds. If the sprouts are thick, it is necessary to thin them out so that there will be a space of about 6 inches between the plants in rows.

Harvesting

The harvesting season comes during the end of July or the beginning of August when the stems begin to yellow and the fruits in the middle part of the umbel take on a greenish-grey hue. Harvesting is best accomplished by a method similar to that of coriander. The seeds must be well protected from moisture and rain; otherwise they would darken. Crop yield is from 1200 to 2400 pounds per acre.

Distribution of Oil

The commercially important part of the plant is the seed which contains the aromatic oil.

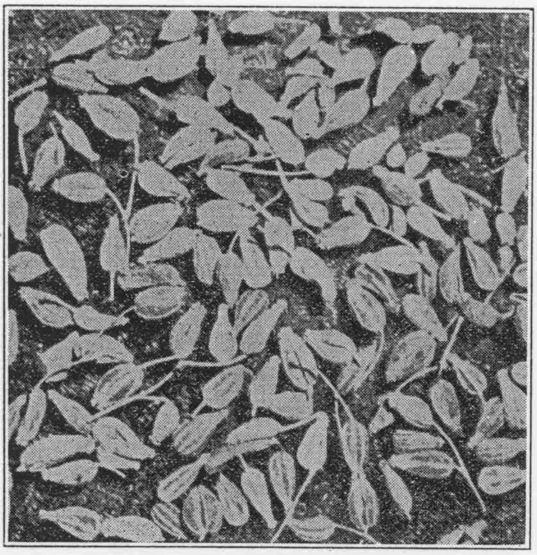


Figure 3a
ANISE SEED
(enlarged 2 X)

Properties of Seeds

Ready-for-market fruits should be greenish-grey, well cleaned, with a characteristic smell (no moldy odor), and no black seed content. The seeds must be very well dried and should be kept in covered buildings until they are ready to be sold. An acceptable moisture content is from 10% to 14%. Ash content should be not over 9%, with 1.5% insoluble in hydrochloric acid. Acidity must be between 6 mg. and 6.5 mg. normal potassium hydroxide per gram. Oil content shall be from 2.0 to 3.5%. According to its purity, anise is divided into two kinds. One kind must not have any foreign material higher than 4% (such as hemlock, a very poisonous plant which frequently contaminates the Italian aniseed), and not more than 1% sand and earthy material. The other kind must have not over 6% of foreign material. Coriander, which usually grows near anise and has a similar color, must not be present in either kind more than 2%. There are from 300 to 350 seeds per gram of anise.

Besides essential oil in fruit, fatty acids (16 to 22% dry), sugar, anise acid, mineral salts, proteins (16 to 22%) are found.

Properties of Oil

Physical Properties

Soluble in 90% alcohol 1:3 to 1:5
Sp. Gr. at 15° C. 0.975 to 0.990
Opt. Tor. at 20°C. 0° to —2°
Refr. index at 20°C. 1.5530 to 1.5600

Chemical Composition

fenchone 1%
anethole 80 to 90%
anise aldehyde
anise ketone of almond-like flower
anise acid

Uses

Culinary uses of anise oil relate to pastries, candies, fruit preserves, pickles, spice oils, and canned foods such as soups and meats. As for pharmaceuticals, it is used in cough mixtures and in bactericides. Anise fat is used in mild expectorants. Anise oil is used to flavor tobacco products. Anise fruit is employed in the manufacture of gins, cordials, and apéritives. The oil having been extracted, the dried anise seeds may be used as cattle and chicken feed.

FENNEL — *Foeniculum officinale L.* — *Umbelliferae*

History

The herb, fennel, was known to the ancient Chinese and Egyptians, and was used by them for culinary and medicinal purposes. It was highly esteemed by the ancient Saxons and Greeks. The former believed it to be one of the nine sacred herbs which had power against the nine venoms, or causes of all disease. In the eighteenth century, fennel was used as an ingredient for cure of gallstone.

Regions of Cultivation

The plant grows almost anywhere, particularly in Spain, southern France, Macedonia, Moravia, southern Russia, Rumania, Saxony, Galicia, south Africa, etc., but it grows best in sunny localities on well-drained soil. The best soils for fennel are black, sandy, and sandy-clay soils with sufficient lime, not too damp (too much moisture develops commercially undesirable parts of the plant.)

Botanical Description

Fennel is a biennial or perennial grass-like plant, belonging to the family Umbelliferae.

STEM. Its stem is round, of a bluish shade, with very fine striations, inverted toward the bottom, and lacking hair, and grows more than six feet high.

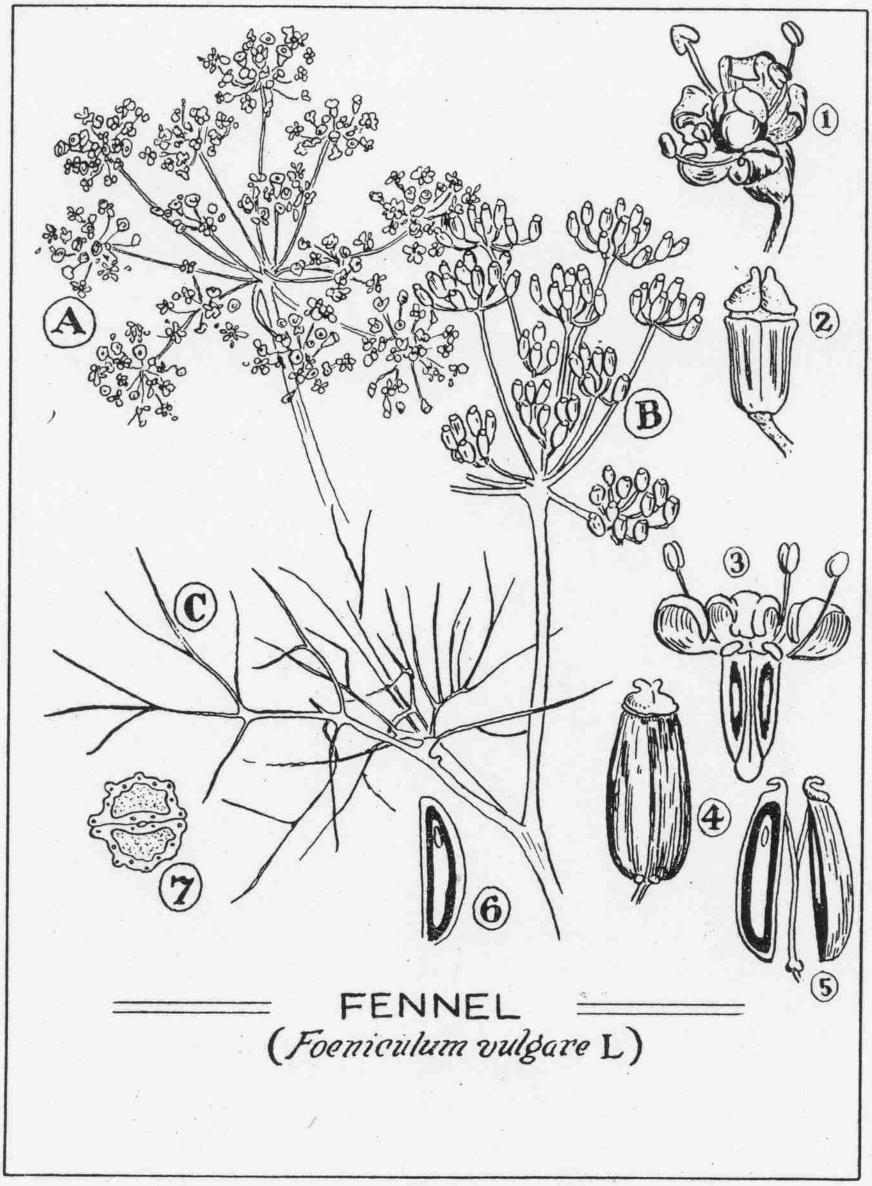


Figure 4

FENNEL (*Foeniculum vulgare L.*)

A. Inflorescence
B. Fruit cluster
C. Leaves
1. Flower
2. Immature fruit
3. Longitudinal section of flower
4. Mature fruit
5. Separated carpels
6. Longitudinal section of single mature carpel
7. Cross-section of fruit

ROOT. It has a fleshy, striated root, only slightly branched.

LEAVES. It has finely divided, narrow leaves, almost threadlike in appearance.

FLOWERS. Fennel possesses compound umbels of yellow flowers carrying 10-20 rays, each ray ending with a smaller umbel. The flowers are small and contain five petals. It flowers in July. After the petals fall off, the fruit develops.

FRUIT. The fruit, or seed, develops from two mericarps tightly connected. The fruit is roughly oval-shaped, and varies in length from 4-10 mm. Each half of the fruit exhibits five primary prominent ridges. The fruit is greenish in color and sweet to the taste.

Methods of Cultivation

Fennel is usually cultivated during the colder seasons of the year. Only in periods of extreme coldness, the dormant seed must be protected from frost and other detrimental climatic conditions by means of a protective layer of fallen leaves or a remaining stem-cutting of the previous crop. Similar to coriander, a high fertility of the soil may result in a vigorous growth of leaves and stems. The aim of the farmer being to have an optimum seed production rather than a profuse growth of other parts of the plant, the usual method of cultivation consists of a fertilization of the soil for a previous crop. Fennel is sown early in the fall, either broadcast or in rows. It is usually sown in shallow drills, about 1 inch deep and 24 to 28 inches apart. The sprouts are thinned out to a distance of 15 inches. Later care consists in weeding. The plant is usually raised directly through seeding; however, it also may be propagated by means of root plantings. Fennel requires 7 to 9 pounds per acre for planting.

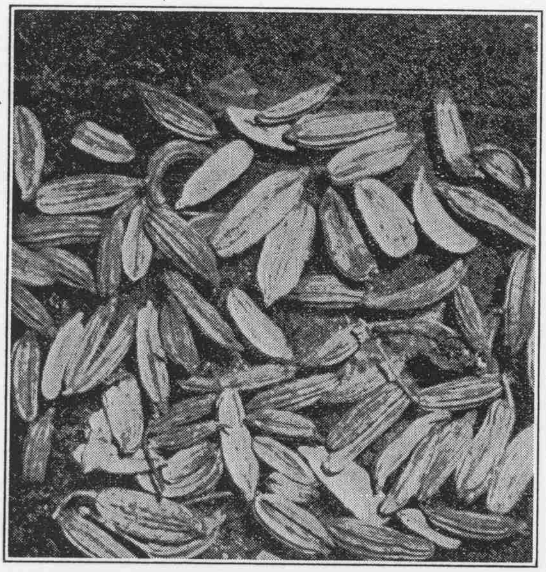


Figure 4a

FENNEL SEED
(enlarged 2 X)

Harvesting

The harvesting takes place when the seeds are sufficiently hard and acquire a greenish-grey color. Since the ripening period of the seeds is not uniform, it is better to harvest the crop in two stages. At first only the middle part of the umbel is cut, later the remainder is harvested. The plants are then piled in heaps, and after three to four days are threshed. The threshed seeds are sifted and then dried before they are placed in bags. The crop yield is from 1000 to 1400 pounds per acre.

Distribution of Oil

The fruit contains an aromatic oil similar to anise oil.

Properties of Seeds

Ready-for-market goods must consist of dry, ripe fruit, of a light greenish-gray color; it must contain no foreign seeds or soil, and very few yellow seeds. The moisture content must be 12 per cent; total ash should not exceed 8 to 10 per cent; the insoluble matter in hydrochloric acid should not exceed 2 per cent; acidity should be from 6 to 6.5 mg. of normal potassium hydroxide per gram; the seeds should contain 4 to 6 per cent oil. After extraction the dried seed can yield 12 to 18½ per cent butter fat and 14 to 22 per cent proteins. There are 120 to 130 seeds of fennel per gram.

Properties of Oil

Physical Properties

Soluble in 80% alcohol 1:5 or 1:8
Soluble in 90% alcohol 1:5 or 1:7
Sp. Gr. at 15°C. 0.860 to 0.995
Opt. Rot. at 20°C. +6° to +24°
Refr. index at 20°C. 1.5250 to 1.5500

Chemical Composition

50 to 60% anethole
fenchone
dipentene
alpha–pinene
delta–pinene
camphene

Commercially, two kinds of fennel oil are distinguished: i.e., "Sweet" and "Bitter" fennel oils. The former has a flavor which closely resembles that of anise oil. The bitter oil resembles turpentine. The chief constituent of the sweet oil is anethole (60%) and fenchone (10-15%). Bitter oil has less anethole and more fenchone. Both kinds contain the terpenes alpha and delta pinene, camphene, dipentene, etc.

Uses

Fennel is used in the manufacture of culinary products such as bakery goods, candies, canned foods, cheese, condiments, meats, fruit preserves, pickles and spice oils. In medicinal use, the United States Pharmacopoeia recognizes as official only the fruit of *Foeniculum vulgare* Miller; however, there are other varieties of fennel. For flavoring purposes, the commercial variety known as "French Sweet Fennel" is preferred. Fennel is also used in the manufacture of perfumes, gin, and cordials. After the essential oil has been extracted, fennel seed can be utilized as cattle and chicken feed.

ANGELICA — *Archangelica officinalis Hoffm.* — *Umbelliferae*

History

Angelica was first used as a spice during the fifteenth century. Its use for the preparation of distilled angelica water was described in 1500. In Great Britain it was once to be found in every herb garden, but today it is little cultivated there. The plant is said to be native to Syria, but has now spread to many cold European countries, especially to Lapland and the Alps, and to Asia. It grows in humid places, on stagnant pools, lakes and rivers.

Regions of Cultivation

Angelica is now grown commercially in Germany (Thuringia and Saxony), in England, Poland, Russia, Belgium, Italy and France.

