ences and with the desire to have uniform definitions for the different indices the French Commission proposed to unify the peroxide number and to define it as the number of milligrams of peroxide oxygen contained in a gram of sample. The Commission agreed to this proposal but because of the small quantity of oxygen decided to express it in micrograms. Hot and cold methods with and without the use of $CO_2$ were considered. It was decided that they will be tried on the semi-drying oil used for the study of the thiocyanogen number.

IX. SOAPSTOCK (PATES DE NEUTRALISATION)

The Commission decided not to take any position on the analysis of these products as they are of interest only to certain National Commissions, are not actually objects of international trade, and there is a tendency in the industry to transform them to fatty acids or true soaps. However, it is interested in the determination of neutral oil in strongly acid fatty materials. It retains four methods which will be tried on a sample of olein and on a sample of acids from the decomposition of soapstocks which will be distributed by the Swiss Commission.

**Composition of the Commission for 1947-48**

President—Dr. G. L. Voerman
Vice President—Foster D. Snell and Dr. Sturm
Secretary—J. Vizern
Members:
Denmark—K. Helholt
United States—Foster D. Snell, H. E. Longenecker, and V. C. Mehlenbacher
France—Prof. L. Margaillan, J. Vizern, G. Wolff
Great Britain—K. A. Williams, Dr. Lee, G. J. Robertshaw (observer for the leather chemists)
Italy—Prof. S. Fachini, Prof. S. Anselmi, Dr. G. Balestrini
Netherlands—Dr. G. L. Voerman, Dr. I. S. H. Bertsam, Dr. H. A. Boekenoogen
Switzerland—Dr. Sturm, Weder
Czechoslovakia—Prof. Vesely, Dr. Herites, Dr. Metzl

**Meeting 1948**

Considering that the success of the work is due in a large part to the frequent contacts between its members, the Commission decided to meet in 1948 in Paris with preference for the month of July.

PUBLICATION OF THE UNIFIED METHODS, 1947

The view is expressed that the International Union of Chemistry will publish the unified methods as adopted to date.

*(To be continued in Part II and III)*

# Minor Oil-Producing Crops of the United States

E. B. KESTER, Western Regional Research Laboratory,[1] Albany, Calif.

IN the six years since the author and G. R. Van Atta published an article under the present title,[2] new developments in the processing of minor oil-bearing materials, derivable principally from agricultural waste, have made it desirable to review the subject and include revisions in the data, descriptive matter on present practices and trends, and reports of technological investigations in the more recent literature.

The third year after V-J day finds the United States in a period of inflated prices and costs. Vegetable oil and meal prices in 1940 and 1947 are listed in Table I. Alhough 1940 and 1947 prices differ widely, showing a two- to three-fold increase, with few exceptions, for the period, the trend has been, by no means, consistently upward. During the spring of 1947 there was a general marked drop in oil prices, followed by a rapid rise to a maximum in January, 1948, and then a sharp decline along with other commodities. These broad fluctuations, of course, may continue until the postwar turbulence, which affects all phases of our national life, has subsided.

TABLE I

Oil Prices (Cents per Pound)
(From *Oil, Paint, and Drug Reporter* and from *Feedstuffs*)

| | June 1940 | Nov. 1947 |
|---|---|---|
| Corn | 8⅜ | ...... |
| Cottonseed | 9 | 32 |
| Almond (sweet) | 75 | 180 |
| Soybean | 7 | 31 |
| Linseed | 9.8 | 31.8 |
| Tung | 22 | 28 |
| Apricot kernel | 38 | 65-85 |
| **Feed Prices (Dollars per Ton)** | | |
| | May 1940 | Nov. 1947 |
| Cottonseed (43%) | 32.50 | 93.00 |
| Coconut | 21.00 | 78.50 |
| Linseed | 24.00 | 83.00 |
| Soybean | 38.50 | 103.00 |

However unstable may be our present scale of prices, the existence of enormous tonnages of agricultural wastes that pollute streams, create health hazards, and are a liability to the food manufacturer, suggests with some insistence that something should be done to reclaim at least a part of their values. The observer, speculating on what he can recover or make from such wastes, would probably consider one or more items in the following list:

| | |
|---|---|
| Feed | Plastic filler |
| Substrates for microbiological processes | Tannin |
| Syrups | Mucilage |
| Vegetable oil and meal | Pectin |
| Citric, tartaric, or other organic acid | Furfural |
| Vitamins | Charcoal |
| | Fertilizer |

He may then attempt to sample a waste and have assays made to learn how much useful material he might recover and, with the data, estimate his gross returns per unit of weight. At this stage disillusion often comes, as he multiplies a number of conversion factors and ends with a pitifully small percentage of starting material in the form of salable goods. The vegetable oils in waste often hold more promise than some others in the list, as they usually involve mere segregation of the oil-bearing part, such as seeds, followed by expression or extraction, but even in this case the various economic factors bearing on a proposed enterprise should be carefully considered

[1] Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.
[2] Oil and Soap *19* (7), 119-125, July, 1942.

before a decision is made to set up a plant. In no instance has it been possible to process a single waste product for oil and obtain a fair return on the investment without the support of other salable merchandise such as feedstuffs or fertilizers. Minor oils are usually recovered in plants capable of handling a variety of materials or in which oil extraction is only incidental to other operations.

The geographic location of processing plants for waste, with respect to the supply, is exceedingly important, because haulage costs (often on a high-moisture-content material) are frequently a deciding factor between profit and loss. The amount of raw material that is obtainable at centralized locations and can be counted on from year to year, is important, as well as the costs of separating and drying (if necessary) the fractions bearing oil. Then there is the problem of properly storing bulk quantities of materials with a minimum of decomposition, which is sometimes difficult to solve. The size of the oil plant itself should be given serious thought. A plant too small is uneconomical because of excessive labor costs, and a large plant is equally bad if a given waste, obtainable in the radius of economic haulage, is only sufficient for short-time operation, unless the same equipment can be utilized for recovery of other oils. Solvent extraction *vs.* expression must be evaluated in terms of type of material under consideration. Low-oil-content, highly abrasive substances like rice bran cannot be handled to best advantage in either screw or hydraulic presses. For solvent extraction, an investigation of the various types of manufactured units should be made (1).

The prospective processor of oil-bearing materials, in computing probable returns from his products, must be concerned with quality and feeding value of his press cake and by-products of his operations. The Feed Control Laboratory, State of California Bureau of Field Crops, Sacramento, has issued a table of analyses covering a number of minor feed materials including grape pomace meal, grape pulp and seed, mustard oil cake meal, olive oil cake meal, prune pits, almond hulls, avocado seed cake meal, tomato pomace, walnut oil meal, rice huller bran, rice polish, rice hulls, raisin oil cake meal, almond hulls, date pits, orange and other meals, in addition to the more common feedstuffs. Analytical data for protein, fat, fiber, and ash are given in each case.

It is possible, under prevailing prices, to realize a substantial profit from certain oil-bearing wastes that were unattractive raw materials in pre-war years. Grape and tomato pomaces are cases in point. These wastes are nuisances which manufacturers are glad to dispose of at little or no cost and, when converted to oils and feed, they return a profit in spite of high labor and processing charges and solve a serious disposal problem.

The sources of minor oils to be considered in this paper are not only those derivable from the wastes which accumulate at canneries, wineries, and drying yards but also seeds and fruits that, under favorable economic conditions, may be grown and processed for oil. In this category we find olives, avocados, sunflower, safflower, rape, and castor. The latter group may be considered potential oil crops of considerable bulk, but thus far they have been of only minor importance in our fats and oils economy.

## Pit and Nut Oils

The pit oils of the apricot, peach, prune, and plum bear a striking resemblance to each other in composition and indeed are not readily differentiated. Cherry pit oil is reported to have a somewhat higher iodine number. These oils are like sweet almond oil, for which they are often substituted (2). If the pits are processed promptly after removal from the fruit, little or no benzaldehyde will be liberated into the oil by hydrolysis of amygdalin. The pit oils command a premium price over the commoner vegetable oils and are usually handled by essential-oil brokers.

Olive, date, and avocado pits are not discussed in this paper. In the pressing of olives for oil the whole fruit, including pit, is crushed. Date pits from the quantity of fruit that is currently pitted would yield a negligible volume of oil, and avocado pits contain less than 2.5% of ether extract on the dry basis (3). In the canning of pitted olives the pits are a waste product for which it would be desirable to find an outlet. The oil content of 3.5 to 7.0% (4) is probably too low for profitable recovery. Inclusion in animal feeds has been suggested, but such a material, unless well ground, would undoubtedly contain hard, sharp particles which might cause injury to the digestive tract.

Tree nuts, although rich sources of oil, must be considered primarily as luxury food items, and edible grades cannot therefore be pressed economically except to fill small demands for specialty oils. Shelling plants, however, reject large amounts of moldy and inedible meats which are entirely suitable for oil recovery. The shells contain a considerable percentage of unseparated meats, which are reclaimed by reducing the shell in hammer mills and subsequent screening.

Although tung nuts are not ordinarily classed with fruit pits and certainly not with edible tree nuts, they are included in this section because they bear a resemblance to both. The development of an industry from domestically grown nuts is still in its early stages, but crops are increasing as new acreage comes into bearing. Between the years 1936 and 1940 new plantings raised the acreage of tung orchards from 75,000 to more than 175,000.

