Physicochemical Properties of Citrus Essential Oils from Florida

Richard W. Wolford,* James W. Kesterson,[1] and John A. Attaway

Among the most significant byproducts of the massive Florida citrus industry are the several essential oils which are of major consumer and economic importance. Investigations leading to the production of Florida citrus oils of highest quality and uniformity are reviewed with emphasis on the chemical and physical properties of the various types of oils. Some of the factors shown to affect the chemical and physical properties of expressed citrus oils were fruit variety, degree of maturity, seasonal variations, storage of fruit prior to extraction, seasonal rainfall, budwood, rootstock, method of extraction, and yield of oil. The chemical compounds identified to date in both Valencia orange and grapefruit cold-pressed oils are listed. Brief mention is made of the more recent use of spectrophotofluorescence in the identification and classification of citrus oils, along with ultraviolet spectra and evaporation residue relating to method of extraction and geographical origin.

The processing of citrus fruits is a very important part of Florida's economy. In conjunction with the processing of citrus fruit into products such as frozen concentrated, canned, and pasteurized "chilled" juices and other juice products, a very significant byproduct industry has developed over the past 25 years. In 1969–1970, approximately 128 million boxes of oranges, 90% of the crop, and approximately 23 million boxes of grapefruit, 62% of the crop, went into processing channels. Among the most significant byproducts of this massive processing industry are the several essential oils which are of major consumer and economic importance.

Many investigators have shown that the quality of citrus oils is dependent upon several factors. Some of these are soil, climate, method of extraction of the oil, weather, maturity, and variety of the fruit, each of which has some direct bearing on both the physical and chemical properties of the several citrus essential oils from Forida and elsewhere.

Citrus oils are contained in oval, balloon-shaped oil sacs or vesicles located in the outer rind or flavedo of the fruit. Winton and Winton (1935) described the exact location of these oil sacs in their discussion of the microscopic structure of the flavedo of the orange. Hood (1916) found a wide variation in the oil yield of Florida oranges, reporting values of 0.11 to 0.58% with reference to the weight of the whole fruit. He stated that the oil content did not reach its maximum until the oranges were fully mature, but was present in quantities feasible for extraction before the fruit was ready for harvest. He also noted a decrease in oil content immediately after a period of rainfall.

Bartholomew and Sinclair (1946) studied the effect of age, size, and environment on relative amounts of oil in California oranges. Hendrickson *et al.* (1969) showed that maturity brought about an increasing quantity of oil per unit of surface area as well as per unit of fresh weight for Florida Valencia oranges. The yield of oil from various citrus cultivars has been presented. Additional studies of effects of budwood and rootstock on peel oil content of Valencia oranges were reported by Hendrickson *et al.* (1970).

Methods of oil extraction used in Florida were investigated by von Loesecke and Pulley (1939). They showed that the method of extraction had an effect upon the physical characteristics of the oil. However, they did not find any relationship between the time of year and physical characteristics.

The physical and chemical characteristics of Florida orange, grapefruit, tangerine, and lime oils have been reported by many early investigators. Foote and Gelpi (1943) noted variations in the properties of different lots of Florida orange oil.

Nelson (1934), Nelson and Mottern (1934), and Markley *et al.* (1937) have carried out investigations relative to the chemical constituents of orange, grapefruit, and tangerine oils produced in Florida.

When oranges were kept in cold storage for periods longer than 6 weeks previous to extraction of the oil, de Villiers (1930) found an increase in specific gravity, optical rotation, iodine number, and saponification value, but a decrease in the aldehyde content of the oil. Kesterson and Hendrickson (1953, 1971) provide a comprehensive review of studies relative to the physicochemical and related properties of orange, grapefruit, tangerine, lemon, lime, Murcott, and tangelo oils produced in Florida.

Deterioration of orange oil, as well as the effects of antioxidants, has been investigated by Kesterson and McDuff (1949) and Kesterson and Hendrickson (1951a,b), Proctor and Kenyon (1949), Kenyon and Proctor (1951), and Flores and Morse (1952). Bacteriological contamination of some citrus oils during processing has been investigated by Murdock and Hunter (1970).

In recent years with developments in gas chromatography, many investigators have done extensive work to elucidate the chemical composition of citrus oils. More than 200 different chemical compounds have been found in the orange oil, of which more than 100 have been identified.