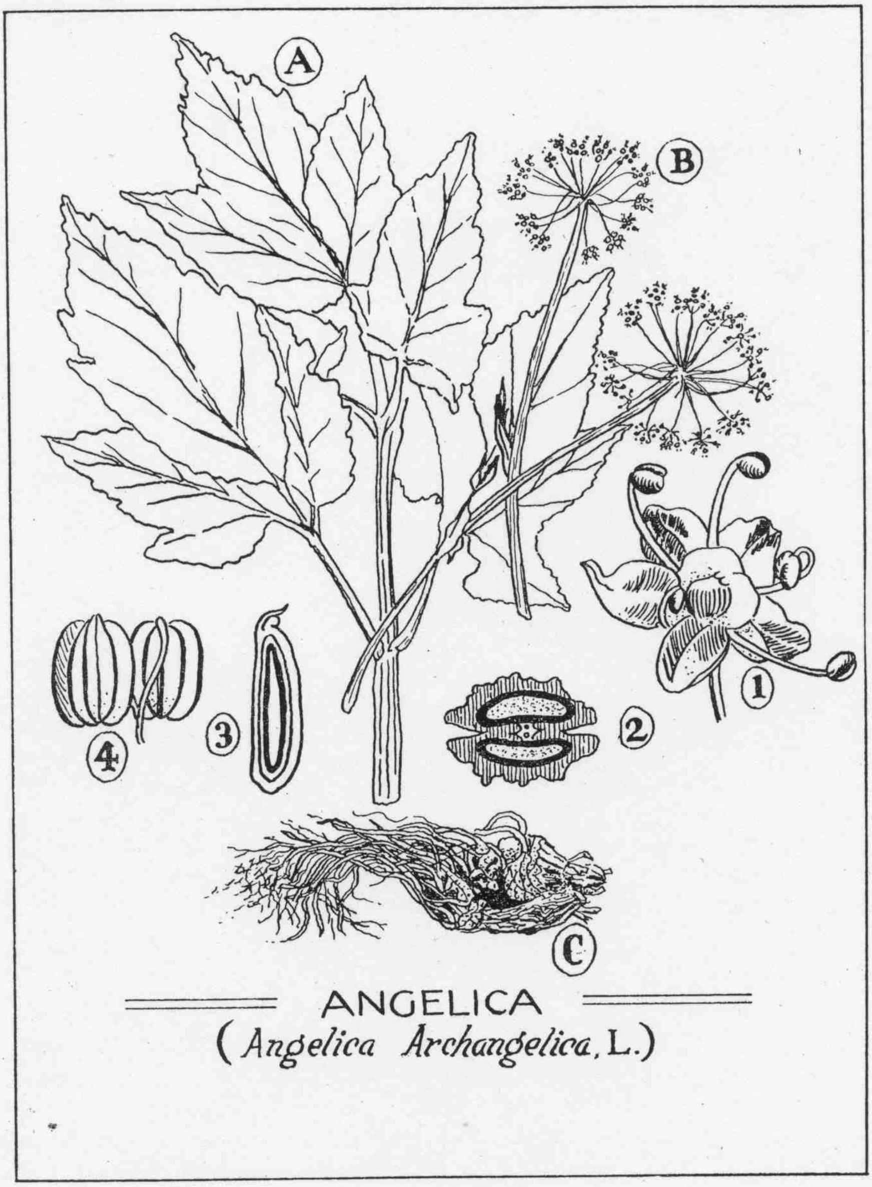


Figure 5

ANGELICA *(Angelica Archangelica L.)*

A. Leafy shoot
B. Inflorescence
C. Rhizome and roots
1. Flower
2. Cross-section of fruit
3. Mature carpel showing single seed
4. Mature fruits

Botanical Description

The plant is a typical Umbelliferae. There are two species of angelica: (1) *A. officinalis Hoffm.*, and (2) *A. silvestris*, or common, both being biennial.

(1) *A. officinalis Hoffm.*

STEMS. The stem of *A. officinalis Hoffm.* is tall and sturdy.

ROOTS.[1] The root is very well developed and has many rootlets.

LEAVES. The leaves are very large and bipinnate.

FLOWERS. The flowers are composed of large compound umbels of small greenish flowers.

FRUIT. Fruit, called seeds, are 6-7 mm. in diameter, greenish in color, with five visible striations.

(2) *A. silvestris*

A. silvestris has darker leaves; its flowers are white and bloom later in the year.

Method of Cultivation

Angelica can be raised by seeding in the usual manner, but germination is apt to be slow and irregular. It is best to store the seeds in moist sand several weeks before sowing. Seeds are sown in rows 11.8 inches apart, with a distance of 24 to 28 inches between the rows. Sowing may also be accomplished through the use of small pieces of root, in which case the distance between rows should be 6 to 19 inches. In order to insure better results, the roots are transplanted after the first year of growth at a distance of 16 to 35 inches between rows, or even 16 to 47 inches. Turning and layering of the top soil is the practice used to prevent weed growth. The seeds are not damaged by this procedure because they are deeply planted (7 to 8 inches.) In order to produce a more aromatic root, the tops of the plant are cut off to prevent formation of seeds. Angelica grows best in a moderate climate, in good soil, preferably with a deep layer of moist clay. Angelica requires 8 to 11 pounds of seed per acre for planting.

Harvesting

Harvesting seasons are the fall (September, October) and the spring (April). After one year of growth, the root is carefully harvested, washed, and dried by air. It is advisable to keep the roots ready for sale in closed containers to protect them against loss of aroma and also against insects. The crop yield is from 900 to 1300 pounds per acre.

Distribution of Oil

Essential oil is distributed throughout the whole plant, being found in the seeds, leaves, stems, and roots. Commercially, the important parts are the roots, then seeds and stems.

Properties of Root

Both cut and whole angelica are found on the market. Normal angelica root which is ready for sale must be one year old, from the spring

[1] Throughout the description of angelica, the rhizome is referred to as root, according to commercial terminology.

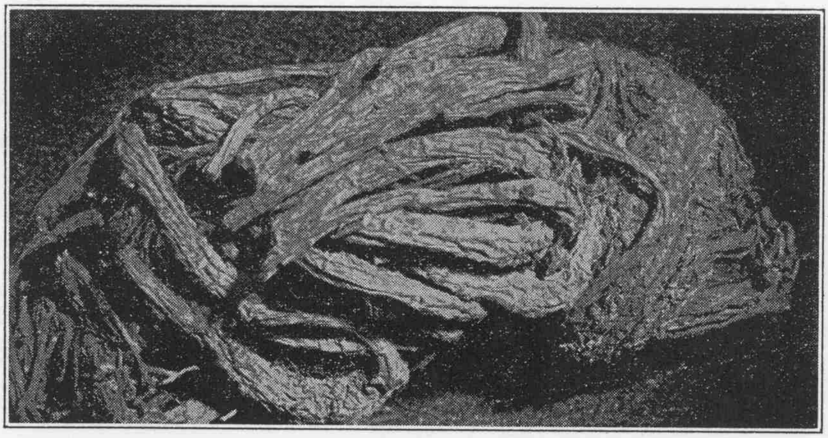


Figure 5a
ANGELICA ROOT

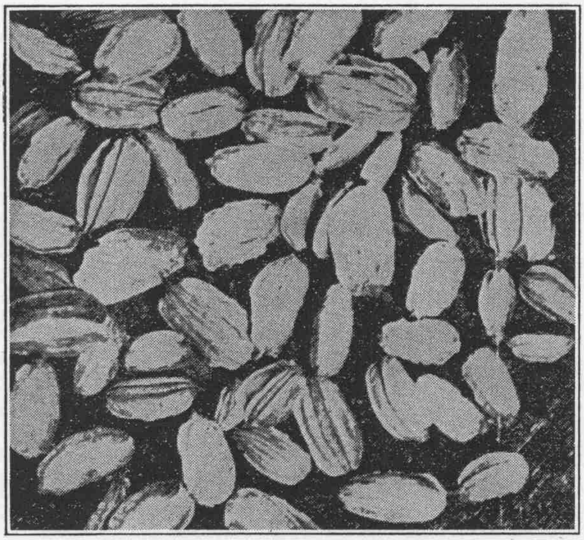


Figure 5b
ANGELICA SEEDS
(enlarged 2 X)

harvest, whole, twisted, with the percentage of tared dust not exceeding 2 per cent. Total moisture should not exceed 10 per cent. Acidity must be less than 5 mg. normal potassium hydroxide per gram. The percentage of oil must be from 0.5 to 1.0 per cent. Angelica has a very strong, musky odor.

Properties of Seed

The seeds are oblong, round at the ends, and whitish in appearance. They contain about 1 per cent of oil. There are from 210 to 240 seeds per gram of angelica.

Properties of Oil

Physical Properties

	Root Oil	*Seed Oil*
Soluble in 90% ethyl alcohol	1:2–1:3	1:5
Sp. gr. at 15°C.	0.857 to 0.918	0.851 to 0.890
Opt. rot. at 20°C.	+16° to +32°	+11° to +13°
Refr. index at 20°C.	1.4770 to 1.4880	1.484 to 1.491

The oils have pleasant spicy odors resembling the smell of oil of pepper. The diluted root oil develops a very agreeable musky aroma.

Chemical Composition

Root Oil	*Seed Oil*
esters of oxy-penta-decylic acid	d–phellandrene
d–phellandrene	esters of valerianic acid
esters of valerianic acid	esters of oxymyristic acid
pinene	
angelicine	
angelic acid	

The lactone of oxy-penta-decylic acid seems to account for the musky odor of root oil.

Uses

The roots, young stems, and leaves are steeped in syrups to make candied angelica. The seeds are used for the flavoring of beverages, cakes, and candies. It is used in medicine as a tonic for digestion. The essential oil is used in perfumes, for flavoring bitters, certain liqueurs such as Chartreuse, some varieties of Vermouth, and gin.

LICORICE — *Glycyrrhiza glabra L.* — *Leguminosae*

Regions of Cultivation

Licorice grows wild in Italy, Spain, southern France, Sicily, Turkey and Russia. It is cultivated in Germany. In the latter part of the nineteenth century successful attempts at cultivation in the United States were made at Sarpy, near New Orleans, Louisiana, and at Riverside, California.

Botanical Description

STEMS. It is a perennial plant with several grey fuzzy tall stems, slightly sinuous.

ROOTS.[1] The roots are heavy and short, have many rootlets which grow deep into the soil and sometimes spread out over a large area.

LEAVES. Leaves are prolonged and oval.

FLOWERS. Flowers, which are of a white-violet color, are collected in moderately long clusters. The plant flowers from July until August.

Method of Cultivation

A suitable soil for licorice is clay. The plant is cultivated by sowing the seeds or planting roots too small to be sold. Roots are cut in pieces 9 to 12 inches long and are planted in furrows spaced at 23.6 inches. The roots are placed at a distance of 18 to 24 inches from each other in the furrows. In order to have a good supply of roots, it is advisable to have a nursery where the roots are planted closely to each other, just far enough apart to develop sprouts. This is accomplished in the spring, the soil having been deeply ploughed and well fertilized in the previous fall. Licorice does not suffer from dryness nor from severe cold. It needs little care except for an occasional thinning. Licorice requires 8 to 10 pounds of seed per acre for planting.

Harvesting

After three or four years the first crop is collected. The difficulty in removing the roots *in toto* is considered among gardeners as the ultimate in gardening art. The roots have to be removed without the slightest damage. If a portion of the root is left in the soil, an eventual establishment of a plant may take place and may be detrimental to future crops other than licorice. In the event of a continuous planting of licorice for a single type

[1] Throughout the description of licorice, the rhizome is referred to as the root, according to commercial terminology.

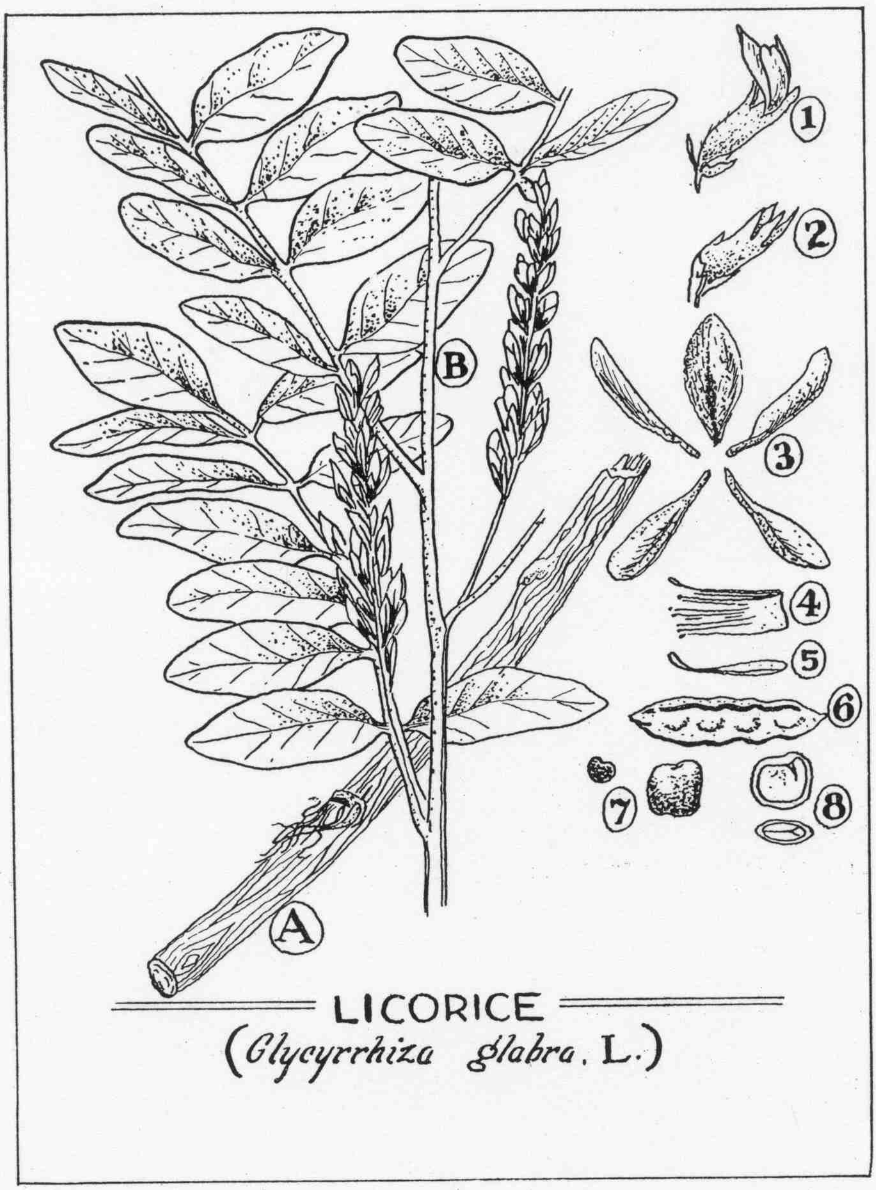


Figure 6

LICORICE *(Glycyrrhiza glabra L.)*

A. Rhizome with roots

B. Leafy stem with inflorescences

1. Flower
2. Calyx
3. Petals
4. Stamens
5. Carpel
6. Mature fruit
7. Seeds
8. Sections of seeds

crop, no harm can come to the next year's crop from leftover roots. Should the farmer, however, desire to plant a different crop, ensuing difficulties in the way of root growth of licorice can readily be imagined. For that reason, licorice usually is planted four to six times in succession in the same place, so that one field is occupied by it for twenty or twenty-five years. Licorice is harvested at different times in different regions. In southern Europe, in places where there is no snow, harvesting can be done even in the winter, so long as the roots are not affected. Harvesting may take place at any time between April until October. The roots are carefully freed from soil and are temporarily placed in piles in such a manner that air can reach all the roots. The roots should be re-stacked every once in a while so that every part of the root will have been exposed to the air. Ten or fifteen days after being harvested, the roots are soaked in cold water, then cut in pieces 12 to 14 inches long, and cleaned. The cleaning process consists in removing both soil and epidermis, or, in some cases, just the soil. The roots then are well dried in ovens at 86° to 104° F. Crop yield of licorice root is 850 to 3360 pounds per acre.