Reported percentage composition figures for fruit pits and tree nuts, as well as characteristics of the oil in each, are included in Table II.

*Almond.* Comparatively little almond oil has been produced in this country. One company in the Los Angeles area is pressing and marketing specialty oils at this time, including small amounts of almond oil. When the shelled nuts, which contain about 50% of oil, are dried, coarsely ground, and cold pressed, about 75% of the oil is removed at 450 pounds of pressure. The oil is low enough in free fatty acids to be refined to a high-grade salable product by the use of bleaching agents alone. American almond oil has an iodine value of about 103, which is slightly higher than U.S.P. specifications; nevertheless it is commonly accepted by the trade.

The meal, containing about 20% of oil, is edible but slightly bitter because of the skins. It is ground, sieved to various sizes, and sold to the cosmetic and baking industries.

Commercial production of almonds in the United States is confined to California. Their production is

TABLE II

Pit and Nut Oils

| Kind | Pit in Fruit, % | Kernel in Pit, % | Oil in Kernel, % | Characteristics of Oil | | | | |
|---|---|---|---|---|---|---|---|---|
| | | | | I.V. | Composition of Fatty Acids | | | Unsaponifiables, % |
| | | | | | Oleic Acid,% | Linoleic Acid,% | Sat. Acids,% | |
| Apricot | 5.6 | 20-25 | 40-47.4 | 100-108.7 | 63.3 | 31.1 | 3.7 | 0.7 |
| Cherry | 12-15 | 28 | 32-40 | 92.8-122 | 49.0 | 42.0 | 8.1 | 0.3-0.7 |
| Peach | 7.5-12 | 5-8 | 40-45.8 | 96-110 | 57.5-62.5 | 15.7-20.9 | ........ | ........ |
| Plum | 2.3-6 | 5-26.7 | 30-42.9 | 100-105 | 72.0 | 22 | 6 | 0.4 |
| Prune | 12.5 | 21 | 33-41.5 | ........ | ........ | ........ | ........ | ........ |
| Almond | ........ | 33-70 | 48.9-60 | 95-107 | 77 | 17.3-19.9 | 3.1-5.7 | ........ |
| Filbert | ........ | 40-48 | 50-60 | 84-90 | 78.2-88.1 | 2.87-9.1 | 4.9-8.0 | 0.3-0.55 |
| Pecan | ........ | 47-60 | 70-75 | 100-106 | 70.9-77.8 | 15.8-25.2 | 4-5.1 | 0.4 |
| Persian Walnut | ........ | 27-42 | 60-70 | 132-161.7 | 17.6 | 73.0 Linolenic, 3.2 | 5.3 | 0.5 |
| Tung | about 50 (air-dry basis) | 63.8-67 | 40-62.7 | 160-170 | 3.9-16.4 | 0.6-9.7 Elaeostearic, 74 5-86.3 | 2.6-8.9 | 0.4-0,8 |

a major industry in certain sections of the state although the total crop represents only about 8 or 9% of the world crop. From the standpoint of acreage almonds rank third among the deciduous tree fruits and nuts of California, being exceeded only by prunes and walnuts. About 95,000 acres are in bearing at present and recent plantings will increase this acreage within a few years. Although the market has thus far easily absorbed the entire crop, which has amounted to as much as 23,800 tons (1944 crop) in recent years, the industry is conscious of the possibility of surpluses, particularly in view of the increasingly large amounts of Spanish nuts being imported, and is seeking to find possible new outlets for its product. In 1944 imported almonds amounted to 37,500 tons, a figure in excess of any prewar imports.

High market prices make it difficult to devote first-grade nuts to oil production. In shelling and grading plants, however, a noteworthy amount of shriveled, resinous, or otherwise inedible meats is rejected and could be used for oil recovery. There is also a noteworthy quantity of bitter almonds, brought in by growers from trees planted many years ago because of their supposed value in pollinating sweet varieties. Such almonds find little use except as seed for root stocks. In experiments conducted at the Western Regional Research Laboratory the fixed oil from the bitter almonds was found to have a bitter flavor and odor but could be rendered quite bland by ordinary methods of deodorization.

About 60% of the almond industry of California is represented by one cooperative. Approximately two-thirds of the annual U. S. almond crop is shelled. Nearly 10% of the meats, which amount to nearly 650 tons, are moldy, shriveled, or otherwise inedible and contain 40% of oil, which means that about 260 tons of oil is annually available from this source. The present price quotation on almond oil ($.80 to $1.10 per lb.) is considerably less than in prewar years as a result of competition with imported almond oil.

*Persian Walnut.* The amount of walnut oil produced in this country is about 300 to 600 tons annually. Raw material for oil pressing is divided between inedible meats rejected during shelling and meat fragments recovered from shell. The former have a slightly lower oil content (55.60%) than edible meats. One plant in Los Angeles produces most of the walnut oil manufactured and obtains its raw material principally from the shelling plant of a large cooperative. Screw-press equipment is used. The shell is reduced to flours of various screen sizes, which are sold as soft grit for blasting of metals, plastic filler, an ingredient of non-skid paints, and as a carrier for agricultural insecticides.

Walnut oil is similar to linseed oil and before the war was sold entirely to the coating industry. During and after the war it has been consumed as a specialty food oil. The outlet for walnut press cake, which contains a considerable amount of shell, is mainly the fertilizer industry although some was used for stock feed a few years ago.

About 90% of the English or Persian walnuts grown in the United States are produced in California; the other 10% are raised in Oregon. The California crop is divided about equally between northern and southern counties. The majority of the growers are members of an association which manages the disposal of the crop.

Production of walnuts in the United States for the period 1939-44, inclusive, averaged 63,350 tons, of which about 23,000 tons, or over 36%, were shelled. In years preceding the war when supply exceeded demand, some edible-grade nuts were pressed for oil, but current production is entirely from rejected meat.

*Pecan.* Pecans are grown in a number of southern states, but principally in Georgia, Oklahoma, and Texas, which accounts for about two-thirds of the national crop (61,000 tons, 1941-45 average). About 300 tons of pecan oil have been produced annually for the past two or three years. As in the case of walnuts, the source material is the discarded meats from shelling plants, which are shipped to a central point for processing. The oil is hydraulically pressed. It has been found fairly high in free fatty acids (8-12%) and some difficulties were experienced at first in refining it, but these have been overcome. The oil has brought from 40 to 50c a pound. Press cake is treated to recover tannin, of which it contains about 14%, and the residue is sold for stock feed.

*Filbert.* Commercial raising of filberts is almost wholly confined to Washington and Oregon. The crop for the five-year period, 1941-45, averaged less than 6,000 tons. Filbert oil is similar to olive oil, having a content in combined oleic acid of 85%, but the amount of inedible meats rejected from shellers would be so small that recovery of significant amounts of the oil would not be practicable unless the acreage devoted to the raising of the nuts was considerably expanded. One cooperative agency has a membership of about 3,500 producers, who grow about two-thirds of the filberts and 70% of the walnuts raised in the Pacific Northwest.

*Tung Nuts.* Commercial production of tung or China wood oil from domestic nuts is a relatively new industry in this country. The first oil mill was established in 1928, but the crop at that time was small and supplied only a minor fraction of our requirements of this valuable drying oil. Most of our needs were met by imports from China directly or indirectly. In 1939 the entire domestic crop of nuts was 1,160 tons, representing about 230 tons of oil, but during the war years when foreign supplies were almost completely cut off, the crop increased manyfold from new acreages coming into bearing and averaged 25,410 tons for the period 1941-46. The price per ton since 1940 has almost doubled, reaching a record high of $102 in 1944. The 1946 crop of 57,400 tons or the equivalent of almost 12,000 tons of oil was the largest on record. Production figures are estimated in terms of air-dried nuts in the husk.

The producing area for tung trees occupies a belt roughly 100 miles wide extending along the Gulf of Mexico from Central Florida through Georgia, Mississippi, Alabama, Louisiana, and into southeastern Texas. Up to the present, Mississippi is the most important producing state, representing about two-thirds of the total annual production.

The important characteristic of tung oil is its high content in combined elaeostearic acid, which contains three double bonds in conjugated relationship and contributes the unique drying characteristics of this oil. Tung oil is obtained principally by expression methods. The press-cake is poisonous to cattle and is suitable only for fertilizer.

*Apricot Pits.* Nearly all of the apricot industry is located in certain well defined areas of California such as the Sacramento, Santa Clara, and San Joaquin Valleys and a few of the southern counties. Washington and Utah also raise apricots commercially, but their combined crop is less than 11% of the national total. The average crop for the five-year period, 1941-45, was about 219,000 tons of which more than 80% was dried, canned, or otherwise processed. Many of the larger growers dry their own fruit and are able to sell the pits at prices as high as $55 per ton. At least four plants in California shell apricot pits to recover kernels, for which there is a strong domestic market. Current prices are 21-23c a pound. The dry-pit equivalent for processed apricots in this country is about 10,000 tons, which would yield about 2,500 tons of kernels. In the cracking process a considerable amount of broken kernels result, which are used to recover oil. The annual production of apricot pit oil is about 100 tons a year. The oil has always sold for a high premium price, is now listed at 65-85c a pound, and is used in the cosmetic and pharmaceutical industries as a substitute for sweet almond oil.