Fundamental information relative to all types of essential oils produced throughout the world is found in Perry (1908) and in Gildemeister and Hoffman (1928–1931). In 1952, Guenther completed publication of six volumes on the essential oils bringing the whole subject up to date. In the present publication emphasis is placed on a partial review of the physical and chemical characteristics of some of the types of Florida citrus essential oils as they are affected by methods of processing, maturity, environmental conditions, fruit variety, rootstock, and budwood.

EXPERIMENTAL

Methods of Analysis. The physical and chemical properties of the original oils and the 10% distillates were determined following the official A.O.A.C. methods (1970). Specific

State of Florida, Department of Citrus, P.O. Box 1088, Lake Alfred, Florida 33850
[1] University of Florida, IFAS, Agricultural Research and Education Center, P.O. Box 1088, Lake Alfred, Florida 33850

Table I. Analyses of Florida Cold-Pressed Valencia Orange Oil Extracted by Four Different Processes[a]

	Pipkin roll	Screw press	FMC rotary juice extractor	Fraser–Brace excoriator
Specific gravity, 25° C/25° C	0.8423	0.8420	0.8431	0.8441
Refractive index, $n^{20}D$	1.4721	1.4722	1.4725	1.4730
Refractive index, 10% distillate, $n^{20}D$	1.4711	1.4711	1.4712	1.4713
Difference	0.0010	0.0011	0.0013	0.0017
Optical rotation, $\alpha[^{25}D]$	+97.16	+96.69	+96.19	+96.10
Optical rotation, 10% distillate, $\alpha[^{25}D]$	+97.52	+97.25	+97.21	+97.61
Difference	+0.36	+0.56	+1.02	+1.51
Aldehyde content, %	2.02	1.52	1.97	1.65
Ester content, %	0.39	0.53	0.53	0.97
Evaporation residue, %	1.31	1.71	2.09	3.12
Yield, lb oil/ton peel	1.85	4.90	7.00	9.70

[a] From Kesterson and Hendrickson (1953). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

gravity and optical rotation were determined as recommended by the United States Pharmacopoeia (1965). The aldehyde content of the oil was determined by the hydroxylamine method, a standard procedure which is given by Guenther (1952) and adopted by U.S.P. employing a potentiometric end point or by the Essential Oil Association procedure (1965) using the bromophenol blue end point. Evaporation residue was determined according to U.S.P. or E.O.A. procedures. The method of Seeker and Kirby, as reported by Poore (1932), was used for the determination of esters. In this method, the aldehydes present are removed with hydroxylamine hydrochloride prior to the saponification of the esters. Methods used to determine those properties required as standards of purity by the U.S. Pharmacopoeia and Essential Oil Association of the U.S.A. are described in detail by Kesterson and Hendrickson (1971).

SPF Data. The Aminco–Bowman spectrophotofluorometer (SPF) was used to obtain luminescence data according to the methods of Kesterson *et al.* (1969, 1970).

Gas Chromatographic Data. Gas chromatographic analyses were conducted isothermally using a Perkin–Elmer 226 with a Sargent recorder (5 mV). The parameters employed were as follows. 0.2-μl sample, temperature: column, 130° C; block, 200° C; detector, 170° C. Carrier gas: helium, 40 psi; hydrogen, 10 psi; air, 35 psi. Stationary phase; Carbowax 20M + Versamid 930 (99:1 w/w) and Apiezon L + Igepal CO-880 + DOPC (95:4:1 w/w); column 300-ft × 0.01-in. i.d. capillary. The recorder had a chart speed of 0.5 in./min. Compounds are listed (Tables VII and X) in the order in which they emerged from the Carbowax 20M column.

Commercial Samples. Cold-pressed oils of orange, grapefruit, and tangerine were secured from plants using different methods for extraction of the oil from the peel. Over 700 samples were taken from lots of oil, representing the production for approximately 1 week. One plant furnished 12 samples of Valencia orange oil for analyses to determine the effect of fruit storage prior to extraction on the physical and chemical characteristics of the oil. One part of the selected lot of Valencia oranges was processed through the oil plant on the day it was picked, while an equal quantity of the same lot of fruit was held in ambient storage from 3 to 5 days prior to oil extraction.

Distilled Oils. Distilled oil of orange was secured from processing plants as a byproduct in the de-oiling of citrus fruit juices. The juice was flashed in the de-oiler operated in a vacuum of 11 in. (gauge) at 87° C to 25.5 in. (gauge) at 54° C. A vapor mixture of oil and water was removed and condensed and the oil was separated by decantation or centrifugation.