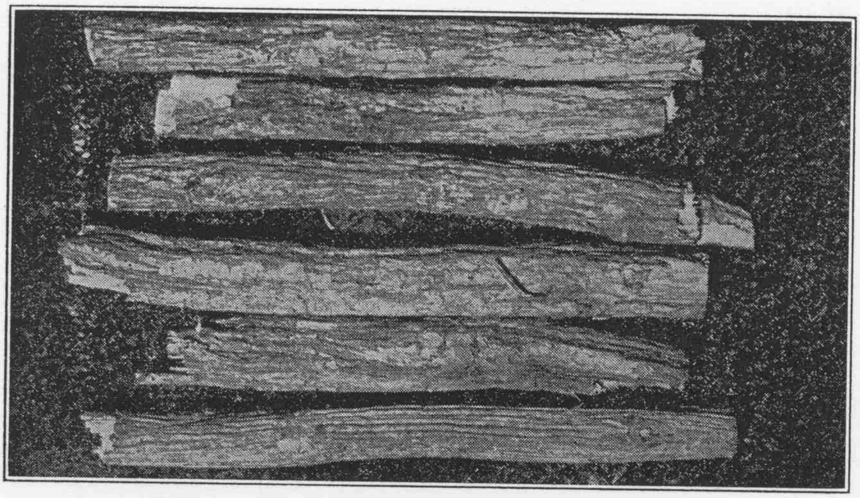


Figure 6a

LICORICE ROOTS
(natural size)

Distribution of Oil

Commercially, the important part of the plant is the root, which has a sweet, irritating taste because of the presence of the essential oil.

Properties of Root

There are three kinds of commercial licorice: (1) uncleaned (simply soil cleansed); (2) cleaned (epidermis, root hair, scrapings, and soil removed); (3) doubly cleaned (remainder of epidermis scraped off with special knives so that a lemon-yellow shade is reached). Ready-for-market goods must be in (1), simply cleaned of soil; in (2), of light color and containing no foreign matter such as *G. echinata L.;* in (3), 14 inches long, thick, without dark spots or trace of epidermic layer, of lemon-yellow color, completely free of foreign matter.

In all cases the roots must be well-dried, have no odor, and have a slightly acrid, sweet taste. They must contain not more than 10 per cent moisture, have 2 to 3½ mg. of normal potassium hydroxide per gram acidity, 0.03 to 0.05 per cent volatile oil, and not less than 5 per cent glycyrrhizine.

Properties of Oil

Physical Properties

Soluble (60%) in cold water

Chemical Composition

glycyrrhizine	starch
asparagine	proteins

Uses

Licorice is used in the manufacture of candies and chewing gums; in medicinal materials such as laxatives and cough mixtures; for flavoring tobacco products; and in the beer and liquor industries. In addition, licorice is a stabilizer of the foam in some fire extinguishers. The licorice pulp is used in insulation board.

II

THE ROLE OF THE AGRICULTURAL ENGINEER IN THE PRODUCTION OF ESSENTIAL OILS

by PAUL J. KOLACHOV

Director of Research
Joseph E. Seagram and Sons, Inc.
Louisville, Kentucky

II.

The Role of the Agricultural Engineer in the Production of Essential Oils[1]

by DR. PAUL KOLACHOV

The growth of plants for the production of essential oils has been confined strictly to foreign nations. These nations over a period of many years have developed the planting, cultivating, harvesting, and processing of aromatic plants until their industries not only supplied growing world demand by gradual expansion, but also vied with each other on the basis that their product was superior. Both place of growth and methods of processing supposedly had contributed to this so-called superiority of product. The industry, in fact, was an art.

With the application of scientific methods to agriculture, it has been learned that results obtained in any one place can be reproduced and ofttimes bettered by selecting proper locations and controlling growth conditions. This is true in the planting of peanuts, grain, fruits, trees, flowers, and certainly is true for other plants. Investigations conducted by the author at the distillery of Joseph E. Seagram and Sons, Incorporated, on the growth of various aromatic plants, have definitely indicated that better grades of oils are obtained under controlled conditions.

This paper proposes to show where and how aromatic plants can be grown in the United States, and how they can be processed to supply the American market demand for essential oils. In order to limit the length of the article, there have been chosen for discussion six species of aromatic plants which have been studied in order to show in detail the role of an agricultural engineer in their cultivation. The plants chosen are:

Coriander (Coriandrum sativum, L.)
Caraway (Carum carvi, L.)
Anise (Pimpinella anisum, L.)
Fennel (Foeniculum vulgare, L.)
Angelica (Angelica archangelica, L.)
Licorice (Glycyrrhiza glabra, L.)

The chart (Figure 1) shows the regions of the natural growth of these six selected plants, and their cultivation throughout the world. Strangely enough, as will be noted, there is no cultivation of these plants in the United States, although this country consumes a great amount of aromatic raw material. That is why this article shall stress the introduction of aromatic plant culture in the United States, and shall indicate the role the agricultural engineer will play in the essential oil industry.

The selection of a proper locality is very important in starting a plantation for the growth of aromatic raw materials. However, the various aromatic plants require such different conditions for their growth that

[1] From lecture delivered before American Society of Agricultural Engineers, Power and Machinery Division, Pennsylvania State College, June 18, 1940.

GEOGRAPHICAL DISTRIBUTION OF AROMATIC PLANTS USED IN THE EXTRACTION OF ESSENTIAL OILS

(1) CORIANDER		(2) CARAWAY		(3) ANISE	
Wild	Cultivated	Wild	Cultivated	Wild	Cultivated
Italy France Asia Minor S. Russia Turkestan India Nepal	Russia Moravia England Africa N.W. Italy Germany Holland Mexico Persia Syria Palestine	Asia Persia All of Russia Africa Australia N. & C. Europe	Holland Russia Germany Scandinavia France England W. Asia	Asia Minor	W. Europe S. Russia Spain England France Germany Bulgaria Cyprus India Turkey Chile Syria

(4) FENNEL		(5) ANGELICA		(6) LICORICE	
Wild	Cultivated	Wild	Cultivated	Wild	Cultivated
Spain France Rumania Saxony N. America W. Asia S. Russia S. Africa	S. France Germany E. Europe Parts of Asia	Lapland Alps Asia	E. Germany England Belgium Italy France Hungary	S. Russia Turkey Italy S. France Sicily Spain Syria	Germany

Figure 1

it is impossible to give any specific rules that would be applicable to all such plants. It may be said, in general, that the best location is flat country having small hills, since the mechanical processes of cultivation can be most easily handled on flat ground.

One of the first conditions for the cultivation of any aromatic botanical is a fertile soil. Table 1 shows the most suitable soils for the cultivation of each of the species under consideration. The soil must have a good physical structure, it must be easily penetrated by air, and must have a

SOILS

CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
1. BLACK SOIL. 2. SANDY BLACK. 3. RICH LIMY. GROUNDS WITH SWAMPY DRAINAGE ARE UNSUITABLE.	1. BLACK TOP SOIL WITH SUBSANDY OR SUBCLAY SOIL. 2. SANDY. 3. SWAMPY.	1. MARLIKE & LIMY SOILS RICH IN HUMUS. 2. BLACK SOIL.	1. SANDY BLACK SOIL 2. RICH LIMY AND WET. 3. SANDY CLAYS. 4. CAN THRIVE ON LIMY STONY SOIL.	1. BLACK TOP SOIL WITH SUBSANDY SOIL. 2. SANDY SOIL. 3. SWAMPY SOIL 4. SOIL BORDERING RIVERS AND LAKES.	1. SALT MARSHY TYPE. 2. SANDY BLACK. 3. SOIL BORDERING RIVERS AND LAKES.

Table 1

sufficient amount of humus or organic material, so that it will be neither powdery nor caked on the surface. Therefore, the best soils are black soils, such as black top soil with sandy or light clay subsoils having a sufficient amount of humus. Soils containing over 50 per cent clay, all clay, or poor sandy material, are less suitable. Usually a certain amount of lime (10-15 per cent) is considered desirable in soils where aromatic plants are grown. In order to obtain the necessary moisture, porosity of soil is important, since this type of earth holds the water for a long period of time, absorbs heat easily, and permits free passage of air to the roots, all of which are requisites for the normal growth of aromatic raw material. Table 2 indicates the type and amount of fertilizer to be used in each case.

PLANTING REQUIREMENTS

FERTILIZER IN POUNDS PER ACRE (ONE YEAR PRIOR TO PLANTING)

CORIANDER		CARAWAY		ANISE	FENNEL	ANGELICA	LICORICE	
TYPE	AMT.	TYPE	AMT.				TYPE	AMT.
HUMUS AND PHOSPHORUS IN FORM OF SUPER PHOSPHATES.	250 TO 350 POUNDS	POTASSIUM IN FORM OF POTASSIUM CHLORIDE.	90 TO 130 POUNDS	SAME AS CORIANDER	SAME AS CARAWAY	SAME AS CORIANDER	HUMUS AND NITROGEN IN FORM OF AMMONIUM SULPHATE	150 TO 250 POUNDS
NITROGEN IN FORM OF AMMONIUM SULPHATE.	90 TO 130 POUNDS	NITROGEN IN FORM OF AMMONIUM SULPHATE.	90 TO 130 POUNDS					

Table 2

The soil is prepared for planting by more or less deep ploughing, which must be deep enough to insure a fast growth of roots, and, therefore, of the plants themselves. Ploughing can be effected by ordinary implements.

Success in planting aromatic raw materials depends upon several factors, the more important being:

1. Thorough preparation of the soil at the proper time.
2. Planting in the season established as most suitable for each type of aromatic plant.
3. Thorough but not very deep planting of clean, grade A seeds or roots into a wet ground. The seeds must have shown high percentage of germination in the previous year's crop.
4. Planting using a technique (which will be explained later).
5. Good care of the plantation after seeding.

Table 3 illustrates the depth of ploughing required by each of the raw materials under consideration.

In order to have a good crop, the seeds used must have previously a high percentage of germination; i.e., a high ratio of seeds which will grow to those that will not. The following is a simple method for determining this germination. After the seeds have been carefully cleaned, the germ-

PLANTING PROCEDURE DEPTH OF PLOWING					
CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
5"-8"	7"-8"	7"-8"	7"-8"	7"-10"	12"-18"

Table 3

inative capacity is checked by pouring some water into a small plate, placing a glass or wooden piece in the middle of the plate, covering the latter with blotting paper, and soaking its ends in water. On the paper 100 seeds are deposited for testing. These seeds are covered again with the blotting paper and placed in a warm, not too dry place with the temperature between 60° and 75°F. As the seeds germinate, or sprout, they are removed, and the date noted. After the whole germination process is finished, the final number of sprouts are counted and the ratio of sprouted to not-sprouted seeds is calculated (in percentages). This is known as the germination percentage, and it indicates how many seeds will grow when planted. Table 4 gives the amount of seed required in the planting of each species.

PLANTING REQUIREMENTS SEEDS IN POUNDS PER ACRE					
CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
10-13 lbs.	10.5-13 lbs.	10.5-12.5 lbs.	7-9 lbs.	8-11 lbs.	8-10 lbs.

Table 4

The planting of aromatic material is usually accomplished either in the spring or in the fall of the year, depending on the species. Early spring planting begins as soon as the earth thaws and is sufficiently dry so that particles of soil will not cling to the planting implements. In this season the seeds of those plants which do not suffer from temperatures below freezing, are sown; for example, caraway and anise.

Immediately after early spring planting, late spring planting begins, and continues until the end of April or the beginning of May, depending on the quality of soil. Crops adaptable to late spring planting are coriander and fennel, although on the whole, early spring planting gives better results.

Fall planting must begin either *very* early (the middle of August and the beginning of September) or *very* late (just before the frost begins). In the first instance, the sprouts will have time to develop and get firm before winter sets in (examples are caraway and fennel); in the second case, the seeds are not given a chance even to begin sprouting or germinating (examples are coriander and fennel). However, late fall planting is rather dangerous, since early spring storms may be so violent that the whole crop, together with the porous black soil, could be blown away, whereas seeds planted in early fall have enough time to establish themselves and grow strong enough to withstand storms.

PLANTING PROCEDURE
DISTANCE BETWEEN ROWS

	CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
SEEDS	15"-22.5"	15"-22.5"	15"-22.5"	22.5"-27.5"	24"-28"	24"-28"
ROOTS					6"-18"	25"

Table 5

With regard to the actual planting, an ordinary seeder can be used for the sowing of seeds, but for planting roots the use of a tomato planter is recommended, which, with some experimentation, can be adjusted for the automatic planting of any root. Table 5 indicates the distance necessary between rows. It is very important to have the rows of seed or roots absolutely straight, as on this depends a successful weeding and re-ploughing. Almost all aromatic plants belong to the group which requires at least one additional shallow ploughing after planting. In order to insure a very uniform distribution of seeds, it is advisable to mix them with dry sand.

Because almost all aromatic plants have seeds of very small size, they

PLANTING PROCEDURE
DEPTH OF PLANTING & SPACING ARRANGEMENTS

	CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
SEED PLANTING	1.2"-1.6"	1.0"-1.2"	.8"-1.2"	.8"-1.2"	.75"-1.2"	1.2"-2.0"
ROOT PLANTING	——	——	——	——	1.5"-2.4"	1.5"-2.4"
SPACING ARRANGEMENT	8"-10"	10"-12"	8"-10"	14"-16"	12"-12"	18"-24"

Table 6

require a shallow planting, which is hard to effect in soft black soil. When such soil is used, it is advisable to press the earth down slightly with a land roller just before seeeding. The depth of sowing is of great importance, since an entire crop can be ruined from planting the seeds too deeply, especially if the seeds are very small. Table 6 shows the requisite depth of planting and spacing arrangements for the various plants.