To recover apricot kernels and oil, the pits are first air dried, then cracked between steel rolls; then the mixture of shell and kernels is introduced into a brine tank of such density that the shell sinks and kernels float. The kernels are skimmed off, washed, dried, and sorted. The whole kernels are ultimately used in making bakery goods, such as macaroon paste, after the amygdalin is hydrolyzed and large portions of the benzaldehyde and hydrogen cyanide are driven off. Apricot pit oil is recovered in screw-press or solvent extraction equipment. It is easily refined and bleached to a very pale product.

*Prune Pits.* No prune-pit oil is produced at present although small amounts have been pressed in years past. It is similar to apricot- and peach-pit oils. The prune industry differs from the apricot industry in that the bulk of the crop is dried without removal of the pits. Some prune products other than the dried fruit are now sold, but these are of small volume. About 80% of the national prune crop (594,000 tons, 1941-45 average) is raised in California, the remainder coming principally from Washington, Oregon, and Idaho. Over 90% of the total production is dried, canned, or otherwise processed. Oil from the pits of all the prunes processed would amount to more than 5,000 tons, but it is useless to consider prune pits as a source of oil unless present marketing practices were to change in favor of pitted prunes. Agnes Fay Morgan of the Home Economics Division, University of California, College of Agriculture, found that prune pit oil is relatively rich in vitamins A and E.

*Peach Pits.* Growing of peaches is a widely distributed industry. Leading states in this respect are New York, Pennsylvania, Michigan, Virginia, South Carolina, Georgia, Washington, Oregon, and California, which account for about 70% of the total United States production.

Of the entire crop (1,600,000 tons, 1941-45 average), about one-third is canned, dried, frozen, or made into jams, preserves, jellies, and spirits. California accounts for 90% of the processed peaches in the United States, and about half of its own peach crop is processed. The bulk of the peach crop in eastern and middle western states is marketed as fresh fruit, and it is only in California that one could consider peach pits as a source of oil and other by-products.

Disposition of peaches by type of product and method of processing in California appears in Table III (average data for 1941-45). The total of these figures is 531,018 tons, which represents about 1,400 tons of kernel oil.

TABLE III
Disposition of California Peach Crop
(Average for 1941-45)

| | Canned, Tons | Dried, Tons | Frozen, Tons | Other, Tons |
|---|---|---|---|---|
| Clingstones | 359,480 | 13,520 | 2,414 | 1,562 |
| Freestones | 24,660 | 123,500 | 5,400 | 482 |

At the present time peach pits can be obtained from canneries and drying yards for little or nothing except cost of hauling. High-proof spirits are made from the juice extracted from clingstone pits, and the pit itself is converted into char for poultry feeds and fuel briquets. Very little peach kernel oil is extracted for the following reasons: 1. most of the pits available for by-products are from canners' clings, which in the original wet condition yield less than 1% of oil, and 2. modern canning practice in many plants utilizes a saw which cuts the entire peach, including pit, in two, badly crushing the kernel and making it difficult to recover the fragments.

It will be noted that while clingstone varieties are favored for canning, freestone peaches are preferred for drying. The pits are usually obtained from the latter in whole condition, which renders them more suitable for cracking and recovery of kernels. Brine flotation is used as for apricot pits. Eleven tons of dry freestone peach pits yield about one ton of kernels. Oil may be recovered in screw presses, but in the one plant now operating in California on peach pits hydraulic presses used in hot pressing of olive oil are employed.

*Cherry Pits.* Types of processed cherries marketed pit-free that will be considered here are canned, frozen, miscellaneous packs of sour varieties, and also brined sweet and sour cherries. The major states in production of processed cherries in these classifications are Michigan, New York, Wisconsin, Washington, Oregon, California, and Pennsylvania, which account for 85% (78,000 tons, 1941-45 average) of the total. California raises no sour cherries commercially, but there is extensive brining of sweet varieties. The oil represented by the total of pitted cherries amounts to about 1,100 tons per year, but no plant is at present recovering any of it. One company in Wisconsin is known to have produced a few tons of cherry kernel oil a few years ago. Another used cherry pits as a source of "oil of bitter almonds" produced by steam distillation of the ground meats.

*Plum Pits.* The amount of plums processed per year amounts to about 4,000 tons (1937-46 average). The pit oil represented by this quantity of fruit is insignificant. The principal areas where canning of plums is practiced are Michigan and California.

## Fruit Pulp Oils

The oils to be considered under this heading are those from avocado and olive. The latter is a particularly important oil of commerce, although supplies obtainable from olive growing in this country are not large when compared with world production. The two oils are quite similar in their high content of combined oleic acid. Reported percentages of oil in the fruit and composition of the combined fatty acids of olive and avocado oils are found in Table IV.

TABLE IV
Fruit Pulp Oils

| Kind | Oil | I.V. | Characteristics of Oil — Composition of Fatty Acids: Oleic Acid,% | Linoleic Acid,% | Sat. Acids,% |
|---|---|---|---|---|---|
| Avocado | 3-21 | 70.6-94.4 | 77.4 | 10.75 | 7.2 |
| Olive | 15-60 | 74-94 | 69.1-84.4 | 3.9-12.0 | 9.3-17.2 |

*Avocado.* The culture of avocados in the United States is confined to Florida and California, the latter state accounting for 83% of the total production (20,270 tons, 5-year average, 1941-45). From 1,000 to 6,000 tons a year are imported, principally from Cuba during the summer months. Commercial plantings in California are located almost entirely in climatically favored areas in the southern part of the state, particularly in the coastal belt from San Diego to Santa Barbara.

As the avocado is a luxury fruit, most of the crop is sold at prices which make oil recovery from any considerable portion of it out of the question. It is only in the spring that a small tonnage can sometimes be utilized for this purpose. The cull portion of the crop ordinarily represents only about 1% but occasionally may be considerably higher because of adverse conditions such as the heavy freeze of 1937.

The oil content of avocados may vary from 3 to 21%. The normal range is about 9 to 15%. The percentage of oil is a criterion of grade, being highest in fruits considered best for table use. Avocados are reputed to have a high content of vitamin A, which naturally passes into the oil, and the oil itself has been found to contain sufficient vitamin D to be protective against rickets (5). Because of the pulpy character of the fruit avocado oil is not easily recovered by expression methods unless a fibrous medium such as peat moss is added, which of course increases retention of oil in the press cake. A process recently developed at Lindsay, California, for recovery of olive oil has been successfully used on avocados. With this method the fruit is pitted and macerated with water in a ball mill; the resulting slurry is screened and centrifuged to separate the oil. Yield is approximately 25-30 gallons of oil per ton of fruit, or about 10-12%.

Avocado oil has been reported to have a definitely favorable effect on skin disorders, such as eczema (6), but it is not sold and formulated for this purpose. The current market price for the oil is from $1 to $1.10 a pound. It is sold almost exclusively to the cosmetic trade.

*Olive.* Olive oil occupies a unique position among vegetable oils in that it is valued for its distinctive odor and flavor as well as for its low melting properties and adaptability to many cosmetic and pharmaceutical formulations. Unlike the common bland oils, such as cottonseed and corn oils, high-grade virgin olive oil is not alkali-refined or steam-treated but merely water-washed and clarified. If the fruit has been properly stored prior to crushing, the oil will have a low content of free fatty acids.

California accounts for almost the entire U. S. production of olives (99.8%). Major producing areas are the San Joaquin and Sacramento valleys and a group of southern counties, particularly Los Angeles, Riverside, San Bernardino, and San Diego. Principal varieties grown are the Mission, Manzanillo, and Sevillano. About half the acreage is planted to Mission, which is a fairly good variety for oil. In prewar years California produced only 5 to 10% of our national olive oil requirement. Olives devoted to this purpose have never yielded the domestic grower as high a price as those sold for canning, but demand became so great when Mediterranean supplies were shut off that the price of the former rose from about $75 a ton in 1940 to about $220 a ton in the early months of 1947. Olive oil prices in this period rose from about $2 to about $8 a gallon. From 1941 to 1945 inclusive about 50% of the total olive crop was used for oil production. With resumption of olive oil imports oil prices have begun to decline.

The oil content of ripe California olives ranges from about 10.7 to about 33.7%, averaging 18 to 25%. Of the total present about two-thirds is recoverable as "virgin" oil. Table V summarizes U. S. production, utilization, and import figures for olives and/or olive oil through the years 1939 to 1945 inclusive.