Essence Oils. Essence oils were obtained from juice evaporators during concentration of citrus juices. The principle of essence or volatile component recovery from citrus juices has been described by Wolford *et al.* (1968, 1969). Essence oils were separated from the aqueous essences in on-line decantation systems and from the surface of essences during chilled storage in large stainless steel tanks fitted with weirs.

Experimental Oil Samples. Experimental oil samples were prepared at the University of Florida Agricultural Research and Education Center's pilot plant utilizing the FMC in-line extractor with mist spray attachment and auxiliary equipment previously described by Kesterson and Hendrickson (1953). Ten box lots of fruit were used to prepare oil samples from the orange-type cultivars, while 15 box lots were used for the grapefruit cultivars. The Fraser–Brace excoriator was used to prepare oil samples from small immature fruit since the immature fruit was not suitable for processing in the FMC in-line extractor.

Commercial Processing Methods. Citrus peel oils have been expressed in Florida by seven different types of equipment: Pipkin roll, screw press, Fraser–Brace excoriator, FMC rotary juice extractor, FMC in-line extractor, AMC scarifier, and Brown peel shaver. The FMC in-line extractor, the screw press, and the Brown peel shaver represent the commercial systems now in use in Florida. The various types of equipment for expressing peel oils and the general processing procedures used after extraction for production of high quality cold-pressed citrus essential oils are discussed by Kesterson and Hendrickson (1953, 1971).

RESULTS AND DISCUSSION

One of the factors found to influence the physical and chemical properties of cold-pressed oil of orange to the greatest extent was the yield of peel oil. Data obtained by Kesterson and Hendrickson (1953) and presented in Table I show some typical analyses of samples of Florida cold-pressed Valencia orange oil extracted by four processes: Pipkin roll, screw press, FMC rotary juice extractor, and Fraser–Brace excoriator. Yields of oil obtained using these four different methods of processing were shown to vary from 1.85 to 9.70 lb per ton of peel. Examination of these data point up some of the effects of yield on specific properties of the oil, such as the lowering of the optical rotation while other physical properties, specific gravity, evaporation residue, and refrac-

tive index increased. They reasoned that the decrease in optical rotation might be due to the presence of more high molecular weight substances extracted under higher yield conditions with a concomitant lower yield of *d*-limonene, which would result in a lower optical rotation. The percentage increase in evaporation residue appears to follow that of the difference in optical rotation between the 10% distillate and the optical density of the whole oil. Likewise, these data show an increase in ester content with increased yield. The variability in aldehyde content is related to methods of oil extraction and recovery.

Effect of Aqueous Phase on Aldehyde Content. In the extraction of citrus oils, the oil cells are ruptured by pressure or abrasion and the oil is washed away employing adequate amounts of water. A relationship determined by Kesterson and Hendrickson (1953) between aldehyde content and the amount of aqueous phase which influences yield is represented in Figure 1. Apparently, the aldehyde content of the oil would be affected to the greatest extent up to approximately 100 gal of aqueous phase per gallon of oil, or a relative 1% oil concentration in the emulsion from the finisher to the centrifuging operations. It is evident that in the production of orange oils with high aldehyde contents, the amount of aqueous phase should be reduced to as small a quantity as is practical under operating conditions. Kesterson and Hendrickson (1967) showed an increase in insoluble solids with increasing amounts of aqueous phase. These insoluble solids, primarily pulp particles, presumably act similar to ion-exchange resins and selectively absorb constituents from the oil. Loss of aldehyde in operations using large quantities of water might then be explained by absorption loss, as well as by loss through solubility. In addition to the above, there remains the possibility of enzymatic degradation.

Storage of Fruit Prior to Oil Extraction. Results obtained indicate that the length of time fruit was stored prior to oil extraction was another factor which influenced the characteristics and quality of the oil.

There were no significant differences in the physical properties of cold-pressed oils of orange extracted from fruit on the same day it was harvested and those extracted from fruit stored in fruit bins for 3 to 5 days before the oils were extracted. However, significant differences were found in the chemical properties. The ester content of the oil from stored fruit was 31.3% higher than that extracted from fruit which had not been stored. The evaporation residue of the oil from stored fruit was 9.6% higher and the aldehyde content was 4.6% lower.