Care after planting consists mainly in weeding, re-ploughing, and watering. Weeding is the most important of these three processes as weeds are the worst enemies of aromatic crops. It is better to give the land one or two extra weedings than to take the chance of losing an entire crop. A field tiller or weeder can be used, the necessary adjustment being made for each plant.

In the process of cultivation, the land must be harrowed, but the furrows should not be made deeper than 1.5 to 2.5 inches, especially in dry weather, since otherwise the soil will dry out.

Optimum harvesting time varies with the plant, since the degree of ripeness desired differs with the species. Table 7 concerns growth data,

GROWTH DATA

VEGETATION PERIOD & TIME REQUIRED TO REACH MATURITY

	CORIANDER	CARAWAY	ANISE	FENNEL	ANGELICA	LICORICE
VEGETATION PERIOD	52-60 DAYS	50-154 DAYS	130-140 DAYS	150-160 DAYS	150-175 DAYS	150-170 DAYS
GROWTH TO MATURITY	1 YR.	2 YRS.	1 YR.	1 YR. SMALL CROP / 2 YRS. REGULAR	2 YRS.	3-4 YRS.
HARVESTING PERIOD	FALL	FALL	FALL	FALL	SEED-FALL; ROOT {SPRING or FALL	ROOT {SPRING, FALL or WINTER in (SNOWLESS REGIONS)

Table 7

including the vegetation period, the interval before maturity is reached, and harvesting time. Ripeness depends not on the amount of herbage or flowers or seeds, but on the quantity and quality of essential oil contained at the various periods of growth and ripeness. Furthermore, the amount of oil in the plant depends on the time of day during which harvesting is done. There can be considerable loss in yield of essential oil if the crop is collected at the wrong time of day. It is obvious, therefore, that each species must be studied thoroughly before harvesting time can be determined. Usually, the best period of the day is just before the morning dew disappears. Table 8 shows approximate minimum and maximum yield in pounds per acre. For harvesting, equipment similar to the "All-

APPROXIMATE YIELD IN POUNDS PER ACRE		
SEEDS	MINIMUM	MAXIMUM
CORIANDER	750 POUNDS PER ACRE	2200 POUNDS PER ACRE
CARAWAY	900 " " "	1100 " " "
ANISE	1200 " " "	2400 " " "
FENNEL	1000 " " "	1400 " " "
ANGELICA	900 " " "	1300 " " "
ROOTS	MINIMUM	MAXIMUM
LICORICE	900 POUNDS PER ACRE	3300 POUNDS PER ACRE

Table 8

Crop Harvester" can be used, after adjustment to the type of aromatic raw material being cultivated.

After the crop is harvested, the seeds must be threshed, graded, and dried, or, in the case of roots, cleaned and dried. A special tool for cleaning licorice roots must be developed, since the present craftsman's method used in Europe is not satisfactory. This tool could be similar to a potato cleaner, except that the knives should be replaced by metal brushes.

Raw material having a moisture content higher than that indicated below must be submitted to drying as soon as possible, in order to prevent any alteration in its active principle, i.e., essential oils. For this purpose, a drying machine that can be moved from plantation to plantation should be employed. The temperature of drying the aromatic plants mentioned should be 86° F. For licorice, a temperature as high as 104° F. may be used. When dry, the maximum moisture content permissible in coriander, caraway, anise, fennel and angelica seeds, is 12 per cent. Licorice root may have as high as 15 per cent; angelica root, 10 per cent.

Aromatic raw material that is ready for sale must be kept in dry, dark warehouses which have a low temperature and good ventilation. Low temperature and darkness help to avoid losses of essential oils through evaporation and structural alteration of the fragile components contained in the plants. The author's investigations show that there were losses in essential oil content of materials stored under uncontrolled conditions, whereas the amount of essential oils in properly stored seeds and roots remained constant. For this reason, this company stores its aromatic raw material for gin production in a warehouse having an air-conditionng system with a constant humidity of 65 per cent, and a temperature of 48°

F. Figure 2-A shows the automatic recorder which regulate these conditions. Figure 2-b shows the Seagram storage room and shipping containers.

Aromatic materials can be used in the form of either seeds or roots, as condiments for food, or as ingredients in perfumes, medicines, liquors, etc. When crushed, extracted, and concentrated, they provide essential oils. In order to obtain the oil properly, crushing is necessary first, as this process opens the cavities inside the seeds and roots which contain the essential oils, thus allowing the oils to be separated from the plant proper. Figure 3 pictures equipment for cutting roots.

The main practicable methods for the removal of essential oils are: a) water extraction, b) steam distillation.

The most important difference between the two types of atmospheric distillation consists in the use of water in the first case, and dry steam

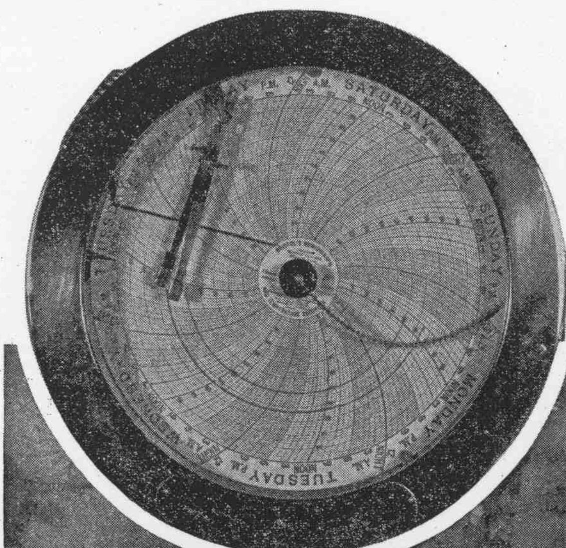


Figure 2-A (left) Automatic recorder-controller for temperature and humidity.

Figure 2-B (below) Storage room and shipping containers for aromatics.

Courtesy of
Joseph E. Seagram & Sons, Inc.
Louisville, Kentucky

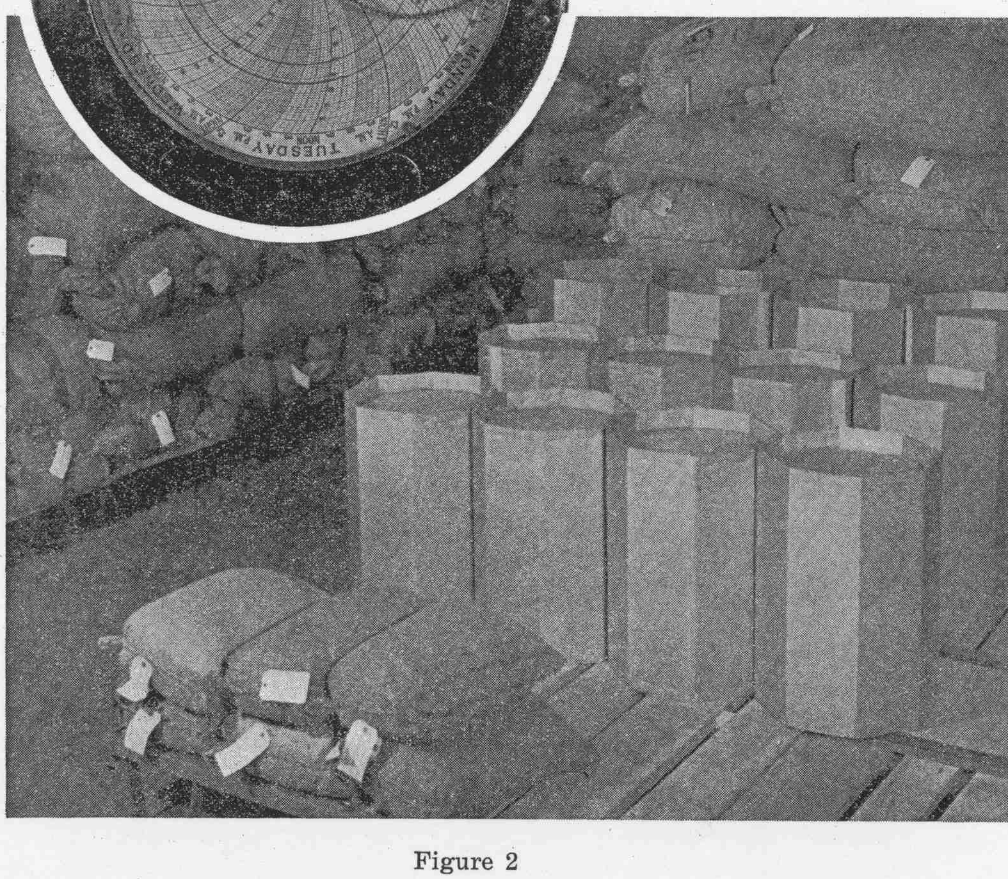


Figure 2

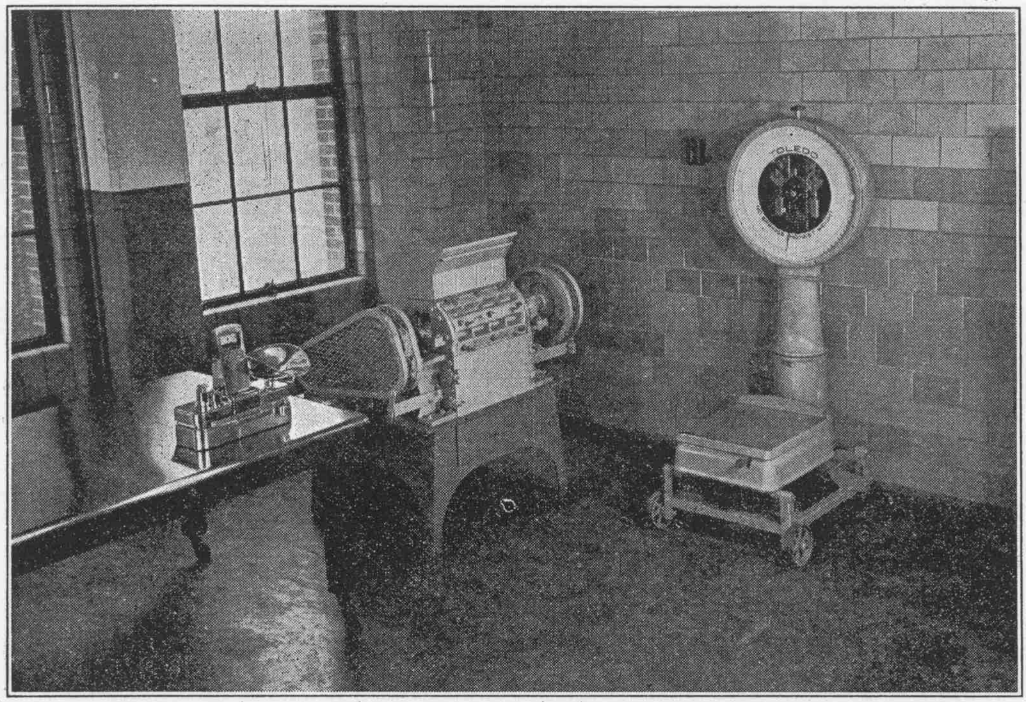


Figure 3

in the second. The former has several inconveniences. In water extraction, steam is formed within a kettle containing water and aromatic material. Heat is applied either by a double bottom or by a direct flame. In this process, part of the raw material may burn along the walls of the kettle, thus adding volatile products of decomposition to the distillate, or, in some instances, the essential oil itself, after being in contact with the hot walls of the kettle, may become decomposed.

The second and most widely accepted method is dry steam distillation, which consists in placing the dry raw material on perforated partitions situated inside a kettle at even distances from each other. In contrast to the water extraction method, steam is produced outside of the kettle in boilers, whence it is injected into the kettle from the bottom by means of perforated copper coils, and travels through the partitions. Since the steam is evenly spread inside the kettle, the danger of localized overheating is avoided. The presence of several baffles or partitions also helps to overcome this difficulty.

The average charging capacity of a steam still is 3000 to 4000 pounds of raw material, which furnishes from 10 to 175 pounds of essential oil, according to the percentage of oil in the aromatic plant used. In many instances, different aromatic plants require different still capacities; for example, while 1 pound of dry mint requires a corresponding capacity of 1 gallon in the still, 1 pound of coriander needs only 0.5 gallon. The thickness with which the raw material is laid on the partitions varies also with the plant used and its unit size. The finer the particle and the softer the material, the thinner the layers on the partitions should be. The average capacity of a partition or plate is 800 to 1000 pounds per square meter

(10.764 sq. ft.) The vapors of steam, saturated with essential oils, pass into an air cooler and then into a condenser, where the two mixed liquids drain into a special decanting container, the oil and water being separated by difference in specific gravity.

The decanting container may be one of two types, depending on whether the oil is heavier or lighter than the water. The oils from the six plants discussed in this paper are lighter than water, and are removed from the top of the container. In this instance, it is very important to drain the water from the bottom of the container as rapidly as possible. Some of the water may be recycled to the kettle. The capacity of the average decanting container is approximately 2 to 2.5 gallons. It is usually made of copper or glass, and is hermetically closed to avoid loss of oil by evaporation.

There are numerous types of condensers, but the best for these oils are straight copper shell-and-tube types, because they are the easiest to clean. Cleanliness is vital in the essential oil industry.

The rate of distillation is very important, as a distillation that is too fast will not permit the steam to make a thorough extraction, while one that is too slow may lead to overheating of raw material, and, in consequence, to alteration of its quality. It is necessary to establish the appropriate rate of distillation for each aromatic plant.

The water which forms the steam must be of the best quality. As condensation inside of the kettle is undesirable, the kettle walls should be insulated and covered with a resistant paint.

For convenience a portable still is proposed for the extraction of essential oils from 10,000 pounds of seeds a day. This still is illustrated in Figure 4.

The still, 8 feet in diameter and 8 feet in height, consists of ten plates 8 inches apart, each 50 square feet in area. Seeds cracked with an ordinary grinding mill are piled upon each plate to a depth of about 5 inches so that the total amount is equal to 200 cubic feet a charge. Cracked seeds afford more surface exposure and prevent their being used again as whole to adulterate other lots. The still will accommodate three charges a day. Steam needed per hour is 2500 pounds; water for boiler and condenser is 50 gallons per minute. If copper material is used, the installation of the still costs from $4000 to $5000. Installation of a 50 H.P. boiler costs approximately $1500 to $2000. By using farm waste such as corn cobs, wood, etc., for fuel, minimum expenses are incurred.