Many attempts were made during the years of olive oil shortage to extend the limited amounts

TABLE V
Olives and Olive Oil
(U. S. Production, Utilization, and Imports, 1939-45)

| Year | Production, Tons | Av. Price, Dollars | Crushed for Oil, Tons | Utilization Canned, Tons | Other, Tons | Oil Imports | |
|---|---|---|---|---|---|---|---|
| | | | | | | Edible Tons | Inedible, Tons |
| 1939 | 23,000 | 74.00 | 6,100 | 12,200 | 3,700 | 32,300 | 18,360 |
| 1940 | 69,000 | 76.20 | 39,100 | 16,200 | 12,000 | 10,850 | 7,420 |
| 1941 | 55,000 | 153.00 | 28,300 | 16,700 | 9,400 | 3,200 | 1,550 |
| 1942 | 57,000 | 122.00 | 34,200 | 11,100 | 10,100 | 3,250 | 420 |
| 1943 | 57,000 | 178.00 | 28,600 | 15,500 | 11,900 | 480 | 70 |
| 1944 | 42,000 | 200.00 | 19,800 | 13,800 | 7,600 | 3,850 | 50 |
| 1945 | 30,000 | 269.00 | 9,800 | 13,500 | 6,400 | ........ | ........ |

available from the California crop. No doubt considerable adulteration with cheaper oils occurred as well as addition of refined hot-pressed or solvent-extracted olive oils to virgin grades. One process for producing an olive-flavored oil was publicized and practiced, in which olive paste was first macerated with a cheaper oil, such as cottonseed or corn oil before pressing (7). This process was claimed to provide a blend carrying more of the typical olive-flavor than would be possible by merely mixing a bland oil with olive oil.

In addition to the conventional hydraulic pressing which has been used for many years to recover olive oil and the centrifugation process described in the section on avocado oil, a number of other methods have been operated or proposed, several of which are reviewed briefly in Dr. G. S. Jamieson's book on vegetable oils (8).

## Seed Oils

More numerous and variegated than the oils from the pits and fleshy portions of fruits are those from a host of miscellaneous seeds which are or could be by-products of food-processing industries. Included in this list are several oil-bearing seeds which are raised for oil production (although in small amounts compared with soybeans and flaxseed, for instance) or used for other purposes such as feed. Others are still in the development stage. Omitted are the seeds of fruits and vegetables which are sold mainly in the fresh condition or, if processed, without removal of seed from a substantial portion of the product. Of those omitted watermelon, cantaloupe, and fig seeds are examples. Table VI summarizes reported figures for percentage of seed in fruit or vegetable, kernel in seed, oil in seed, and characteristics of the oil.

### Seeds of Fruits

*Citrus Seeds.* In the years 1941-45 an average of 19% of the oranges, 53% of the grapefruit, and 30% of the lemons grown in the United States were processed for juice, concentrates, fruit segments, and other citrus products. These figures, however, include noteworthy amounts of seedless grapefruit and oranges.

Of the four states (Florida, Texas, Arizona, and California) that produce grapefruit in commercial quantities Florida and Texas by far exceed the other two, but only Florida processes seeded varieties in tonnages large enough that the seeds might be considered a source of vegetable oil. The average tonnage of seeded grapefruit processed in Florida during the five seasons, 1941-42 to 1945-46, was 476,600 tons, of which the dry seed equivalent, at 1.9%, is 9,050 tons. This would be capable of yielding 2,850 tons of oil, based on 31.5% for recoverable oil in the seed. At the present time a plant is operating at Lakeland, Florida, for manufacturing citrus by-products, which includes the recovery of seed oil in considerable amounts.

Of the oranges processed in this country only the Valencias need be considered as sources of oil-bearing seed. For the period 1943-44 to 1945-46 the processed Valencias (annual averages) were divided as follows:

| | |
|---|---|
| California | 219,000 tons |
| Arizona | 65,500 tons |
| Florida | 377,460 tons |
| Total | 661,960 tons |

The seed content for oranges is greatly variable, but 0.3% is a fairly good average figure. Based on this factor, the seed equivalent for the total of the Valencia oranges processed is about 1,986 tons, which at 40% would yield about 790 tons of oil.

In a study at the Western Regional Research Laboratory (9) it was learned that orange-seed oil could be refined, bleached, and deodorized to a pleasing bland product, useful for foods and other purposes. In composition of glycerides it resembles cottonseed oil.

Lemons, unlike oranges, are not a seasonal crop. Fruit may be found on a given tree in all stages of ripeness. Seed content of lemons is an exceedingly variable quantity, depending on climatic conditions during ripening. It is an interesting fact that a cold snap or freeze will be followed, after a fixed time interval, by a crop of lemons having an abnormally high seed content.

Lemon seeds cannot be counted on as a prolific source of oil. The tonnage of processed fruit is subject to wide variation from year to year, and, as the seed content is also variable, a figure for potential annual oil yield is difficult to reach. Based on the three-year average, 1943-44 to 1945-46, a possible oil yield of about 540 tons can be computed if it is assumed that the content of dry seeds is 1.25% and the recoverable oil in the seed is 31.5%.

*Grape Seeds.* More than 90% of our national grape crop is grown in California, where the 500,000 acres in vineyards is more than double that devoted to oranges. Here we find principally the Vinifera or Old World type of grape whereas the Great Lakes region, particularly New York and Ohio, is a center for the production of the Labrusca or slip-skin type, of which the Concord is representative.

Although California grapes are divided into three main classes (wine, table, and raisin varieties), the distinction between them is by no means absolute. For example, any variety of each of the three groups is suitable for wine, but the so-called wine varieties, with rare exceptions, are suitable for wine only. Raisin varieties are the most versatile as they are also fine table grapes and are used plentifully for the manufacture of wine, brandy, and for canning.

Grape seeds that result from commercial processing, and therefore may be considered sources of oil and other by-products, are to be found in the wastes from both the raisin and wine industries, particularly the latter. In former years, raisins were made almost exclusively from seed-bearing grapes, principally Muscat, and large quantities of seed were rejected from drying plants and used for making brandy, oil, tartrates, and stock feed. When the Thompson seedless was developed, the tonnage of Muscats converted to raisins dwindled, and in 1946 less than 27,000 tons of this variety were dried. In terms of vegetable oil this is equivalent to about 120 tons. Another factor that has drawn Muscat grapes away from the raisin field is the demand for them in the manufacture of Muscatel wine.

The flourishing wine industry in California has made available large quantities of grape pomace, from which seed can be readily separated by either dry or wet methods. At least three mills in California are now producing grape-seed oil by extraction from the seeds of wine pomace. It has been estimated that on the basis of an annual crush of a million tons of grapes the quantities of recoverable oil would be in the neighborhood of 3,200 tons if all the pomace were processed and all the seeds obtained therefrom were extracted.

The wine interests of California for a number of years have been increasingly interested in possibilities of by-product manufacture from their pomace piles, which have constituted a serious disposal problem. The commonest practices in the past have been to dry and burn the pomace or use it directly in the vineyards for fertilizer, but neither of these is satisfactory. If pomace piles stand idle for even a short time, putrefaction sets in and they become a health hazard to the community. Certain factors are important in a consideration of the economics of by-product recovery from a low-grade, high-moisture material like pomace, chief among which is the geographical distribution of the wine industry. The following pertinent facts are therefore set down as an aid in evaluating this factor:

1. Of a total of more than 400 wineries in California, about 60 account for 80% of the annual crush. All of these handle in excess of 1,000 tons of grapes per year. In one district, the southern San Joaquin Valley, 90% of the wine and brandy coming from that area is made by the larger wineries.
2. There are five major producing districts in California: the northern San Joaquin and Sacramento Valley, southern San Joaquin Valley, North Coast, Central Coast, and southern California.
3. The total crush for the Sacramento and San Joaquin Valley districts is about 80% of the state total.
4. The total crush, excluding Thompson seedless grapes, by the larger wineries in the Sacramento and San Joaquin Valley districts is 43% of the state's total for all grapes crushed and 62% of the total seed-bearing grapes crushed.
5. The oil represented by the pomace from the larger wineries in the San Joaquin and Sacramento Valley districts is about 2,000 tons per year, and from all wineries in those districts, about 2,400 tons.