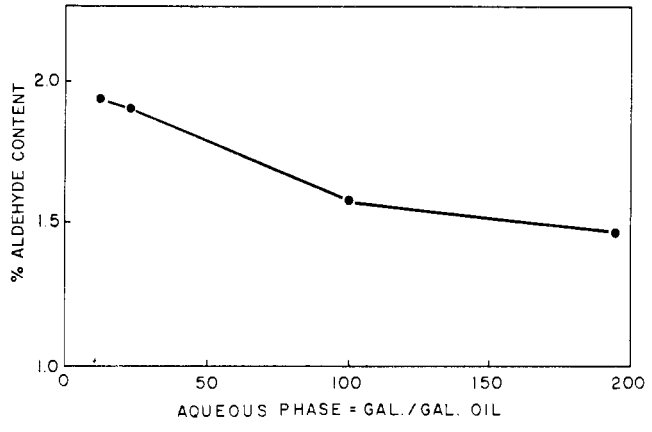

Figure 1. Influence of the quantity of aqueous phase which comes in contact with the oil during processing of the aldehyde content of cold-pressed orange oils. After Kesterson and Hendrickson (1953). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences

Effect of Maturity on Properties. In the studies by Kesterson and Hendrickson (1953, 1971) of the effect of fruit storage on oil quality, all samples of oil of orange were from the same variety of fruit and were extracted by the same process. Therefore, over a period of 4 months information was obtained in reference to the effect of fruit maturity on properties of the oil. Here, again, differences were noted in chemical characteristics rather than in physical properties. Aldehyde content of Valencia orange oils increased as fruit maturity increased, reached a maximum when extracted during the early part of the Valencia season from fruit that just passed the maturity standards, and then decreased after peak maturity had been reached. The ester content of these oils was lowest when extracted during the early part of the Valencia season and gradually increased as the fruit matured. Valencia oranges that had passed peak maturity produced an oil with the highest ester content.

Relation of Fruit Variety to Properties. Kesterson and Hendrickson (1953, 1971) showed that oils manufactured by any one process fell within a particular category of their own and remained there throughout the season. Differences in physical properties of expressed orange oils obtained from different varieties of fruit by any particular process were not significant. Their study of the chemical properties showed the aldehyde content of cold-pressed oils of orange was highest when made from Valencia oranges. Mixtures of pineapple and seedling oranges gave an oil with a lower aldehyde content, and mixtures of Hamlin and Parson Brown varieties yielded the lowest aldehyde content.

Variety of fruit apparently had very little effect on the ester content of orange oils. Oil of orange produced by the Fraser–Brace extractor from midseason varieties of fruit that were partially green in color was considerably higher in ester content than that made by the same process on the same varieties later in the season when the fruit was completely orange in color. It was also higher in esters than oils produced by the other methods. High evaporation residue values also were found for oils produced by the Fraser–Brace extractor.

Effect of Yearly Variations on Properties. Yearly or seasonal variations in physicochemical properties of expressed orange oil have been concerned mainly with two factors: refractive index and aldehyde content. A definitive comparison between oils produced in 1947–1948 (a very wet season) and 1948–1949 (a very dry season) showed the average refractive index to be 0.0008 of a unit higher during the dry year, while a 16.1% reduction in aldehyde was experienced in the dry season of 1948–1949. Kesterson and Hendrickson (1953, 1971) reported on the aldehyde content of Valencia orange oil as related to total rainfall (Figure 2). Comparison was made between the aldehyde contents of commercial samples of Valencia orange oil produced from 1951 to 1964 and the total rainfall data each season based on official records of the Lakeland Weather Bureau, as reported by Johnson (1965). A positive correlation of 0.603 with significance at the 5% level was obtained for the relationship between aldehyde contents and rainfall. The authors concluded that in view of some exceptions to the rule the distribution of rainfall within a season may be a factor that also influences the aldehyde content. They presented a regression analysis of their data to provide annual predictions of aldehyde contents of Florida Valencia oils and stated that proper use