Judging by the mint essential oil industry in Indiana, no engineer's license is required to run the still and no union labor contract is necessary for this type of enterprise.

Essential oil obtained from the charge will depend upon the content in the different seeds. In the table below there is shown the minimum and maximum amounts of essential oils extracted from the various aromatic plants.

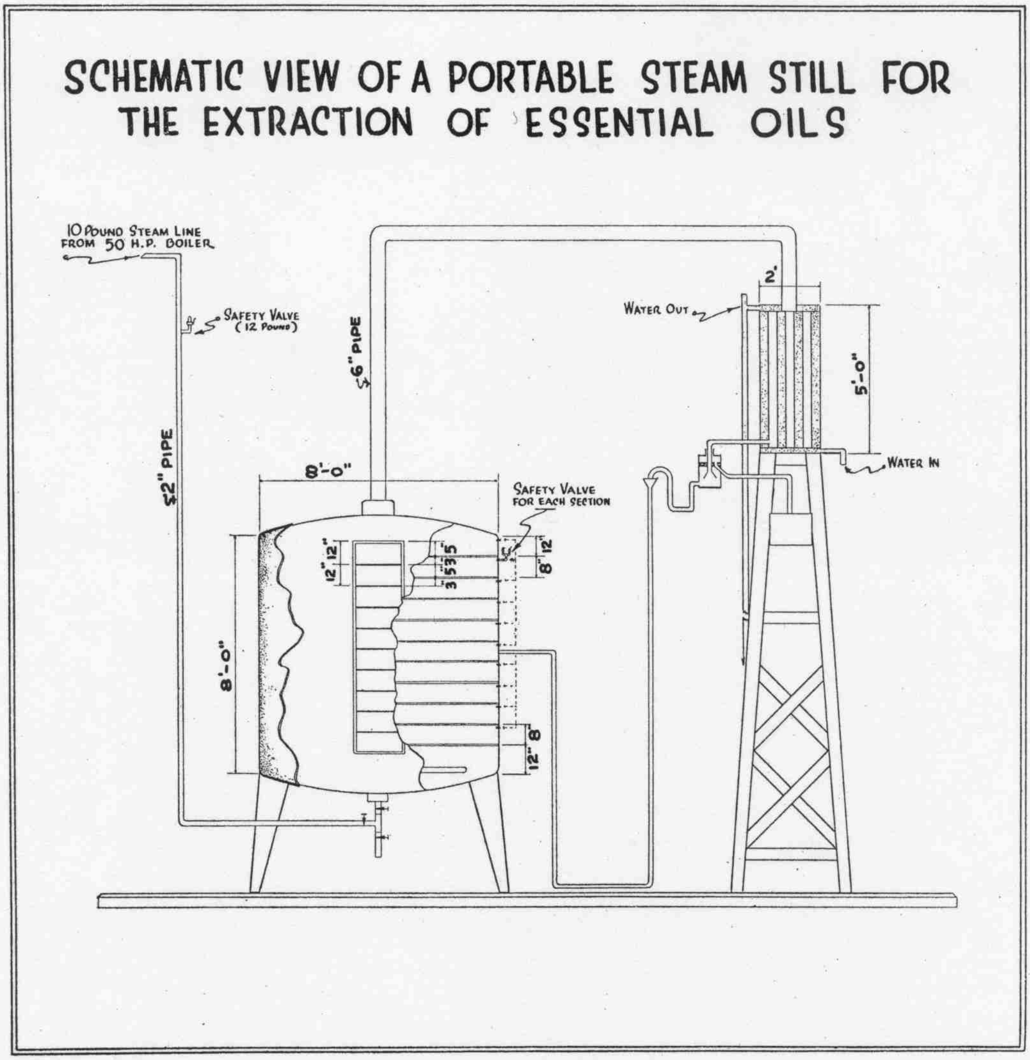


Figure 4.

	Per cent Essential oil		Oil from 3333 lbs. lbs. min.	max.	Wholesale price per lb.
Coriander	0.5	— 1.0	17	34	$12 50
Caraway	3	— 5	100	170	3.25
Anise	2.5	— 3	85	100	3.50
Fennel	4	— 5	150	170	2.25
Angelica Seed	0.75	— 1.0	25	34	48.00
Angelica Root	0.3	— 0.5	8	17	68.00
Licorice Root[1]	....	....	....	....	2.50 (resinol)

After distillation of these materials for essential oils, the seeds contain considerable amounts of butter-fat and protein. This residue could be utilized profitably as cattle and chicken feed.

In the two types of distillation mentioned, the resultant essential oil must be rectified. The best method for rectification is a vacuum fractionation of oil.

Seagram's Research Department has experimented on the application

[1] Production of essential oil from licorice root is not in commercial practice; only the extract and resinol are prepared.

of vacuum extraction to essential oils on a laboratory scale and has obtained an oil with a mild aroma, because of the following facts:

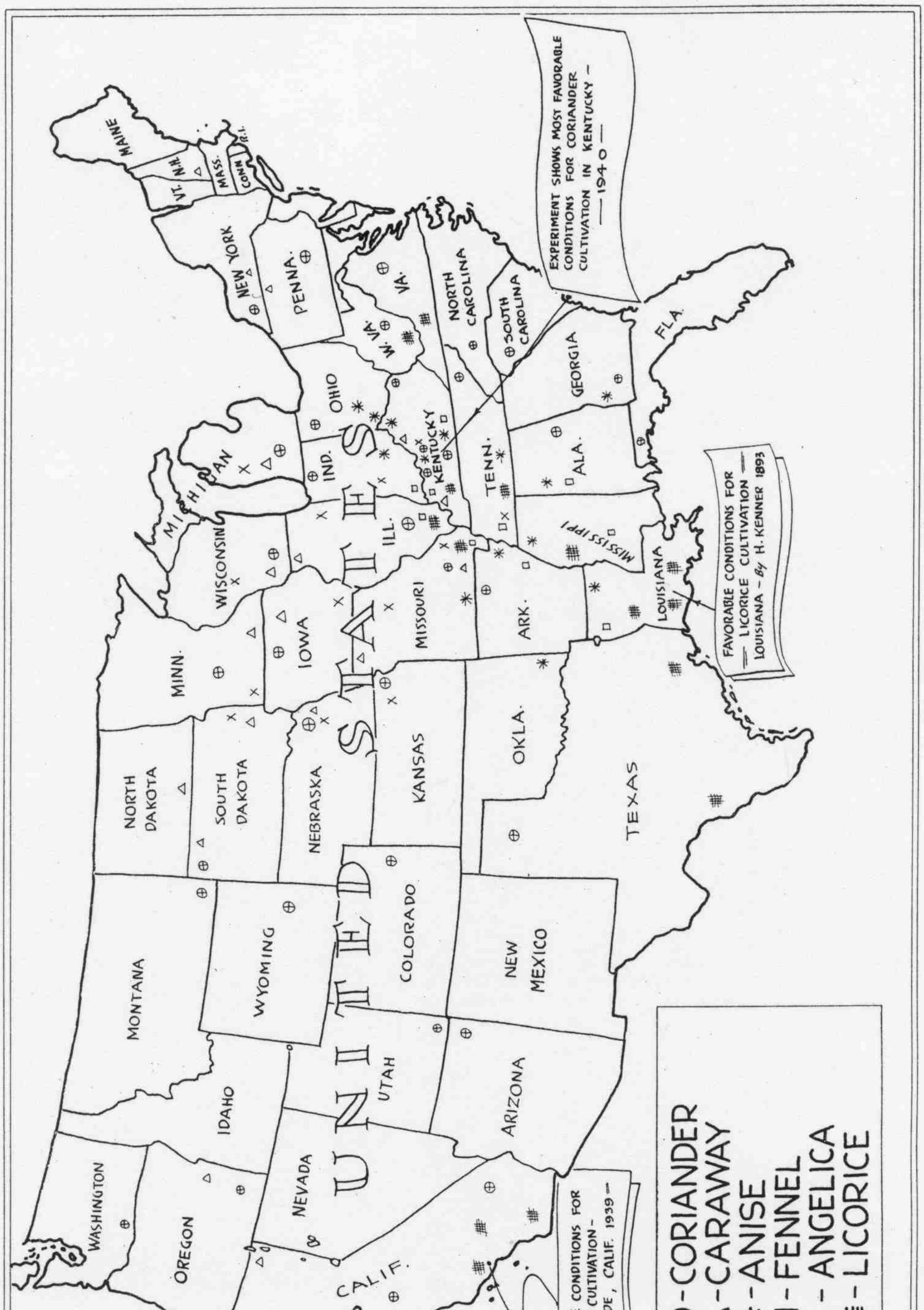
1. The temperature of extraction is low and, therefore, decomposition of materials is minimized.
2. Decomposition of terpenes is eliminated as well as of carbohydrates.
3. The splitting of higher esters and alcohols is avoided.
4. The most volatile parts of organic material are carried over in the distillate.

To emphasize the profitable production of aromatic plants, the cost of production of corn and cotton is compared with that of the aromatic plant. The cost of production for corn averages $15 an acre and this includes the seed, labor and power, manure and fertilizer, and other costs. The cost of production of cotton per acre is $15 and includes seed, land preparation, fertilizers, planting, cultivation, picking, ginning and miscellaneous. The cost of production of an aromatic plant averages $20 an acre, including the extra expenses for weeding. The average return per acre for corn is 50 bushels (2800 pounds); for cotton, 250 pounds; and for the aromatic plant, 1000 pounds. The farmer, realizing $0.50 a bushel for corn receives $25 an acre, netting him a profit of $10 an acre. At $0.08 a pound for cotton, he receives $20 return an acre, nettting him a profit of $5. If it is assumed that the average farmers price for the aromatic plant under discussion is $0.08 a pound, from the yield of 1000 pounds an acre, the total return would be $80 an acre, with a net profit of $60 an acre. In this latter case, the farmer's price is assumed to be a reduction of $0.04 a pound from the market value which was obtained from botanical house price lists for 1937.

The proposed regions of domestic cultivation for the six aromatic plants discussed in this article are displayed on the map of the United States (Figure 5).

In conclusion, the author suggests that:

1. American farmers should start cultivating aromatic plants.
2. Small farms should increase the variety of their crops. Under the existing agricultural system of a single type crop on a whole farm, in good years there is overproduction, and in poor years, entire loss of income because of crop failure.
3. By planting simultaneously a greater variety of crops, the farmer multiplies his chances of being protected against total crop failure.
4. Farms on which aromatic plants are grown, should be run on a cooperative basis, so that the movable agricultural implements may be interchanged.
5. The United States Department of Agriculture should experiment with the entire field of aromatic plants in order to investigate the possibilities of their cultivation in this country.



The Proposed Regions for Aromatic Plant Cultivation

Fig. 5

III

THE PREFERENTIAL ODOUR RATING OF ESSENTIAL OILS FROM CORIANDER SEEDS

by E. H. SCOFIELD

Director of Quality Laboratory
Joseph E. Seagram and Sons, Inc.
Louisville, Kentucky

III.

The Preferential Odour Rating of the Essential Oils of Coriander Seeds

By Dr. E. H. Scofield

I. Introduction

In order to complete the description of the properties of the essential oils of coriander seeds, some form of psychological analysis must be included. This simply means that the materials should be exposed to human sense organs under well defined conditions of stimulation followed by an accurate account of the odour, taste, and certain accompanying perceptions to which the materials give rise. This type of analysis is important from at least two fundamental viewpoints.

In the first place, chemical methods are relatively insensitive when compared with human sense organs in the detection of the particles emitted by odour and taste exciting substances. For example, according to Allison and Katz (1919) chloroform is detectable, in terms of odour, to the extent of 0.0033 ounces per cubic foot of air. The same investigators report that oil of peppermint and artificial musk are detectable respectively to the extent of 0.000023 ounces and 0.0000000409 ounces per cubic foot of air. The sense of taste is likewise a very delicate function. According to Skramlik (1926) (After Kiesow) cane sugar, a typical sweet substance, is detectable on the tip of the tongue in a molecular concentration of 0.014; and hydrochloric acid, a sour substance, in one of 0.0029. He reports also that table salt and quinine sulphate are detectable respectively in molecular concentrations of 0.43 and 0.00000039. The amazing sensitivity of the odour and taste functions, and especially the former, surpass by far the vast majority of the chemical methods of detection. They are approached in sensitivity only by spectroscopic techniques of which, beyond a doubt, we shall hear more and more in the future.

In the second place, it is almost needless to point out that chemical and physical analyses tell us nothing whatsoever about the type of reaction which a given odour or taste excitant will induce when it is exposed to one sense organ or another. In other words, such analyses would give us no clue as to what we ordinarily imply by the term "quality". For instance, all of the chemical and physical data available at the present time describing ordinary quinine would not enable us to predict that this material when placed on the tongue induces a unique experience described by the human observer as "bitter". The same generalization in essentials applies to salty, sweet, and sour substances. By the same token, the chemical analysis of the essential oils of coriander and peppermint would not enable us to predict the unique experiences induced when their vapors contact the olfactory membranes of the nose. This whole problem is further complicated when one takes into account certain additional factors which appear in the combined operation of two or more odours or two or more tastes. These factors include fusion, compensation, and adaptation. According to Boring, Langfeld, and Weld, (1935) the fusion of two odourous substances gives rise to what one might describe as a psychological mixture. These authors claim that a mixture of eucalyptol and pyradine in a ratio of 9.5 to 1 resembles the odour of camphor. When compensation operates, the resultant odour is much weaker than either of the odours of its separate components. Titchener (1926) states that the odour of red India-rubber neutralizes the odours of cedar wood, gum benzoin, paraffin, beeswax, and tolu balsam. Adaptation to odour is a very common phenomenon. This process operates in such a manner that odours of the most insistent kind fade out if the sense organs of the nose are exposed to them for a sufficiently long period of time. For example, the repeated inhalation of the vapors of a strong perfume leads temporarily to an almost complete disappearance of its scent. Fusion, compensation, and adaptation operate also in the taste mechanisms. Thus the taste of a weak lemonade is a fusion of sour and sweet. In addition, lemonade is less sour and less sweet presumably because the acid and sugar weaken each other's effect. This latter phenomenon is an example of taste compensation. Adaptation is illustrated by the gradual weakening in the intensity of the flavor of a salt solution if it remains in the mouth for a relatively long period of time.