TABLE VI

Oils of Various Seeds

| Kind | Seed in Fruit, % | Kernel in Seed, % | Oil in Seed, % | Characteristics of Oil | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | Composition of Fatty Acids | | | | |
| | | | | I.V. | Oleic Acid, % | Linoleic Acid, % | Other Unsat. Acid, % | Sat. Acids | Unsaponifiables, % |
| Orange | 0.3 | 62-70.8 | 34.2-45 | 98-105.4 | 36.6 | 36.5 | Linolenic, 0.6 | 26.3 | 0.95 |
| Grapefruit | 1.9 | 77 | 28-35 | 101-106 | 19.7 | 48.8 | | 26.6 | 0.7 |
| Lemon | 0.5-2 | ........ | 30-35 | 103-109 | ........ | ........ | ........ | ........ | ........ |
| Tomato | 0.5-0.6 | ........ | 18-23 | 107-125 | 45 | 34.2 | ........ | 18.3 | 0.7 |
| Grape | 3.5-4 | 54 | 6.8-22 | 105-160 | 10.0-34.5 | 54-71.4 | ........ | 8.4-15.0 | 0.8-2.8 |
| Squash | 1.5 | ........ | 36.6 | 121 | 36.6 | 43.3 | ........ | 18.9 | ........ |
| Pumpkin | 1.5 | ........ | 30-35 | 120-130 | 25 | 45 | ........ | 30 | ........ |
| Sunflower | ........ | 54-61 | 27.5-30.8 | 122.4-136.6 | 21.2-39.1 | 51.2-68.3 | ........ | 9.7-12.8 | ........ |
| Safflower | ........ | 47.6-52.4 | 24-33.8 | 143.3-149.8 | 14.7-17.3 | 76.6-79.0 | ........ | 5.0-6.7 | ........ |
| Apple | $<1$ | 63.5-71.5 | 18.0-25 | 104-122.4 | ........ | ........ | ........ | 6.6 | 1.1-1.7 |
| Pear | $<1$ | 60.5-69.3 | 21.4-25 | 124.1 | ........ | ........ | ........ | 9.8 | 1.0 |
| Rice bran | ........ | 8-9.5 (Bran in rice) | 8.7-20.4 (In bran) | 92-109 | 38.7-45.3 | 26.4-42.0 | ........ | 14.8-21 | 0.7-5 |
| Watermelon | ........ | ........ | 20-30 | 133.8 | 13 | 63.4 | ........ | 15.2 | 1.2 |
| Okra | ........ | 50 | 12.8-22.0 | 91.1-100.3 | 41.5-43.7 | 25.5-29.7 | ........ | 28.8-30.0 | 1.1 |
| Mustard (white) | ........ | ........ | 25-30 | 94-106 | 28 | 19.5 | Erucic, 52.2 | 4 | ........ |
| Castor | ........ | ........ | 35-55 | 82-90 | 7-9 | 3-3.5 | Ricinoleic, 80-86 | Dihydroxy stearic, 2.1 | 1.8 |
| Tobacco | ........ | ........ | 30-46 | 117.8-151.6 | 12.0-30.2 | 54.6-77.3 | ........ | 7.4-15.2 | 0.6-3.1 |
| Cranberry | ........ | ........ | 20 | 169.2 | ........ | ........ | ........ | ........ | ........ |
| Asparagus | ........ | ........ | 12.8 | 137 | ........ | ........ | ........ | ........ | ........ |
| Wild plants. | | | | | | | | | |
| Brassica arvensis (Charlock oil) | ........ | ........ | 25.0-25.6 | 121 | ........ | ........ | ........ | ........ | ........ |
| Brassica juncea | ........ | ........ | 31.8-34.0 | 129 | ........ | ........ | ........ | ........ | ........ |
| Tumbling mustard | ........ | ........ | 32.6 | 151 | 5.2 | 19.0 | Palmitoleic, 1.0 Erucic, 25.3 Linolenic, 34.0 | 14.1 | 0.5 |
| Hare's ear mustard | ........ | ........ | 29.5-36 | 99-107 | 20-30 | 15-20 | Erucic, 35-45 | 4 | ........ |
| Pennycress | ........ | ........ | 32-35 | 117.3-123.2 | 12.1-12.5 | 32-33 | Erucic, 49-53.4 | 2.5-5 | 0.8-1.5 |
| Gopher plant | ........ | ........ | 47.3 | 82.0-84.7 | 84.3-90.6 | 2.3-7.0 | ........ | 5.7-7.7 | ........ |
| Wild cucurbits | ........ | ........ | 28-31 | 122.7-131 | ........ | 37-43 | ........ | ........ | 1.1 |
| Jojoba | ........ | ........ | 48-50.8 | 81.7-86.3 | 0.7 | ........ | Eicosenoic, 30.3 Dicosenoic, 14.2 | 1.6 | 48.3 |
| Cattail seed | 35-40 | ........ | 17.9 | 130.8 | 11.6 | 73.6 | ........ | 14.8 | ........ |
| Pinus monophylla | ........ | ........ | 21 | 102.1 | 56.7 | 31.6 | ........ | 8.5 | 1.9 |
| Pinus sabiniana | ........ | ........ | 11.5 | 120 | 50.5 | 45.2 | ........ | 4.3 | ........ |

The northernmost of the two areas mentioned (the northern San Joaquin and Sacramento valleys, not including the counties of minor importance) is an area roughly 80 miles long and about 50 miles wide. The southern San Joaquin district is about 40 by 100 miles in extent. Between is the county of Merced, in which wine production is relatively small. It is logical, therefore, that separation of grape seeds and recovery of oil has developed in these two areas. In the southern there are already two solvent plants which extract grape seeds, besides other oil-bearing materials, and another close to the northern district. Some lots of grape seeds were brought into the San Francisco area during the war years when oil supplies were short, but the results obtained with screw presses were not favorable. It has been our experience at the Western Regional Research Laboratory that ground whole grape seeds cannot be handled well in this type of equipment. Oil yield is low and the cake tends to clog at the choke ring. Decorticated seeds, however, were quite satisfactory and contain upwards of 30% oil. The solvent extraction of grape seed as now practiced is not wholly satisfactory because of difficulty in filtering the miscella.

At the present time only a fraction of the available wine pomace in the two districts mentioned above is worked up for oil and by-products. The skins and portions of the pomace other than the seeds are dried and sold as a low-grade stock feed.

Grape-seed oil is excellent for culinary purposes. A product obtained from raisin seed is used to coat packaged raisins to make them free flowing. Although a wide range of iodine values from all sources are reported in Table VI, American grape seeds usually yield an oil between the limits of 125 and 137. It has been used together with other oils in paint formulations.

*Apple Seeds.* Seventy per cent of domestic apples used in processing are raised in the Western and North Atlantic states. The portion of the annual apple harvest converted to canned, dried, frozen, and miscellaneous products is about 620,000 tons (1943-45 average), which, at 0.25%, would yield 1,550 tons of seed, the oil equivalent of which at 25% is 385 tons. Economical recovery, however, would probably be limited to the larger districts mentioned above, in which case the maximum oil recovery would be only 270 tons. Only large packing centers would be justified in separating seed from waste and would in most instances dispose of the seed to oil mills.

Apple-seed kernels contain about 1% of amygdalin (10) and are therefore a potential source of "oil of bitter almonds." It has been reported that apple-seed oil contains a muscle regulating substance, probably a vitamin, but no confirmation of this statement has been found in the literature.

*Pear Seeds.* California and Washington are centers of production of pears for canning, drying, and other uses, accounting for 90% of the national total or 240,000 tons (1944-45 average). The output of processed pears in California is roughly double that of Washington.

The seed content of pears is greatly variable, but if the same figure (0.25%) is used as for apples in computations, the amount of recoverable oil represented by all the pears processed in the two states is about 150 tons.

Pear-seed oil is very similar to apple-seed oil in iodine value and saponification value, but no determinations have been made on composition of the fatty acids. Unlike apple seeds, pear seeds contain not more than a trace of amygdalin (11).

*Cranberry Seed.* Principal producing states for cranberries are Massachusetts, New Jersey, Wisconsin, Washington, and Oregon. The two western states account for only about 3 to 4% of the total. Average total production of cranberries for the five-year period, 1941-45, was 32,500 tons. Of this amount only 11,000 tons were made into seed-free manufactured products (principally sauce), the seed equivalent of which was about 110 tons containing about 22 tons of oil. The oil is said to be rich in vitamin A (12).

By-product manufacture from cranberry wastes is carried on by a cooperative, operated by grower members in the three largest producing states, at a plant in South Hanson, Mass. More valuable than the seed oil is the ursolic acid in the skins, which is potentially available to the extent of 10-15 tons a year (13).

Ursolic acid in the form of its sodium salt is a powerful agent for producing water in oil emulsions (14).

## Oils from Seeds of Vegetables and Other Domestic Plants

*Rice.* As a supplementary bulk oil of commerce, the oil of rice bran is much greater in potential volume than any other described in this paper with the exception of tobacco seed oil. It occurs in the bran in the amount of 15% although practical yields are about 10-12. In general characteristics it resembles cottonseed oil.

Annual production of rice in the United States amounts to about 1,400,000 tons (1941-44 average). The area devoted to its cultivation is fairly large, 1,450,000 acres, and is confined to relatively few geographical districts: the San Joaquin and Sacramento valleys of California, southwestern Louisiana, eastern Texas, and Arkansas. Practices in cultivating, harvesting, and milling vary somewhat with the locality, variety of grain, and season, and these differences may be reflected in variations in the quality of both products and by-products.

Rice is usually milled at or near its point of origin. Milling consists in dehulling, separation of the bran from the endosperm, and finally polishing the kernels to yield the familiar white milled rice of commerce. The fractions of product and by-products are obtained in the following ranges:

| | |
|---|---|
| Hulls | 17-21% |
| Bran | 8-9.5% |
| Polish | 1-3% |
| Milled rice (including broken grains) | 64-68.5% |

The oil of rice is concentrated almost entirely within the bran, which is also a storehouse for vitamin B. The bran is used widely in poultry feeds but it is not entirely acceptable because of its tendency to become rancid in a short space of time. The oil is affected and develops large quantities of free fatty acids. It is common knowledge at rice mills that whole rice which has been adjusted to a moisture content of about 15% withstands storage very well, but if the hull is removed, lipolytic and/or oxidative changes in the bran oil begin to take place immediately, and this is true of both stored brown rice and rice bran. Extracted bran, on the other hand, can be stored quite well and is more acceptable as a constituent of feeds than the oil-containing bran. In more than one instance it has been used as a source of B vitamins.

Rice bran can be solvent-extracted without pretreatment of any sort. A number of plants produce rice bran oil, including at least three in California. Although refining losses are almost invariably high because of rapid development of fatty acids, the oil, if obtained from bran of sound grain and promptly extracted after milling, is reputed to be quite stable and is excellent for culinary purposes. It has long been used in the Orient.