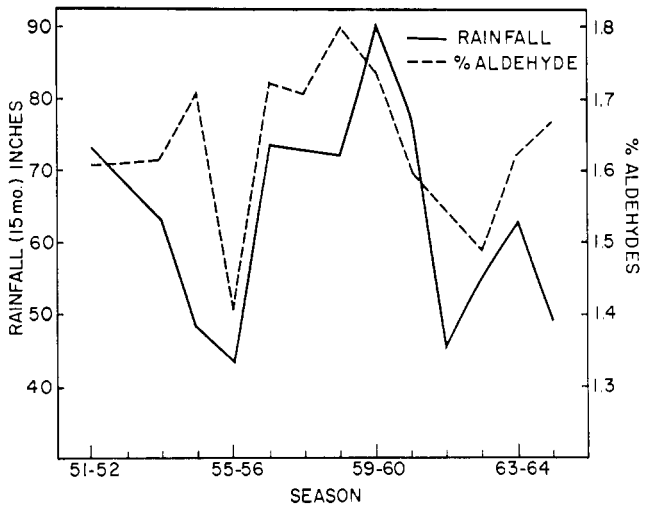

Figure 2. The aldehyde content of Valencia orange oil as related to total rainfall

of irrigation would make it possible for the aldehyde content to be maintained at an optimum value for high quality oils.

Cultivar, Budwood, and Rootstock as Related to Oil Yield and Quality. Hendrickson and Kesterson (1970) collected data relative to the quantity of oil found in the peel of various citrus cultivars (Table II). Identical fruit trees were used for studies conducted both during the 1968–1969 and 1969–1970 fruit seasons. Peel oil contents are shown to be quite variable from season to season and further work is necessary to establish these limits.

Hendrickson *et al.* (1969, 1970) studied the peel oil contents from 34 different Valencia budwood selections on a common rootstock and indicated peel oil contents ranging from 11.1 to 15.7 lb per ton of fruit. Differences in yield were found to be statistically significant. These data suggested that the peel oil yield could be increased approximately 4.6 lb per ton of fruit by proper choice of budwood selections. However, more information is needed to recommend the ideal selection. In their studies, they showed that the peel oil content

Table II. Peel Oil Content of Various Citrus Cultivars[a]

Citrus cultivars	Season	No. of samples	Range in lb oil/ton of fruit			% Oil in fruit		
			Max	Min	Avg	Max	Min	Avg
Hamlin Orange	1968–1969	66	9.2	6.7	8.4	0.46	0.34	0.42
	1969–1970	30	8.4	5.0	6.0	0.42	0.25	0.30
Parson Brown	1968–1969	30	19.6	10.7	14.8	0.98	0.54	0.74
Orange	1969–1970	30	10.5	7.1	8.3	0.53	0.36	0.42
Pineapple	1968–1969	53	23.9	7.0	13.4	1.20	0.35	0.67
Orange	1969–1970	30	7.7	5.8	6.6	0.39	0.29	0.33
Valencia	1968–1969	204	28.5	9.2	18.0	1.43	0.46	0.90
Orange	1969–1970	139	18.7	9.3	13.2	0.94	0.47	0.66
Temple	1968–1969	...	...	...	...	...	...	...
Orange	1969–1970	27	13.0	6.2	8.5	0.65	0.31	0.43
Duncan	1968–1969	28	7.1	4.7	5.9	0.36	0.24	0.30
Grapefruit	1969–1970	24	7.6	4.8	6.1	0.38	0.24	0.31
Marsh	1968–1969	32	9.4	4.3	6.8	0.47	0.22	0.34
Grapefruit	1969–1970	30	7.8	5.1	6.1	0.39	0.26	0.31
Foster Pink	1968–1969	...	...	...	...	...	...	...
Grapefruit	1969–1970	21	9.4	5.3	7.0	0.47	0.27	0.35
Tangerine	1968–1969	21	17.6	7.9	14.0	0.88	0.40	0.70
Lemon	1968–1969	8	15.7	12.0	13.2	0.79	0.60	0.66
Lime	1968–1969	3	6.9	6.5	6.7	0.35	0.33	0.34

[a] From Hendrickson and Kesterson (1970).

Table III. Average Values for Properties of Commercial Citrus Essential Oils from Four Cultivars Produced in Florida During Three Seasons[a]