One should not create the impression that no attempt has been made to correlate taste and odour with chemical constitution. Many such studies have been conducted. A typical one is that of Oertly and Myers (1919). For example, they have listed a number of sweet producing molecular arrangements and groupings. Crozier (1934) in discussing the threshold concentrations of the odourous materials of Allison and Katz (1919) points out that the higher members of homologous chemical series of compounds are more effective as excitants than the lower members. While the trends of such investigations merely have been indicated here, it is certainly true that the results so far are not very convincing or encouraging. One is inclined to agree with Crozier (1934) when he comments that: "There are, however, at the present time too many puzzling irregularities in the taste exciting properties of organic molecules to permit useful rationalizations of the details of gustatory (taste) excitation."

This introduction, while brief, will serve to emphasize the necessity of employing the human observer for the purpose of completing the description of such materials as the essential oils of coriander.

II. Problem

The most general objective of this study was that of determining the quality of six different oils of coriander in terms of human reactions. In particular, it was considered of importance to study the influence of the materials on the sense organs of the tongue and nose, thus giving rise to a description of their taste and odour properties. It should be possible by scientific methods to determine the differences, if any, in the flavor intensities of the six materials. In the most practical sense, this simply means that the oils would be rated in such a manner that they could be arranged on an intensity scale ranging from the most intense to the least intense. In the strictly operational sense, this would mean conducting the experiment with the objective of determining eventually the minimum amount of the oil which must be present in a specific volume of solution before it would just give rise to the odour or taste experience. For example, and to state the matter in a somewhat abstract manner, say that one should find that x milligrams of one oil in B volumes of solution can just be tasted. Also let us say that it requires twice as much or 2x milligrams of another oil in the same volume of solution to give rise to the same experience. Thus, the first oil would be rated as having twice the flavor strength of the second. In other words, the more oil required in a constant volume of solution to induce an odour or taste experience, the weaker it is. We shall not describe the technique or any of its further possibilities in this report. It will suffice to say that technically the procedure is known as threshold determination and now has a long and involved scientific history.

Another possibility of quality rating would consist in employing some kind of system or systems leading to the rating of the materials in terms of relative odour or odour-taste pleasantness. This is very important from the industrial viewpoint and especially so when one considers that the essential oil of coriander is employed on a large scale basis as a flavoring material, (Clarke, 1922). It should also be of special interest to know how the essential oils of coriander from different countries and from different sources in the same country compare with one another. In other words, which oil is the most pleasant, the next most, and so on down to the least pleasing? The experiments described in this report were limited to a solution of this problem.

It will be noticed that throughout this discussion differentiation has been implied between the sense of odour and the sense of taste. Rossi (1940) in his article with the title "Wine Tasting" quotes from Andre L. Simon: "The tongue, although it reacts to heat and cold, has no means of detecting flavors and savors; it cannot taste." This statement as it stands is most perplexing in view of the vast amount of anatomical, physiological, and psychological work which has been accomplished in the study of the odour and taste functions. Howell (1933) and many others point out very conclusively that the tongue reacts to at least four fundamental tastes including sweet, bitter, sour, and salt. If we accept the general classification of Henning (1921), the odour experiences fall into six fundamental classes which include: spicy, burnt, resinous, flowery, fruity, and foul. Presumably, the vast majority of the experiences resulting from the introduction of foods and liquids into the mouth cavity are a combination of these various taste and odour qualities.

In order to limit further the problem described in this report, an attempt was made to determine if taste, as the term is technically defined, would enter into the picture to an appreciable extent. Three observers were instructed to taste the oils with their nasal cavities blocked. It was found that the only experience resulting from the introduction of the oil into the mouth was describable in terms of a "slight burning quality". Discrimination under such conditions became impossible. Therefore, it was considered sufficient to limit the rating of the six substances to odour alone, thus obviating the necessity of placing the material on the observer's tongue. The general procedure will be described in the next section.

III. Method

The physical properties of the coriander seeds from which the six oils employed in this study were extracted are presented in Table I on page 19 with the title "Coriander Seeds Assayment". The pyrex containers into which the oils were introduced will be referred to as inhaling beakers. An inhaling beaker has a shape very much like an elongated brandy inhaling glass with a diameter of about 2.2 inches at the center and 1.5 inches across the top and bottom. Its capacity is equal to approximately 9.15 cubic inches. The glass proper is mounted on a stem with a circular base. Such properties provide for a container which is extremely convenient to manipulate both for the experimenter and the observers. All traces of odour were removed from the beakers by a thorough cleansing. This included washing with a special soap preparation followed by rinsing in tap water. Finally, they were rinsed in distilled water and dried by heat in an electric oven. The six essential oils of coriander, which were stored in small weighing bottles, were introduced into the beakers by pipettes. Individual pipettes were provided for each beaker. Preliminary observations revealed that the odours were so intense that only two drops of oil per beaker were required. Each beaker was covered with a watch glass immediately after the introduction of the oil. The laboratory in which the experiment was conducted was relatively free of extraneous odours and maintained a temperature of 74° F. with a variation not exceeding plus or minus 2° F. The experiment proper was conducted on a table provided with a horizontal partition for separating the experimenter and the observer. Thus, manipulations of the experimenter were completely concealed from all persons participating in the study. The partition was provided with an opening in its center through which the beakers were passed to the observers.

The procedure of presentation employed may be referred to as the method of paired comparisons. The general characteristics of this method were described by Titchener (1901). Fundamentally, it consists of a technique whereby every sample in the series is compared with every other sample. For instance, say that one had only three samples. This would mean that sample 1 would be compared with 2; sample 2, with 3; and, finally, 1, with 3; making a total of three comparisons per observer. The six samples employed in this study required 15 comparisons per observer. Employing the method as described, the preferential odour reactions of 25 observers, 15 men and 10 women, were obtained. They ranged in age from 22 to 37 years. All represented employees in a variety of capacities of Joseph E. Seagram and Sons, Inc., at Louisville, Kentucky. Many of them had had some experience in previous odour and taste preference experiments. However, the point which must be emphasized is that none of them should be classified as odour or taste experts. In fact, it was argued that the utilization of the conventional expert would be detrimental to the fulfillment of our experimental objectives. In the final analysis, and in an extremely practical sense, the typical consumer is the most critical and the best judge of the merits of a product prepared for human consumption. Thus, in effect, a sampling of consumer odour preference for the essential oils of coriander was obtained. A typical experimental session may be described as follows: The experimenter and observer remained seated opposite one another at the table and separated by the partition. The instructions to the observer were given as follows:

> "This is an experiment dealing with the abstract odour properties of a series of liquids. We wish to rate the liquids in order of odour pleasantness. You will be given these liquids two at a time. Please compare them by inhaling their vapors, first the one on your right, followed by the one on your left, and so on as often as the time allowed will permit. You will be given a total of 20 seconds for making your judgment. Indicate your preference by returning to the experimenter first of all the beaker containing the liquid with the more pleasant odour, followed by the second or lesser preferred. Do not guess. If you are doubtful, indicate accordingly."

A 30 second rest interval was allowed between the presentation of each pair of samples. All time intervals were controlled by the use of a stopwatch. It must be emphasized that the beakers remained covered with the watch glasses except during the time they were in the hands of the observer. The entire series of samples was renewed 5 times during the course of the experiment. All pairs indicated by the observer as "doubtful" were returned to him at a later time in the experiment. The instructions as given were identical for all observers with one exception. This consisted in alternating that portion of the instructions dealing with the beaker to be smelled first. For instance, the first subject smelled first of all the one on his right, the next observer the one on his left, and so on. In this manner, any individual preferences for the spacial position of the beakers were nullified. Thus, 15 comparisons were made by each of the 25 observers. The data were recorded on standard paired comparison charts with the sample numbers indicated on the horizontal and vertical axes and the responses indicated in the body of the chart by arabic numerals.

The experiments were conducted over a period of three days. At the end of this time it was considered desirable and important to re-rate the samples by an independent method. This check method may be described as a rank order one. Here the six beakers were placed on a large table and arranged in a serial order on a line running transversely with the observer. The latter was instructed to inhale the vapors of each beaker as often as required and finally to re-arrange them in order of preference from left to right. Twenty-five observers again participated, all but five of whom were employed in the preceding experiment.

IV. Results and Discussion

The combined results of the 25 observers' preferential reactions on the basis of odour and by the employment of the paired comparison method are presented in Table I.

TABLE I

Numerical Odour Ratings in Terms of Percent Preference of Six Essential Oils of Coriander by the Method of Paired Comparisons

Sample	*Description of Seeds from Which Oils Were Extracted*	*Percent Preference* (Odour)
1	U.S.A., Kentucky overripe	23.2
2	U.S.A., Kentucky unripe	20.7
3	Moroccan	17.2
4	U.S.A., Kentucky ripe	15.3
5	Hungarian	12.9
6	Russian	10.7

This table is based on the following calculations. The total number of preferences for each sample was obtained directly from the 25 paired comparison charts. Each value was then divided by the total number of comparisons made, namely 375. Thus, sample 1, U.S.A. Kentucky over-

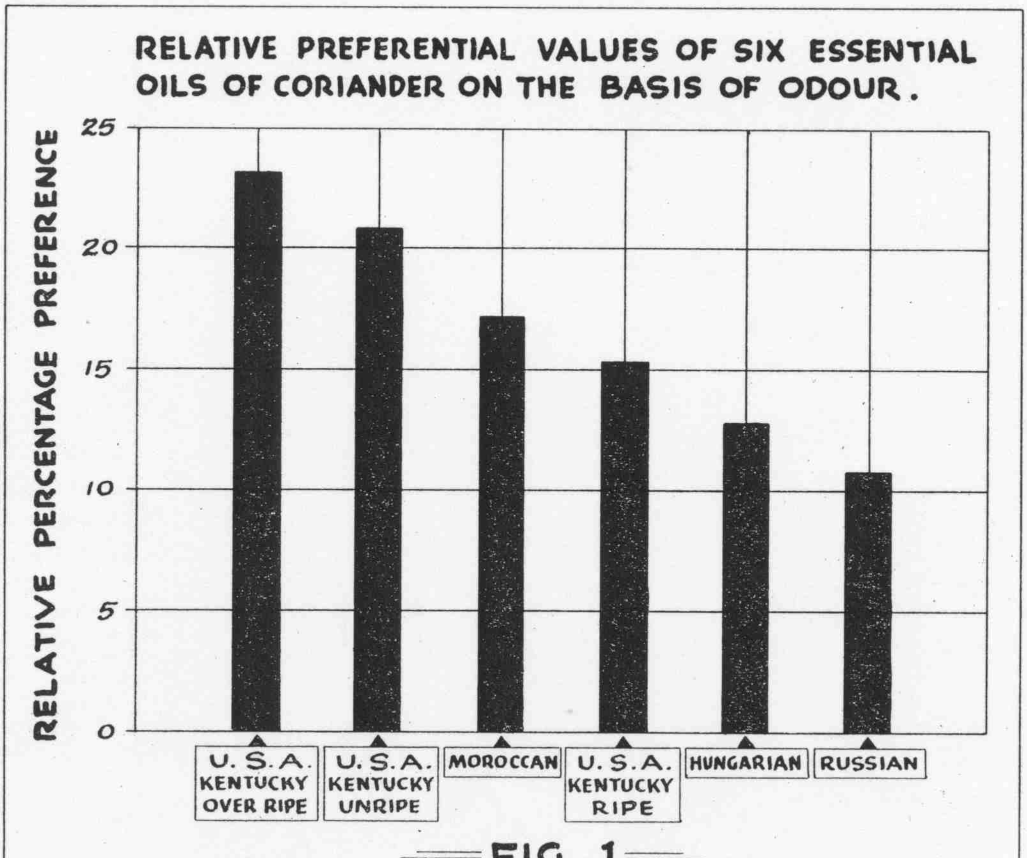


FIG. 1

ripe, was selected 23.2 percent of the time; sample 2, U.S.A., Kentucky unripe, 20.7 percent of the time, and so on. These results are represented graphically in Figure I. It will be noticed that on a relative basis these samples vary from one another by approximately equal amounts. The data indicate that the oil with the most pleasing odour in the series is the one derived from the U.S.A. Kentucky overripe seed. The second most pleasing is the oil derived from the U.S.A. Kentucky unripe seed, and so on with the Russian seed yielding the oil with the least pleasing odour. One must bear in mind in interpreting these results that the oils were rated in relative terms of odour pleasantness and not absolute ones. In other words, the data do not tell us to what extent an odour is pleasing or to what extent it is unpleasant, but rather how it compares with all other members of the group. Since our original objective was simply that of determining the relative merits of the six oils, the data as given are sufficient. Another point is worthy of consideration at this time. It will be recalled that our observers, in the course of the instructions, were told that this would be an abstract odour study. Thus, objectively speaking, the data should not be interpreted in terms of specific industrial uses of oils. This simply means that the data as they appear here signify that the pure odours of some of the oils are more pleasing than those of others. They do not mean that one may immediately conclude that an oil with the highest preferential rating in this report would retain its relative position on a preferential scale if, along with others, it were combined in various mixtures or solutions and the whole experiment duplicated. For instance, the only way one might determine whether or not these materials would retain their relative positions when employed, say, as a flavoring material for beverages, would be to incorporate them in the desired combinations and again obtain a preferential rating. However, one is probably safe in predicting that the chances are that the oils with the superior odours in the abstract sense would yield in general superior combinations or mixtures in the majority of instances.

While no analytical attempt was made in this study to isolate those factors contributing to the superiority of one odour over another, some of the voluntary comments of the observers were interesting. It was found that in general the comments describing the inferior odours were stated in terms which automatically fall into two of Henning's classifications, namely, resinous and spicy. These terms in particular were advanced in connection with the Hungarian and Russian samples. This might be due to the fact that the Hungarian and Russian oils were derived from the 1939 crop. In other words, it is entirely possible that age is to some extent a quality determining factor. While one cannot be sure without additional experimentation, it would seem that it was simpler for the observers to describe the relatively unpleasant odours more readily than the relatively pleasant ones.

The results of the check experiment are shown in Table II:

TABLE II

Numerical Odour Ratings of Six Essential Oils of Coriander by the Rank Order Method

Sample	*Description of Seeds from Which Oils Were Extracted*	*Total Odour Scale Value*	*Average Odour Scale Value*
1	U.S.A., Kentucky overripe	144.7	5.8
2	U.S.A., Kentucky unripe	136.8	5.5
3	U.S.A., Kentucky ripe	132.7	5.3
4	Moroccan	123.8	4.9
5	Hungarian	106.8	4.2
6	Russian	103.6	4.1

The two scale value columns are derived on the basis of the procedure described by Hull (1928) for treating rank order data. For the present purposes, the average column simply represents the order of merit in terms of odour pleasantness of the six samples and the approximate psychological distances between the members of the series. Thus, these results stand in close agreement with those presented in Table I. In fact, there is only one disparity. The U.S.A. Kentucky ripe and the Moroccan rate third and fourth respectively, while in Table I this order is reversed.