Several types of physical and chemical treatments for preventing deterioration of the oil fraction of rice have been proposed, but to date none of them has been found practical for large-scale commercial application. No lipase has been isolated from the bran although it is quite evident that some substance is present that acts like a lipase. The amount of rice bran oil potentially available is from 16,000 to 20,000 tons (computed from the average rice crop for 1941-45).

A recent trend in rice milling has been the production of so-called processed rice. This is an "infused" rice; the unhulled grain is treated with hot water and steam to an extent that a portion of the oil and vitamins is driven from the bran into the endosperm. When the hulls and bran are subsequently removed, the product is lightly colored and vitreous in appearance. In the debranning and dehulling, breakage is considerably less, probably because of gelatinization of the starch granules to such an extent that about a third more first-grade saleable rice is obtained than in the case of white rice. It is finding wider consumer acceptance. The necessary equipment for producing the material is simple in construction and inexpensive to operate, and the increased returns obtainable from the finished product more than offset the additional operating cost. This discussion is introduced to indicate a development in the technology of rice processing which is reflected in the changed character of the by-products and more particularly the bran and bran oil, which are rendered more stable, that is, less susceptible to rancidification.

*Sunflower Seeds.* Up to the present time production of sunflower seed in the United States has been quite limited (2,400 tons per year, 1941-45 average). Localities in which it is grown are principally California, which accounts for about 80% of the total, and Missouri (15%). Small amounts are raised in Illinois. The oil equivalent of the total sunflower seed crop is about 360 tons, but all of the domestic crop at present is consumed entirely in feeds. During the war sunflower seed oil was imported in large quantities, principally from Argentina, and used largely in the manufacture of shortenings. In postwar years these supplies have fallen off.

Considerable experimental development in the raising of sunflowers has been going on in Canada (15) for a number of years and more recently in the United States, particularly Illinois. The investigation by Milner *et al.* (16) includes a comparison of four common varieties of sunflower, which shows no large differences in the oil content of whole seed, but noteworthy variations in iodine value and corresponding variations in the content of oleic and linoleic acids (Table VI).

*Safflower Seeds.* Repeated attempts have been made to establish safflower as an oil-seed crop in the United States, but with varying degrees of success. Yields per acre in test plantings have been low in a number of instances although near Prosser, Washington, 50 to 75 bushels were obtained in one instance (17) and 70 bushels in western Nebraska under conditions of irrigation (18). Rabak (19) reported as high as 40 bushels per acre without irrigation in the northern great plains under favorable climatic and cultural conditions, and as high as 50 to 65 bushels under irrigation. Low yields (20 bushels) in the Yakima, Washington, district have been reported (20). Central and eastern Nebraska (25 bushels) (18) as well as the area near Havre, Mont. (7 to 12 bushels) (21), seem poorly adapted to safflower. For a number of years small acreages of safflower were grown in the area near Deming, New Mexico, and in 1947 about 500 tons were produced in eastern Washington and northern Idaho.

Safflower *(Carthamus tinctorius)* is a glabrous herb yielding seed weighing from 40 to 48 pounds per bushel. It has been cultivated in India and Egypt for many years. The seed, as well as the press cake from oil extraction, is a valuable stock feed.

The oil content of safflower seed is somewhat variable, depending on variety as well as soil and climatic conditions. Although the range of 24 to 33.8% covers the usual oil content, percentages as low as 10.44 have been reported (21).

Extensive tests have been made on the use of safflower oil, which has drying properties, in coating compositions. Carrick and Nielsen (22) reported that safflower oil blended with linseed improves resistance to weathering. The oil, when heated for 150 to 180 minutes at 310-315°C., polymerizes to a stiff gel soluble in turpentine (23). In India this material is known as "roghan" or "afridi wax."

*Tomato Seeds.* Tomato production for manufactured products averaged 2,893,000 tons in the five-year period 1941-45. If it is assumed that seeds represent 0.55% of the whole tomato (8) and contain 15% of recoverable oil (estimate from one solvent extraction plant in California), the potential annual supply of tomato seed oil is about 2,400 tons. The press cake from tomato seed is rich in protein (30-40%) and is acceptable as a stock feed.

In years past the Department of Agriculture has investigated tomato seed as a source of fixed oils and the general subject of utilization of tomato waste. These studies included the important incidental factors, such as handling and sorting tomato waste, cleaning of the seed, extraction and refining of the oil, cost analysis, and possible returns (24). At the time of this investigation only 400 tons of tomato-seed oil were potentially available; since then, as already indicated, the amount has risen considerably and the general economic picture is greatly different. One vitally important factor is the crying need today for some means of disposing of tomato waste other than discharging it into streams where it causes widespread pollution. Ordinances against such practice are becoming increasingly severe, and tomato processors are seeking means whereby the pomace can be converted into products that will at least off-set the cost of handling and processing. A number of plants at present merely dry and sell whole tomato pomace for feed. In at least one instance seed is commercially separated from skins and sold for oil recovery. Both of these materials have been found to have a carotene content greater than 350 p.p.m.

Freshly expressed tomato seed oil is brownish to reddish in color and has a strong odor. After caustic refining, bleaching, and deodorizing, a pale yellow product is obtained which is entirely suitable for culinary purposes.

The tomato processing industry is widespread. Canneries are located in many parts of the country. This lack of centralization, together with the fact that the potential supply of tomato seed oil is small, means that an oil recovery set-up in connection with any one plant would most likely be uneconomical to operate unless other oil-bearing materials are also available for processing.

*Squash and Pumpkin Seeds.* Indiana is the principal processing center for pumpkin and squash (25-30% of national total). Illinois accounts for about 15-20% and California, 10-15%. Other states where canning and/or freezing of these vegetables is practiced are Ohio, Washington, Oregon, and New Jersey. The total annual pack in terms of fresh produce is about 82,000 tons (8-year average, 1937-44) from which about 1,230 tons of seed would be obtainable, the oil equivalent of which is about 430 tons.

No pumpkin or squash seed oil is being produced at the present time in this country, but in Europe it has been recovered and used for edible purposes. A nut-like delicacy is made by roasting and salting the kernels of pumpkin seed.

*Okra Seeds.* Commercial plantings of okra as an oil-seed crop are not yet a reality although several of the southern states, most notably Louisiana and Texas, have been experimenting with the raising of this plant and have reported favorably on yields. Indeed, the results are so promising that farm machinery companies are already developing special combines for harvesting it. Although related to cotton, okra is said to resist attack by the boll weevil and other insects, except nematodes. Separation of hulls and kernels of okra seed is somewhat difficult as the seed is small. Solvent-extracted or expressed oil from crushed whole seed has a greenish color, but this is readily bleached. The oil is reputed to have good stability after refining and hydrogenation.

An excellent discussion of okra seed oil by W. R. Edwards, Jr., and J. C. Miller appeared in the Chemurgic Digest for January 31, 1947. This article discusses use of the oil as an edible fat, feeding value of okra seed meal, processing of okra seed and its products, agricultural economy of okra seed production for industrial uses, and varieties and possibilities of new varieties of okra seed with high oil content.

*Asparagus Seeds.* On the San Joaquin and Sacramento river delta area of California are grown from 65,000 to 70,000 acres of asparagus annually; about 750 to 1,000 lb. of seed per acre could be obtained, or a total of approximately 26,250 tons from which about 3,400 tons of oil are extractable. At the present time none of this oil is being recovered. It has an iodine value of about 137 (25). Land devoted to asparagus culture in California is about one-half the national total. The next largest producing state is New Jersey where about 22,000 acres of asparagus are raised annually.

*Mustard Seed.* Considerable amounts of the Oriental variety of mustard are grown in Montana, the largest mustard-producing state in the country. This variety has an oil content of about 30% and yields a press cake that is not suitable by itself for condiment manufacture but may be incorporated as a filler with condiment mustard. The cake contains about 30% protein but is quite bitter, and, if used in stock feeds, it must be kept below 5%. It is sold principally for fertilizer.

Other varieties of mustard grown principally for condiment manufacture are the brown and yellow mustards, which have oil contents of about 25 and 20% respectively. The larger manufacturers express the oil as a preliminary step in the making of food products. Proportions of the three varieties grown in 1946 were: Oriental, 33.6%; yellow, 54.9%; brown, 11.5%.

Mustard seed production in the United States amounts to about 21,500 tons per year (1941-45 average), having an oil content of about 6,500 tons. The oil, because of its high percentage of combined erucic acid, is frequently used in place of rape oil for special lubricants. Although Montana is the principal producing state, accounting for about 78% of the total crop, significant amounts are grown in California, Washington, Oregon, and North Dakota, which produce mainly the yellow and brown varieties.

*Rape Seed.* The United States imports as much as 15,000 tons of rape-seed oil a year to supplement its meager supplies of high-erucic-acid oils. Although some rape seed is grown in northern Idaho and eastern Washington, it is still a minor crop. The Canadian production, which is sold almost exclusively to the United States, is steadily increasing. The 1947 crop, estimated at 14,600 tons, represents a 66% increase over 1946.