Year	Variety of fruit	Specific gravity, 25° C/25° C	Refractive index, $n^{20}D$	Refractive index of 10% distillate, $n^{20}D$	Difference	Optical rotation $\alpha[^{25}D]$	Optical rotation of 10% distillate, $\alpha[^{25}D]$	Difference	Aldehyde content, %	Evaporation residue, %
1967–1968	Valencia Orange	0.8430	1.4730	1.4719	0.0012	+96.49	+97.49	+1.21	1.67	2.13
	Grapefruit	0.8524	1.4762	1.4719	0.0043	+93.40	+98.11	+4.78	1.43	7.09
	Tangerine	0.8468	1.4744	1.4721	0.0023	+91.28	+93.90	+2.62	1.21	4.10
	Lemon	0.8472	1.4744	1.4729	0.0016	+66.07	+63.07	−2.48	2.45	1.81
1968–1969	Valencia Orange	0.8431	1.4730	1.4719	0.0011	+97.35	+98.31	+1.01	1.72	2.06
	Grapefruit	0.8516	1.4759	1.4719	0.0042	+93.30	+97.77	+4.43	1.46	6.23
	Tangerine	0.8451	1.4740	1.4719	0.0021	+93.73	+95.97	+2.24	1.23	3.52
	Lemon	0.8500	1.4748	1.4738	0.0010	+63.00	+61.40	−1.60	3.30	2.15
1969–1970	Valencia Orange	0.8430	1.4730	1.4718	0.0012	+97.23	+98.11	+0.94	1.50	52.8[a]
	Grapefruit	0.8575	1.4775	1.4713	0.0062	+90.49	+97.37	+6.88	1.23	9.53
	Tangerine	0.8446	1.4737	1.4721	0.0016	+92.12	+93.52	+1.40	1.13	4.69
	Lemon									

[a] Evaporation residue, (mg/3 ml sample) as per present U.S.P. Standards XVII.

Table IV. Physicochemical Properties of Oils Centrifuged from Valencia Orange Juice as Compared with Cold-Pressed Valencia Orange Oil[a]

Type oil	Juice oil		Cold-pressed oil
Extractor	Brown reamer	FMC in-line	FMC in-line
Specific gravity, 25° C/25° C	0.8427	0.8501	0.8427
Refractive index, $n^{20}D$	1.4730	1.4740	1.4730
Refractive index, 10% distillate, $n^{20}D$	1.4723	1.4724	1.4718
Difference	0.0007	0.0016	0.0012
Optical rotation, $\alpha[^{25}D]$	+97.22	+90.82	+97.42
Optical rotation 10% distillate, $\alpha[^{25}D]$	+99.11	+99.31	+97.71
Difference	+1.89	+8.49	+0.29
Aldehyde (decyl), %	0.86	0.72	1.63
Evaporation residue, %	2.56	10.15	1.79
Acid no.	1.17	1.66	0.50
Free acid, %	0.30	0.43	0.13
Ester no. before acetylation	1.94	11.99	0.27
% Ester before acetylation	0.68	4.19	0.10
Ester no. after acetylation	3.83	14.68	3.43
% Ester after acetylation	1.34	5.13	1.20
Free alcohol, %	0.52	0.74	0.87
Total alcohol, %	1.05	4.04	0.94

[a] From Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

of Valencia fruit from a common scion grown on 19 different rootstocks ranged from 13.5 to 17.9 lb of oil per ton of fruit. Some rootstocks seemed to suppress oil yields while others increased oil yields. Mean fruit weight and mean surface area did not show any significant relationship to peel oil content. Analysis of variance showed a significant difference in peel oil content. Brazilian Valencia oil samples obtained from a common scion on different rootstocks had aldehyde values of 1.26, 1.44, and 1.73%, respectively, for trifoliate orange, sweet orange, and Rangpur lime. Rootstock apparently has a profound influence on the aldehyde content of orange oil, the amount of which is considered as an indicator of quality. Average values for properties of commercial citrus essential oils from four cultivars produced in Florida during three seasons are shown in Table III.

Juice Oil *vs.* Cold-Pressed Oil. During the manufacture of baby or infant juices, it is customary to centrifuge juice in order to lower the oil content. Our laboratory has examined one sample each of Valencia orange juice oil prepared from juices extracted by the Brown reamer and FMC in-line extractors. These oils possessed a flavor and aroma that is typical of fresh orange juice and quite different from cold-pressed orange oils. The physicochemical properties for these oils are shown in Table IV and compared with a typical cold-pressed Valencia orange oil.

The ratio of the oxygenated components shows that the juice oils are low in aldehyde content and high in ester content as compared to a cold-pressed oil. The ester content of juice oils is some 7 to 18 times greater than that of a cold-pressed oil, and this change in ratio of flavor components is probably responsible for the fruity note in the juice oils. The two juice oils were quite different in chemical composition, undoubtedly due to the difference in the type of equipment used to express the juice.