The evidence in its entirety rather conclusively points toward the relative superiority of the oils extracted from the U.S.A. Kentucky overripe and unripe seeds and the relative inferiority of those extracted from the Hungarian and Russian ones.

V. Conclusions

1. The essential oil of coriander extracted from the U.S.A. Kentucky overripe seed is superior in odour to the five other oils employed in this study.

2. The essential oils of coriander extracted from the U.S.A. Kentucky overripe and unripe seeds occupy the two highest positions, in terms of odour, on the basis of two independent rating methods.

3. On the basis of the rank order method, the U.S.A. Kentucky oils occupy, in terms of odour, the three highest positions.

4. In general, the two oils with the lowest rating, the Hungarian and the Russian, are describable in such terms as "resinous" and "spicy".

VI. Bibliography

1. ALLISON, V. C., *and* KATZ, S. H.
An Investigation of Stenches and Odors for Industrial Purposes. J. Indus. Eng. Chem., 1919, Vol. 11.

2. BORING, EDWIN G., LANGFELD, HERBERT S., *and* WELD, HARRY.
Psychology, a Factual Textbook. John Wiley and Sons, Inc. New York, 1935.

3. CLARKE, A.
Flavoring Materials, Natural and Synthetic. Henry Frowde and Hodder & Stoughton, London, 1922.

4. CROZIER, W. J.
Chemoreception. In Handbook of General Experimental Psychology. Worcester, Mass. Clarke University Press, 1934.

5. HENNING, H.
Der Geruch. Second Edition, Leipzig, 1924.

6. HOWELL, W. H.
A Textbook of Physiology. W. B. Saunders Co., 1933.

7. HULL, C. L.
Aptitude Testing. World Book Company, Yonkers, 1928.

8. OERTLY, E. *and* MYERS, R. G.
A new theory relating constitution to taste. (Preliminary Paper) Simple relations between the constitution of aliphatic compounds and their sweet taste. J. Amer. Chem. Soc., 1919, Vol. 41.

9. ROSSI, EDMUND A.
Wine Tasting. American Wine and Liquor Journal, June, 1940.

10. SKRAMLIK, E.
Handbook der Physiologie der Niederen Sinne: Vol. 1. Die Physiologie des Geruchs-und Geschmackssinnes. v. 1926. Leipzig: Thieme.

11. TITCHENER, E. B.
Experimental Psychology. The Macmillan Company, New York, Vol. 1, Pt. 1, 1901.

12. TITCHENER, E. B.
A Textbook of Psychology. The Macmillan Company, New York, 1926.

APPENDIX

IV

LABORATORY ASSAYMENT OF RAW MATERIALS FOR ESSENTIAL OILS

IV.

Laboratory Assayment of Raw Materials for Essential Oils

(1) Moisture: a. by desiccator c. total moisture
b. by oven d. by Dean-Stark tube

(2) Acidity: as milligrams of normal potassium hydroxide per one gram.

(3) Oil Content:
a. by the Seagram modification of Johnson's apparatus
b. by the Seagram modification of Clevenger's apparatus

(4) Contamination

1. Moisture

a. *by desiccator*

An empty dish, preferably a standard Petri dish, is tared accurately and 10 grams of the material to be studied is weighed into it. (Weighed to the fourth decimal place.) Seeds are weighed whole; roots, powdered or whole. In the case of angelica root, usually a whole root is weighed, even if it is heavier than 10 grams. Licorice is weighed in sticks. The Petri dish with the material is then placed in a sulphuric acid (66° Bé) desiccator and left over night for 16 hours. (Sulphuric acid must always be fresh.) After that time, the dish with the material is reweighed, and the per cent of moisture in the desiccator is calculated.

Example: Petri dish	38.5910 grams
Dish + material	48.5910 grams
	10.0000
After 16 hrs. in desiccator	48.4178 grams
Difference for 10 grams material	0.1732 grams or 1.732%

It has been noted that a new raw material which has just arrived from Europe or Asia by boat loses too much moisture by the desiccator. It is better, therefore, to keep the sample for 2 to 3 days in the laboratory at room temperature before starting the determination.

Next, all material is ground on a mill. In order to establish a standard dimension of particles for each ingredient the material is sieved through four standard sieves, an average taken and used every time.

	Opening	*Diameter of wire*
No. 8	0.09 in.	0.03 in.
No. 12	0.07 in.	0.027 in.
No. 16	0.05 in.	0.02 in.
No. 20	0.03 in.	0.016 in.

Below are a few examples:

Left on the Sieve	*Coriander*	*Angelica*
No. 8	3.0%	2.0%
No. 12	10.0%	2.0%
No. 16	25.0%	14.0%
No. 20	36.0%	16.0%
through No. 20	26.0%	66.0%

b. *by oven*

An empty evaporating dish, 80 mm. in diameter, 45 mm. in height, is tared to the fourth decimal place. 5 grams of ground material from the previous moisture by desiccator determination is added, weighed to the fourth decimal place and placed into a dry oven for 3 hours at 103° to 105°C. (After much experimentation, it was found that after 3 hours losses are insignificant; hence it is not necessary to keep the material in the oven any longer.)

Finally, the evaporating dish is placed in a calcium chloride desiccator, cooled for 30 minutes and re-weighed. Per cent of moisture in the oven is calculated.

Example:	Empty evaporating dish..................	38.1064 grams
	Dish + material..................................	43.1064 grams
		5.0000 grams
	After 3 hours in oven........................	42.7900 grams
		0.3164 grams or 6.328%

c. *total moisture*

Total moisture is found by adding moisture by desiccator to the moisture by oven.

Example: 1.732% + 6.328% = 8.060%, total moisture

d. *by Dean-Stark tube*

The Dean-Stark apparatus consists of a 250 ml. flat bottom flask, a Dean-Stark tube, a four bulb condenser and a calcium chloride tube, all equipped with standard taper ground glass joints. See Fig. 1.

10 grams of material is accurately weighed into the distilling flask and covered with 100 ml. of pure, dry xylene. The flask is connected with the Dean-Stark tube and a condenser equipped with a calcium chloride tube. See Fig. 2.

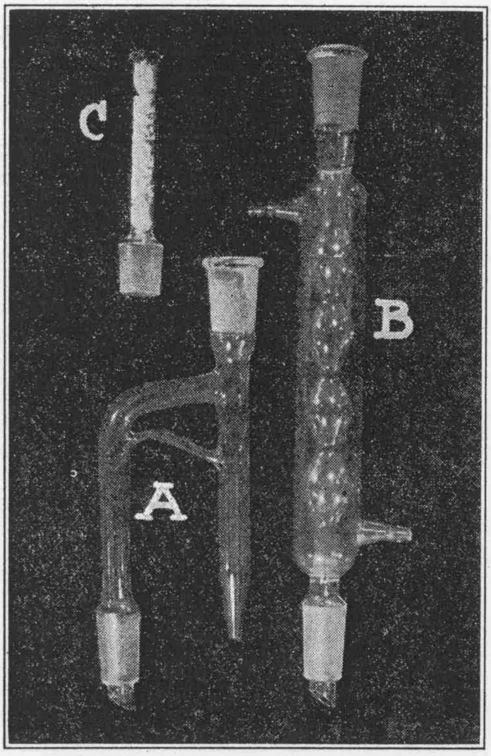


Figure 1

(a) Dean-Stark Tube
(b) 4-bulb Condenser with Ground Glass Joint
(c) Ground Joint Calcium Chloride Joint

The complete apparatus is placed on a small electric hot plate.[1] The heat is increased gradually during 1 hour or more from low through medium to high. The vapors of xylene, charged with vapors of water extracted from material, meet the cold surface of the condenser, condense together in the tube and, separating by specific gravity differential, remain at the bottom of the tube. The excess of xylene returns to the flask and exhausts the material. The end of the determination occurs when for about 10 minutes on "high" there is no increase of volume of water. The reading multiplied by 10 gives the per cent of moisture by Dean-Stark tube. This method is used for

[1] Fisher Scientific Company, Pittsburgh, Pennsylvania, type T 0102, 115 volts, 5.2 amperes.

speed, but is not so accurate as moisture determinations by desiccator or oven.

2. Acidity

The determination of acidity of material is carried out as follows: Place 2 grams of ground material, dried in the desiccator, in a 50 ml. ground joint Erlenmeyer flask, cover with 25 ml. of 95% alcohol and leave for 16 hours. After that period, filter, wash the flask and the filter with 25 ml. of fresh alcohol and titrate with N/20 sodium hydroxide using phenolphthalein as indicator. Titrate 50 ml. of alcohol as a blank. Subtract the blank from the titration figure and calculate as milligrams of potassium hydroxide. Example: 3.3 ml. N/0.0471 sodium hydroxide used for titration of 50 ml. of sample. For blank, 0.3 ml. of same sodium hydroxide. 3.3 — 0.3 = 3.0 ml. 0.0471 x 56 (molecular weight of potassium hydroxide) x 3.0 = 7.9128 milligrams per 2 grams, or 3.9564 milligrams of potassium hydroxide per 1 gram of material.

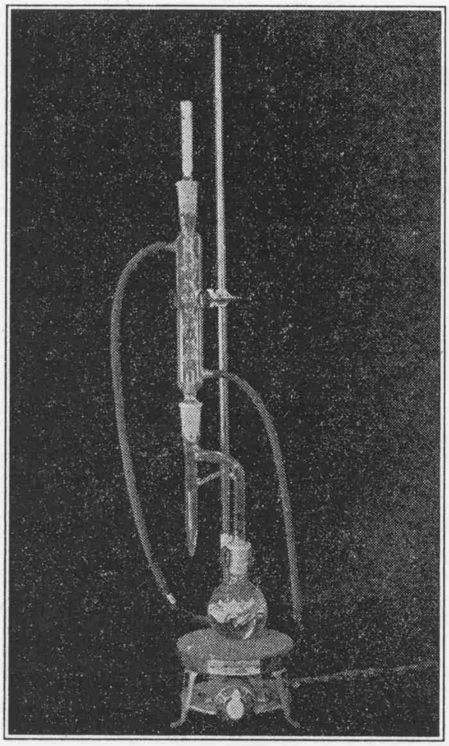


Figure 2
Dean-Stark Apparatus Used for the Rapid Determination of Moisture Content in Raw Materials for Essential Oils

3. Quantitative determination of essential oil

by a. Ether extraction (Seagram modification of Johnson's apparatus)

b. Steam distillation (Seagram modification of Clevenger's apparatus)

a. Apparatus.[1] The apparatus consists of one small 50 ml. flat bottom flask, one central section for extraction, one four-bulb condenser of special design to avoid penetration of condensed water from condenser into the extraction apparatus, and a calcium chloride tube, all equipped with standard taper ground glass joints. The flasks rest in a water bath, heated by an electric hot plate. See Fig. 3.

Procedure. Place 2 grams of material, weighed to the fourth decimal place, into a paper thimble of 16 mm. diameter and 100 mm. height. Place the thimble in the middle part of the apparatus as indicated on the picture, connect the middle part with the flask, pour 25 ml. of anhydrous ether into the thimble, carefully avoiding overflow of particles of material into extraction flask, place in water bath and immediately connect with the condenser equipped with calcium chloride tube and slightly cooled.[2] Increase the temperature. See Fig. 4.

[1] Special Equipment for modified Johnson's and modified Clevenger's apparatus was provided by the Cincinnati Scientific Company, Cincinnati, Ohio, according to designs submitted by Seagram.
[2] If previously cooled too rapidly, sweating may commence and it may be impossible to avoid water drops in the ether.

When the temperature has been raised to 140°F. (60°C.) and the drops are regularly falling in the flask from the thimble, note the time and keep refluxing for 6 hours at 140°-150°F. (60°-65.6°C.) Cover the water surface with paraffin to prevent the evaporation of water. At the end of 6 hours, cool, disconnect the apparatus, remove the thimble with forceps (hold until no drops of ether are falling) and pour quantitatively the contents of the flash into a tared clean evaporating dish.

Rinse the flask and the middle section of apparatus with 5 ml. of fresh ether, and add this to the evaporating dish. Place the dish in a water bath exactly at 47°C. (116.6°F.). Evaporate ether in 14 to 15 minutes at that temperature. The end of the evaporation is reached when there is absolutely no odor of ether. Place the evaporating dish in sulphuric acid desiccator for 15 minutes, and re-weigh. The difference between the empty dish and the dish after extraction is called the *total ether extractive matter*. Then the evaporating dish is placed in a drying oven, where the temperature is slowly raised from 50°C. (122°F.) to 110°C. (230°F.). 2 hours after the temperature of 110°C. is established, place the dishes in the desiccator to cool for 30 minutes and re-weigh. The difference between the previous and the final weighing is called the amount of *essential oil* (per 2 grams.) The difference between total ether extractive matter and essential oil is called *total non-volatile matter*.