*Tobacco Seeds.* An approximation of the potential supplies of tobacco seed oil in this country is difficult because of differences in varieties as well as cultural practices. It has been estimated by Riemenschneider *et al.* (26) that flue-cured tobacco, which is grown on about 876,000 acres would yield 80 to 320 lb. of seed per acre, merely from the sucker growth which develop after the seed heads are topped. On the basis of these minimum and maximum values the potential seed crop would be 35,000 to 140,000 tons, of which the oil equivalent is 12,750 to 51,000 tons. The latter figure is more than three times the potential yield of the next largest minor oil described in this paper (rice bran). Riemenschneider and co-authors mention the possibility of growing *Nicotiana rustica* as a source of nicotine and drying oil. The Brasilia variety, without topping, has produced 100 lb. of nicotine and 1,000 lb. of seed per acre on the west coast. The oil, according to this author, is unusually stable considering its high unsaturation.

Analytical data for tobacco seed oil (Table VI) have rather wide ranges. The limits include values reported by all investigators. It should be mentioned that American tobacco seed oils usually have an iodine value above 135 and a content of combined linoleic acid in excess of 70%.

*Castor Beans.* In the early days of homesteading, raising of castor beans was practiced extensively in scattered localities in the Midwest. By 1850 there were 23 castor-oil mills in the United States located in Illinois, Missouri, Virginia, Tennessee, Pennsylvania, Alabama, and Arkansas (28). St. Louis was the most important commercial center. Another mill, the largest of its kind, was erected in Jersey City in 1857 and began to handle imported seed. Domestic production reached a peak in 1879 and supplied more than the demand. Thereafter growing of castor declined and in some states, Kansas for example, stopped almost entirely. Meanwhile imports began to increase, and today we rely entirely on foreign shipments, particularly from Brazil, which supplies upwards of 90% of our requirements. Table VII summarizes briefly the past trend in domestic production and imports of castor beans.

TABLE VII

U. S. Production and Imports of Castor Beans

| Year | Production, Tons | Imports, Tons |
|---|---|---|
| 1880 | 12,000 | 1,200 |
| 1900 | 3,300 | 3,400 |
| 1920 | 10 | 15,000 |
| 1940 | ........ | 154,000 |
| 1944 | ........ | 171,000 |

The value of castor oil as a lubricant for aircraft was recognized in World War I and large amounts were produced and expressed for this purpose.

With the advent of World War II there arose an urgent need for castor oil, particularly by the armed forces, both as a lubricant and as an hydraulic fluid in the operation of mechanized equipment, and, when dehydrated, as an ingredient of quick drying finishes. In the early years of the war the U.S.D.A., through the Agricultural Adjustment Administration, fostered the planting of castor, and several thousand acres in Texas were devoted to this purpose. Tests in the raising of castor were made in other localities (28), and much valuable information on the culture, selection, and adaptation of varieties was obtained. It was learned that the region where soil, rainfall, growing season, and freedom from disease are most favorable is the Ohio Valley and areas extending southwesterly into Kansas, Missouri, Arkansas, Oklahoma, and Texas. In Florida successful test plantings have been made by a commercial concern. In the latter *Ricinus Communis Africanus* was most satisfactory and under normal conditions yielded 600-900 lb. of beans per acre.

With the termination of the war the raising of castor beans as a commercial crop has practically ceased, yet so much interest is still manifested in this commodity by both government and industry that it was thought desirable to include a discussion of it in this paper.

The raising of castor beans domestically in competition with foreign supplies is difficult, principally because of the labor involved in harvesting. In most strains of castor the capsules holding the beans either drop or shatter when ripe, and for that reason mechanical harvesting has not replaced hand picking. This problem has been intensively studied in the University of Nebraska Chemurgic Project.

Castor is a versatile plant. Its leaves yield ricin, a potent insecticide, and from the stalks a quantity of bast fiber and alpha-cellulose are obtained. Castor oil is unique in being composed for the most part of glycerides of ricinoleic acid (12-hydroxyoleic acid). It has a high density and a low congealing point. It is used widely as a constituent of hydraulic brake fluids, as a low-temperature lubricant, and as a liquid cushion in artillery recoil mechanisms. Its miscibility with low-molecular-weight alcohols is an important property in many applications of this sort.

Acylated castor oil or ricinoleic esters are valuable plasticizers. The sulfonated oil is the well known turkey red oil, which has long been used in the dyeing of cotton textiles with alizarin. By appropriate chemical or pyrolytic treatment castor oil may be converted to a number of useful compounds such as sebacic acid, undecylenic acid, octanol-2, heptaldehyde, and 12-hydrostearic acid.

## Oils from Seeds of Wild Plants

Although wild plants cannot be considered agricultural crops in any sense of the word, nevertheless the oil-bearing potentialities of certain species are sufficiently great to warrant a short discussion of them in this article. The number of wild plants whose seeds contain oil, often in high percentage, is legion. In certain instances cultivation holds definite promise of substantial returns, particularly when the plants in question are resistant to drouth or insect infestations and bear prolifically. Some attempts are in progress at agricultural experiment stations and also in industry to grow various wild plants under the most favorable conditions, and to establish by direct experiment the possible returns derivable therefrom, but these developments are slow to mature.

Few wild oil-bearing seeds growing in the native state are collected today except for food use in local areas. In almost every instance the cost of collecting enough of the seeds at a sufficiently low cost to justify processing is prohibitively high. When, on the other hand, such seeds are more or less constant by-products of another crop, as in the case of grain screenings, and can be obtained without added expense, the picture is quite different. Many of the wild oil-bearing seeds of the United States have been discussed by Dr. Jamieson (8). A few of the more promising ones from the standpoint of the industrialist are included herein.

*Jojoba.* The jojoba *(Simmondsia Californica)* is a desert shrub indigenous to the southern parts of Arizona, New Mexico, and California, where it grows plentifully. Many of the Indians living in these areas gather the jojoba beans and sell them in local markets. Eventually enough of them are assembled in one place to justify shipment to an oil mill for processing.

Jojoba beans are unique among oil seeds in that they do not contain the familiar glyceride type of oil but instead a liquid wax consisting for the most part of long-chain unsaturated esters of long-chain unsaturated fatty acids and alcohols, $C_{20}$ and $C_{22}$, being the predominant length in both. It is considerably more stable to heat than most glyceride oils. Although the oil or wax is non-nutritious, it has a number of important non-food uses. It is readily hydrogenatable to a hard wax, m.p. about 73-4°C., suitable for furniture, floor, and automobile polishes. The beans, containing more than 50% oil, can be processed well in screw-type equipment, which will express the oil down to about 5% in the cake. The latter contains more than 30% protein and is quite acceptable as a feed material.

The wax has been suggested for use as a lubricant for certain purposes (14) and as a transformer oil in the electrical industry (29). The mixture of long-chain alcohols derived from jojoba wax by saponification has been patented as an ingredient of strongly emulsifying shortenings (30). Patents have been issued on the use of sulfurized jojoba oil as an ingredient of quick-drying printing inks (31), as a linoleum base (32), and in the compounding of extreme pressure lubricants (33).

One experimental planting of jojoba was undertaken a few years ago by an industrial concern, but no decisive results on this venture have yet been reported. The shrub is dioecious; that is, staminate and pistillate flowers are borne on separate plants.

*Grain Screenings.* At least two plants in the United States, one at Great Falls, Mont., and one at Fresno, Calif., press or extract grain screenings brought down from western Canada. The screenings are collections of miscellaneous weed seeds together with some broken grain. Anderson *et al.* (34) have made a comprehensive study of the weed seeds in both cereal crops and flaxseed, collected in terminal elevators at Fort William and Port Arthur, including a determination of the oil content of both the individual components and the screenings as a whole. Cereal screenings contained about 14 kinds of weed seed, the principal ones being lamb's-quarters, wild mustard, stinkweed, and hare's ear mustard. The amount of total foreign seeds and of individual species varied widely in different shipments. Refuse screenings from flax contained principally wild mustard, stinkweed, broken flax, hare's ear mustard, and tumbling mustard. The current price paid for grain screenings is about $40 per ton, and the amount of oil recovered by the Montana plant alone in a season is about 1,100 tons (35).

The screenings from cereal grains, cleaned of straw, chaff, and other foreign matter, showed an oil content of about 14.6% with an iodine value of 131. Flax screenings contained about 21.4% oil with an average iodine value of 143. The higher oil percentage and higher iodine value for the flax screenings, of course, are due to the considerable amounts of broken flax seed present. Meal from the pressing of grain screenings can be fed to cattle if mixed with other feeds.

*Wild Mustard.* The wild mustard in screenings is made up of the seed of both *Brassica arvensis* and *Brassica juncea.* The latter is a plant cultivated in India for oil production (36). The proportions of the two show considerable variation in different grain shipments, but the effect of the variation on the constants of the oil is not great, as the total amount of wild mustard present does not exceed 31%, frequently is considerably less, and the iodine value of one is only eight points removed from that of the other (34).

Hare's ear mustard seed oil has been the subject of an investigation by Hopkins (37), who found it had a content in combined erucic acid of 35-40% and an iodine value of 107. The percentage of oil in the seed was reported to be 29.5. Slightly different figures were found by Johnson and Greaves (38).

Tumbling mustard seed (39) has an oil content of about 32.6% and an iodine value of 151. The percentages of combined fatty acids found are listed in Table VI. The low value for erucic acid, 25.3% as compared with that of other mustards, should be noted.