Orange Essence Oil. Orange essence oils are produced commercially in Florida simultaneously with recovery of aqueous orange essences as described by Wolford *et al.* (1968, 1969). Table V shows the physicochemical properties for 15 essence oils obtained from four different processes during the 1968–1969 season. The aroma and flavor of these oils are quite different from other orange oils produced in Florida, having a fruity-like aroma characteristic of fresh juice. Good quality essence oils merit consideration for use in oil added back to juice, perfumes, beverages, condiments, etc.

Table V. Physicochemical Properties of Orange Essence Oils[a]

	Maximum	Minimum	Average
Specific gravity, 25° C/25° C	0.8428	0.8403	0.8415
Refractive index, $n^{20}D$	1.4725	1.4721	1.4723
Optical rotation, $\alpha[^{25}D]$	+99.16	+97.68	+98.42
Aldehyde, %	1.86	1.28	1.57
Evaporation residues, %	1.29	0.34	0.81
Acid no.	0.22	0.11	0.16
Free acid, %	0.06	0.03	0.04
Ester no. before acetylation	3.08	2.94	3.00
% Ester before acetylation	1.08	1.03	1.05
Ester no. after acetylation	6.50	5.43	6.06
% Ester after acetylation	2.27	1.90	2.12
Free alcohol, %	0.97	0.64	0.84
Total alcohol, %	1.78	1.49	1.66

[a] From Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

Orange essence oils contain 0.5 to 2.0% valencene, a sesquiterpene, not appreciably present in cold-pressed orange oils. This terpene can be recovered and converted into nootkatone and used as a flavor enhancer.

Gas chromatographic analyses of the terpeneless expressed or cold-pressed essence and juice oils of orange have been reported by Kesterson and Hendrickson (1971). Quantitative analyses of the chromatographic peaks provided the basis for the ratio of oxygenated components, aldehydes, acids, esters, and alcohols presented in Table VI.

Chemical Composition of Cold-Pressed Valencia Orange Oil. Isolation and identification of the flavor components found in orange peel oil has been a subject of considerable research. Kesterson and Hendrickson (1971) provide an up-dated list of compounds by chemical classification found in orange oil by many referenced investigators. Aldehydes A, B, C, D, and E are α,β-unsaturated R–CH=C–CHO, R, and R′ straight-chain $CH_3(CH_2)_n$; n is 5, 6, 7, or 8. Many of the compounds listed in Table VII have also been identified in orange essence and juice oils.

Oil of Grapefruit. The demand for Florida cold-pressed grapefruit oil has increased phenomenally due to the popularity of carbonated grapefruit beverage drinks. White and red grapefruit oils are currently manufactured in Florida. The white grapefruit oils are produced primarily from Duncan Seedy and Marsh Seedless, and the red grapefruit oils are produced from Ruby Reds and occasionally from Foster

Table VI. Ratio of the Oxygenated Components of Various Orange Oils

	Expressed	Essence	Juice
Aldehyde, %	59.6	46.4	46.0
Acid, %	4.8	1.2	1.1
Ester, %	3.7	29.3	21.0
Alcohol, %	31.9	23.1	31.9
	100.0	100.0	100.0