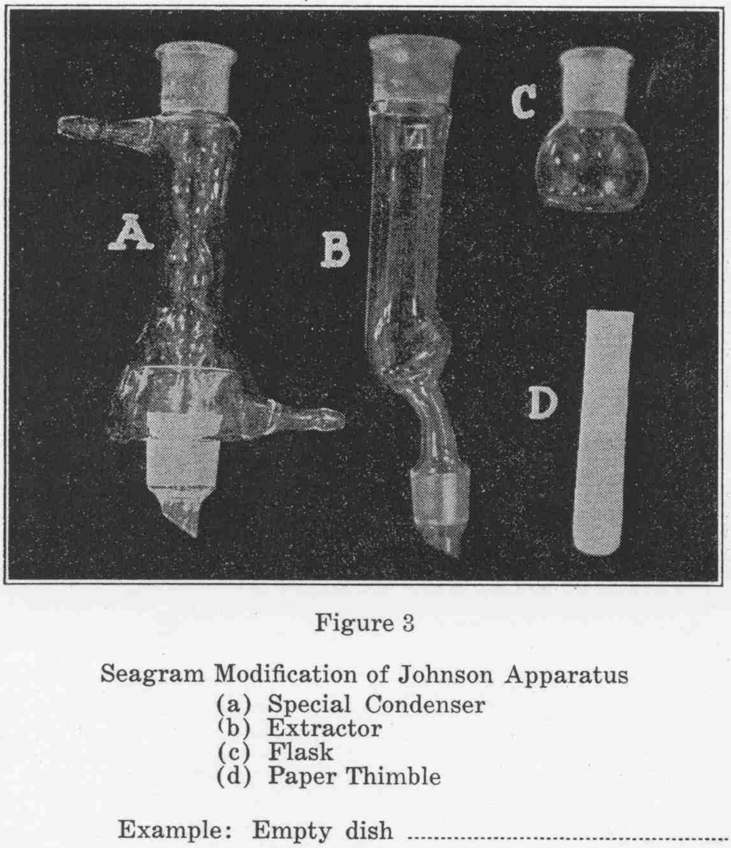


Figure 3

Seagram Modification of Johnson Apparatus
(a) Special Condenser
(b) Extractor
(c) Flask
(d) Paper Thimble

Example:	Empty dish	38.1064 grams
	After extraction and evap. at 47°C.	38.1791 grams
		0.0727 grams
	Weight after 2 hours in oven at 110°C.	38.1323 grams
	Essential oil	0.0468 grams

Per cent of essential oil = 2.34% wet basis or 2.55% dry basis (see calculation below)

0.0727 grams total ether extractive matter
0.0468 grams essential oil

0.0259 grams non-volatile or 1.29%

If per cent moisture by oven was	6.328
If per cent moisture by desiccator was	1.732
then total moisture in per cent was	8.060

and per cent essential oil on the dry basis is calculated as follows:

$$\begin{array}{r} 100.000 \\ -\ 8.060 \\ \hline 91.940 \end{array} \longrightarrow 2.34\%$$

$$\frac{2.34 \times 100}{91.94} = 2.55\%$$

b. *Clevenger method* (Seagram modification)

Apparatus. A flat bottomed, long necked flask of 1000 ml., a four-bulb condenser (15 in. long) and Clevenger tube, all equipped with standard taper ground glass joints comprise the apparatus. Fisher's special high temperature bath wax[1] is used in a copper bath which has round cusps with openings for the necks of the flasks only, and fits tightly in the rest of the bath so that any sweating occurring on the condenser and which could drop into the bath is not dangerous, because the hot wax is protected from the water by an entirely closed container. The bath is heated by gas burners. See Fig. 5.

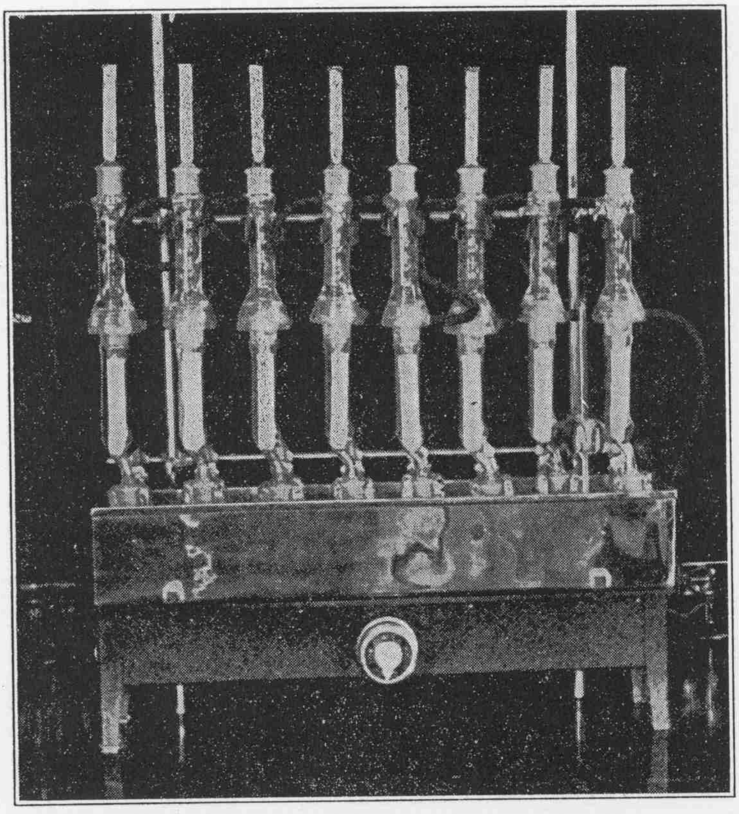


Figure 4

Seagram Modification of Johnson's Ether Extract Apparatus of Essential Oils, Also Special Type of Condenser Designed to Prevent External Sweating from Pentrating Inside of Apparatus.

Procedure. Grind the materials in the same manner as for the ether extraction method. Place 100 grams in the flask and cover with 350-500 ml. of distilled water according to absorption capacity of the material), place the flasks in the wax bath under the covers of the apparatus in such a manner that 1/3 of the flask is immersed in wax, and connect with the rest of the apparatus. See Fig. 6.

1 Fisher Bath Wax No. 15-532. M. pt. 140°F. (60°.C.) Flash point 620°F. (325°C.) Fir point 680°F. (360°C.)

The temperature is raised to 155-160°C. (68.3-71.1°F.) and kept at this level for 1½ to 2 hours. Clevenger's apparatus is an ingeneous combination of cohobation and steam distillation.

Cohobation is a process of rectification. The cohobation procedure requires the use of a special rectification apparatus heated by flame, or indirect steam. In this apparatus, the main part of the oil and also most of the water distills over in the first portion. While the amount of water decreases in the pot, the solubility of oil decreases and the oil can be separated from the solution. Thus Clevenger's apparatus utilizes the same principle — vapors of essential oil condense together with steam and fall down into the Clevenger tube with much smaller amount of water than in the flask. As most essential oils are lighter than water, this fact, in addition to separating essential oils from water, helps to exhaust the material in the flask by returning the excess water back into the flask. The oil remains at the top of the tube.

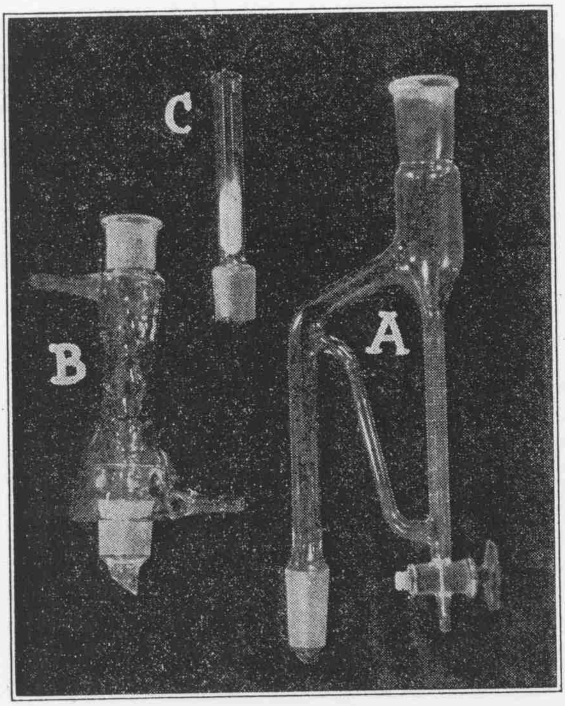


Figure 5

Seagram Modification of Clevenger Apparatus

(a) Clevenger Tube
(b) Condenser (designed to prevent external sweating from penetrating inside of apparatus)
(c) Calcium Chloride Tube (use is interchangeable between Clevenger and Johnson Apparatus; preferred for Johnson.)

When for 15 minutes there is no addition to the volume of oil, the heat may be turned off. Then the bath is cooled. For convenience the reading is done by bringing the level of the oil to 0. Amount read represents the per cent of oil on wet basis. Per cent of oil on dry basis is calculated the same as for the Seagram method, except the reading is previously divided by the specific gravity of the oil. The oil is decanted into small vials (10 ml. capacity), a pinch of sodium sulphate is added, and the vial is covered and left in the refrigerator over night. Thence the oil can be studied for its physical properties such as solubility, specific gravity, optical rotation and refractive index.

4. Contamination

Contamination is also checked by weighing accurately 3 to 5 grams of the material, taking out dirt, broken seeds and badly shaped seeds, and then again re-weighing and calculating per cent contamination.

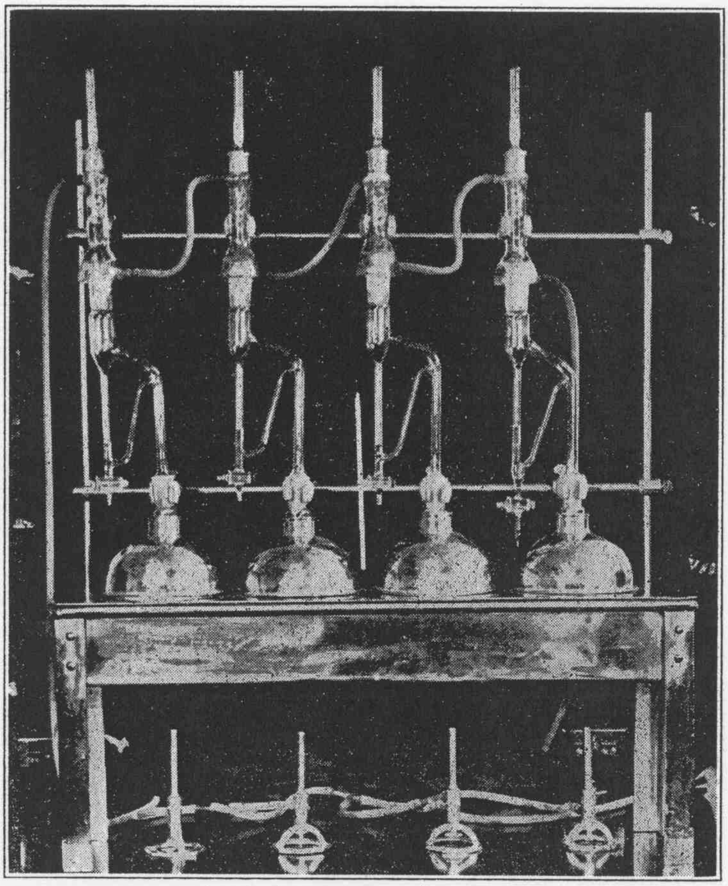


Figure 6

Seagram Modification of Clevenger's Apparatus for the Distillation of Essential Oils.

EXAMPLES OF ASSAYMENT OF RAW MATERIALS

	Coriander Seed	Caraway Seed	Anise Seed	Fennel Seed	Angelica Root	Licorice Root
Experiment No.						
Company						
Age						
Amount represented by lot						
Country	Hungary	Holland	Russia	Spain	Germany	Turkey
Moisture by desiccator	3.691%	1.573%	0.482%	0.524%	3.484%	1.545%
Moisture by oven	6.118%	8.470%	5.454%	7.946%	10.310%	6.504%
Total Moisture	9.809%	10.043%	5.936%	8.470%	13.794%	8.049%
Moisture by Dean-Stark	6.5%	8.5%	6.0%	8.0%	10.5%	7.0%
Acidity as mg. KOH per gram	1.65	4.2	6.7	6.6	4.9	2.8
Total ether extr. matter	0.4137 gm.	0.3599 gm.	0.3662 gm.	0.3965 gm.	0.1343 gm.	0.1015 gm.
Total volatile matter	0.0118 gm.	0.0626 gm.	0.0299 gm.	0.0778 gm.	0.0094 gm.	0.0010 gm.
Total non-volatile matter	0.4019 gm.	0.2973 gm.	0.3363 gm.	0.3187 gm.	0.1249 gm.	0.1005 gm.
% essential oil (dry basis)	0.61%	3.4%	1.5%	4.2%	0.5%	0.05%
% oil by Clevenger (dry basis)	0.60%	3.5%	1.4%	4.0%	0.45%	[1]
Physical properties (Solub., Sp. Gr., Opt. Rot., Refr. Index)						
Contamination %	10.0%	8.0%	6.0%	6.0%	9.0%	1.0%
Quality, accepted (+) or rejected (—)	+	+	—	+	+	+

1 Licorice root is not tested by the Clevenger method.

APPENDIX

V

BIBLIOGRAPHY

V.

Bibliography

Charabot, E., "Les Principes Odorantes des Végétaux," Paris, 1912.

Clevenger, J. F., *J. Am. Pharm. Assoc.*, *17*, 345—9,1928.

Craveri, "Les Essences Naturelles," Paris, 1929.

Durvelle, J. P., "Les Huiles Essentielles," Paris, 1908.

Finneemore, H., "The Essential Oils," London, 1926.

Gildemeister, E., and Hoffman, F., "Die Atherischen Ole," vol. I, Miltitz, 1928,
vol. II, Miltitz, 1929,
vol. III, Miltitz, 1931.

Gibbs, W. M., "Spices and How to Know Them," Matthews-Northrup Works, Buffalo, New York, 1909.

Kolachov, Paul J., "Possibilities of a Domestic Essential Oil Industry," *Manufacturer's Record*, Baltimore, Maryland, August 1940, page 30.

McNair, J. B., "Spices and Condiments," Field Museum of Natural History, Chicago, Illinois.

Naves, Y. R., and Mazuyer, C.. "Les Perfumes Naturels," Gauthier-Villars, Paris, 1939.

Parry, E. J., "The Chemistry of Essential Oils and Artificial Perfumes," vol. I and II, London, 1921.

"Pharmacopoeia of the United States of America," Philadelphia, 1935.

Redgrove, H. S., "Spices and Condiments," Sir Isaac Pitman and Sons, Ltd., London, 1933.

Sievens, Arthur F., "Methods of Extracting Volatile Oils from Plant Material and the Production of Such Oils in the United States," *Technical Bulletin, U. S. Dept. of Agriculture*, No. 16, January 1928.

Tschirch, A., "Handbuch der Pharmakognosie," vol. II, Leipsig, 1917.

Vilikovsky, V., "Agricultural Technology," (Czech), Dep't. of Agriculture of Czechoslovakia, vol. II, Prague, 1928.

Vilikovsky, V., and Kolachov, P. J., *Annals of the Czechoslovakian Academy of Agriculture*, vol. II, Prague, 1937, pages 150-6.

Willkie, H. F., Boruff, C. S., and Althausen, D., "Controlling Gin Flavor," *Ind. and Eng. Chem.*, vol. 29, Jan. 1937, page 78.

Willkie, H. F., and Kolachov, P. J., "U. S. Essential Oil Industry Urged," *The Spice Mill*, July 1940, page 39.

Wood, H. C., and LaWall, C. H., and others, "United States Dispensatory," 22nd. Centennial Ed., Philadelphia, 1932.