*Pennycress.* This weed, also called fanweed, French weed, dish mustard, treacle wart, stink weed, bastard cress, devil weed, and Mithradate mustard, is a principal constituent in the screenings of wheat grown in the northwest and north central states. It is a member of the Cruciferae, to which rape and mustard also belong, and contains, as might be expected, a large amount, about 49%, of combined erucic acid. Oil content in the seed is about 31-35% (40). Test plantings conducted at Montana Agricultural Experiment Station have shown that on irrigated land 1,500 lb. of pennycress seed per acre can be obtained. Harvesting difficulties are encountered, however, in that the seeds do not mature evenly and the pods tend to shatter badly.

Pennycress seed oil is a potential substitute for rape oil, which finds considerable use as an additive in special lubricants, particularly when blown, because of its marked ability to adhere to metal surfaces under heavy loads and to resist flushing by water. Such lubricants find wide application in the operation of both diesel and steam engines. The possible use of pennycress oil in coating compositions has been partially investigated (41).

The residual meal, after the oil has been extracted, is comparable in protein content with undecorticated cottonseed meal and nutritionally is quite adequate as a stock feed. It has the disadvantage, when fed to dairy cattle, of imparting an unpleasant flavor to the milk unless ingestion takes place a number of hours before the cows are milked.

*Gopher Plant.* Gopher plant, also called caper spurge and oily milkweed, is a biennial commonly found in fields and wasteland areas. The seeds yield about 47% of an oil with an exceptionally high content of oleic acid (42), highly laxative in the crude form. Indeed, it is reported to have three times the laxative action of castor oil (43). Private test plantings have indicated the seed might be obtained in yields as high as 1,500-1,800 lb. per acre (44).

From the amount and kinds of fatty acids present (Table VI) there is no indication that the glycerides are responsible for the oil's physiological action, and therefore the possibility exists that it might be suitably refined for edible purposes. The press cake, however, is either poisonous or obnoxious to animals and could not be considered as feed material.

*Wild Cucurbits.* In the arid region of the Southwest three wild gourds grow abundantly: *Cucurbita foetidissima, Cucurbita palmata,* and *Cucurbita digitata.* The plants are perennials and produce large crops of fruit, the seeds of which are rich in oil and protein (45). The first of these was the subject of a study by Wood and Jones in 1943 (46); the other two were investigated by Ault and co-workers (47). The important constants for the oils as shown in these references are included in Table VI. An unusual constituent of the fatty acids of these oils is a conjugated trienoic acid occurring in amounts of 10% or more. The acid is not identical with the elaeostearic acid of tung oil. Curtis (45) estimated that the wild cucurbits might be capable of yielding 1,500 lb. or more of seed per acre.

*Typha (Cattail).* Processing plants with a capacity to handle in excess of 1,500,000 lb. of cattail heads annually are being operated in Minnesota and Wisconsin to remove the fibrous portion for the manufacture of insulation and shock absorbing materials.

The seeds represented by this quantity of cattail heads amount to about 600,000 lb. and would yield about 50 tons of oil (48). In the event that all the available cattail seed in the U. S. were to be extracted, about 17,000 tons of oil could be obtained although, of course, the harvesting of a considerable portion of this wild crop would not be practical. Typha seed oil has an unusually high content of free fatty acids, the cause of which is unknown but may be the weathering of the seeds prior to extraction.

*Pinon Nuts.* Pine nuts of various kinds are harvested annually in considerable amounts in western and southwestern states by Indians and consumed as food. The Forest Service has encouraged this practice in projects for the development of stands of pine with maximum sustained nut production.

In Utah, Nevada, and California, *Pinus monophyllis,* and in New Mexico, eastern Arizona, and southern Colorado, *Pinus edulis* are found in great abundance. Pine nuts from European species are imported into eastern states under the name pignolias.

No reliable data are available on the total potentialities of this wild crop or the practicality of collecting and processing the nuts for oil as they are a food item and hence are probably of more value for this purpose than as a source of oil. One estimate of the harvested pine nuts in the southwestern states is 4,000 tons a year, the oil equivalent of which is about 2,500 tons.

## Conclusion

This discussion of minor oil-bearing crops by no means includes all that were considered and might conceivably have deserved treatment or at least passing mention. Omitted were seeds contained in agricultural products from which, in all likelihood, oil could never be obtained economically. Justification for such reasoning can in most cases be found in the scattered status of an industry processing the commodity from which the oil bearing part is derived, or the low magnitude of processing operations *vs.* that of retail sales. For example, the seeds of watermelon, cantaloupe, and a wide assortment of berries were not included. A vast number of seeds of wild plants could have been discussed, but unless concentration of growth occurs either naturally or as a result of domestication or unless the derived oil seems to have outstanding characteristics, such as that of gopher plant seeds or jojoba beans, it is useless to consider them seriously as oil sources. Certain grain screenings such as water grass, which occurs plentifully in rice, were omitted because their oil content is too low for profitable recovery.

The evaluation of oils must usually be made on the same basis as the commoner bulk oils of commerce which they most closely resemble. As a rule vegetable oils differ from each other only in their contents of individually combined acids and their distribution in the oil as triglycerides or in having a high percentage of an unusual acid such as elaeostearic or hydnocarpic acid. Important variations, however, may occur in content of minor constituents such as oil-soluble vitamins, antioxidants, or prooxidants, and it must be recognized that no oil can be completely evaluated until quantitative measurements covering such substances have been made where their presence is indicated.

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# The Keeping Quality of Elaïdinated Fats*

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DURING a long experience with elaïdinated fats (1) we always noticed that these fats showed a remarkably enhanced keeping quality as compared with ordinary or hydrogenated oils and fats. One of the causes of this behavior is obvious as elaïdinated oils contain considerably more solid glycerides (more in fact than most hydrogenated oils of an equal melting point), which means of course that their percentage of liquid unsaturated oil is less. In a solid fat the oxidation rate will largely depend on the amount of diffusion of the oxygen from the surface to the unsaturated (most liquid) components so one of the causes of better keeping quality (i.e. the larger percentage of solids) is purely *physical*. There may, however, as well be other causes of a *chemical* nature. We may suppose that:

1. Traces of Selenium originating from the elaïdination process may act as an antioxidation catalyst.
2. Known positive oxidation catalysts such as traces of metals like Fe, Al, Ni, etc. may have been eliminated in the course of the elaïdination with Se followed by the usual refining.
3. The elaïdo configuration of the unsaturated acids may prove to be more resistant towards oxidation than the oleic configuration (cis-trans isomerism).

The first supposition may be tested by comparing the rates of oxidation (by means of peroxide formation) of elaïdinated oils containing Se and the same oils after complete elimination of this element. The second supposition might be checked by comparing the velocity of peroxide formation of a selectively hydrogenated oil containing traces of metal and that of the same oil after treatment with selenium and elimination of this element. As selective hydrogenation also causes elaïdination to the equilibrium stage, the only difference between these two oils will be the presence of metal traces in the first. The third hypothesis may be tested by comparing the oxidation rates of an oil or fat before and after elaïdination, provided these fats do not contain traces of metals or selenium.

Now, some investigators (2) have shown that liquid oleic acid is considerably more readily oxidized than solid elaïdic acid. This, however, does not prove necessarily that the space configuration of these acid molecules is of any influence since it might merely be a consequence of the physical condition of both acids, as pointed out by us in the beginning of this article. To prove the influence of space configuration one must compare both the elaïdic and the oleic forms in the liquid condition.

## Experimental

The following oils (for particulars see Table I) were used:

1. Soya bean oil.
2. Elaïdinated soya bean oil containing some Se.
3. Elaïdinated soya bean oil, deselenised.
4. Hydrogenated soya bean oil.
5. Elaïdinated hydrogenated soya bean oil containing some Se.
6. Elaïdinated hydrogenated soya bean oil, deselenized.
7. Palm oil.
8. Elaïdinated palm oil, deselenised.

TABLE I

| Oil | p.p.m. Se | Setting point | Iodine value (Hanus) |
|---|---|---|---|
| 1. Soya bean | 0 | ...... | 126,5 |
| 2. Soya bean elaïdinated | 1105 | ...... | 126. |
| 3. No. 2 deselenised | 0 | ...... | 126. |
| 4. Hydrogenated soya bean | 0 | 30,6 | 74,5 |
| 5. Hydrogenated soya bean, elaïdinated | 470 | 30,6 | 74,5 |
| 6. No. 5 deselenised | 1,5 | 30,6 | 74,5 |
| 7. Palm oil | 0 | 24,8 | 52. |
| 8. Palm oil elaïdinated deselenised | 0 | 33,1 | 51,5 |

All oils were refined, elaïdinated with Se, and deselenised in the usual way. Immediately after deodorization they were subjected to oxidation by blowing dry air at room temperature through them, using an orifice 1 mm. in diam. at 80° C., and a rate of 500 cc. per minute. The peroxide formation was measured at intervals of time by estimating the Lea Index (1 gr. fat, 10 cc. $CHCl_3$, 15 glacial acid, under $CO_2$ 1 cc. saturated KI solution; titration with 0,01 n thio sulphate solution after standing 5 minutes and dilution with 70 cc. water; soluble starch used as

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