Table VII. The Chemical Composition of Cold-Pressed Valencia Orange Oils

Terpenes	Alcohols	Ketones
α-thujene	methyl alcohol	carbone
α-pinene	ethyl alcohol	methylheptenone
camphene	amyl alcohol	α-ionone
2,4-*p*-menthadiene	1-octanol	acetone
sabinene	1-decanol	piperitenone
myrcene	linalool	6-methyl-5-hepten-2-one
Δ-3-carene	citronellol	nootkatone
α-phellandrene	α-terpineol	
α-terpinene	1-nonanol	**α,β-Dialkyl acroleins**
d-limonene	*trans*-carveol	α-hexyl-β-heptylacrolein
β-terpinene	geraniol	α-hexyl-β-octylacrolein
p-cymene	nerol	α-heptyl-β-heptylacrolein
α-terpinolene	heptanol	α-octyl-β-heptylacrolein
α,β-cubebene	undecanol	α-hexyl-β-nonylacrolein
α,β-copaene	dodecanol	α-octyl-β-octylacrolein
β-elemene	elemol	α-heptyl-β-nonylacrolein
caryophyllene	*cis,trans*-2,8-*p*-menthadien-1-ol	**Paraffin waxes**
farnesene	*cis*-carveol	n-$C_{21}H_{44}$
α,β-humulene	1-*p*-methen-9-ol	
valencene	1,8-*p*-menthadien-9-ol	2-methyl-$C_{21}H_{43}$
Δ-cadinene	8-*p*-methene-1,2-diol	
Aldehydes	isopulego	n-$C_{22}H_{46}$
formaldehyde	borneol	
acetaldehyde	methylheptenol	2-methyl-$C_{22}H_{45}$
n-hexanal	hexenol-1	
n-heptanal	terpinen-4-ol	n-$C_{23}H_{48}$
n-octanal	**Esters**	
n-nonanal	perillyl acetate	3-methyl-$C_{23}H_{47}$
n-decanal	*n*-octyl acetate	
n-undecanal	bornyl acetate	n-$C_{24}H_{30}$
n-dodecanal	geranyl formate	
citral { neral, geranial }	terpinyl acetate	2-methyl-$C_{24}H_{49}$
citronellal	linalyl acetate	
α-sinensal	linalyl propionate	n-$C_{25}H_{52}$
β-sinensal	geranyl acetate	
trans-hexen-2-al-1	nonyl acetate	3-methyl-$C_{25}H_{51}$
dodecen-2-al-1	decyl acetate	
furfural	neryl acetate	n-$C_{26}H_{54}$
perillyldehyde	citronellyl acetate	
Aldehyde A	ethyl isovalerate	2-methyl-$C_{26}H_{53}$
B	geranyl butyrate	
C	1,8-*p*-methadien-9-yl-acetate	n-$C_{27}H_{56}$
D		
E	**Acids**	3-methyl-$C_{27}H_{55}$
Oxides	formic	n-$C_{28}H_{58}$
trans-limonene oxide	acetic	
cis-limonene oxide	caprylic	2-methyl-$C_{28}H_{57}$
	capric	n-$C_{29}H_{60}$

[a] From Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

and Thompson pinks. The odor and flavor of grapefruit oil is extremely delicate and characteristic of the fruit from which it is extracted. Therefore, the greatest care is exercised to avoid admixture with other citrus peels or fruit. The same general processing techniques employed for orange oil are used to extract grapefruit oil from residual peel and whole fruit.

Comparison of Red and White Grapefruit Oils. Red and white grapefruit oils were obtained from three different commercial plants employing either the screw press or FMC in-line extractors for oil recovery. These oil samples were dried over anhydrous sodium sulfate and stored in glass for 1 year under nitrogen at 4° C.

The physical and chemical properties for these oils presented by Kesterson and Hendrickson (1953, 1971) (Table VIII) showed that values for specific gravity, refractive index, difference between original oil and 10% distillate for refractive index and optical rotation, and evaporation residue without exception were highest for red grapefruit oils, while the values for optical rotation and aldehyde content were lower than those for white grapefruit oils. Terpeneless oils prepared by the method of Kirchner and Miller (1952) were analyzed by gas chromatography. Chemical composition of the oils was found to be quite variable while the two principal components were octyl and decyl aldehydes. In white grapefruit the ratio of octyl to decyl aldehyde was found to range from 1:1.1 to 1:1.4. In red grapefruit these values were reversed and ranged from 1.2:1 to 1.3:1. Compositional changes of this magnitude would undoubtedly alter the flavor characteristics of the oils. Red grapefruit oils contained a small quantity of linalool while linalool was apparently absent in white grapefruit oils. Nootkatone, a sesquiterpene

Table VIII. Average Values for the Physical and Chemical Properties of Cold-Pressed Red and White Grapefruit Oils[a]

Extractor type oil	FMC in-line		Screw press	
	White	Red	White	Red
Specific gravity, 25° C/25° C	0.8539	0.8590	0.8534	0.0852
Refractive index, $n^{20}D$	1.4760	1.4782	1.4759	1.4766
Refractive index, 10% distillate, $n^{20}D$	1.4719	1.4719	1.4718	1.4718
Difference	0.0041	0.0064	0.0041	0.0048
Optical rotation, $\alpha[^{25}D]$	+93.07	+90.77	+92.67	+91.07
Optical rotation, 10% distillate, $\alpha[^{25}D]$	+97.12	+96.92	+97.04	+96.84
Difference	+4.05	+6.15	+4.37	+5.77
Evaporation residue, %	6.34	8.71	6.32	7.12
Aldehyde content, %	1.56	1.23	1.56	1.38

[a] From Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

Table IX. Nootkatone Concentration and Physicochemical Properties of Expressed Duncan Grapefruit Oil as Related to Fruit Maturity[a]

Sample no.	1	2	3	4	5	6
Processing date	11/19/63	12/31/63	2/11/64	3/24/64	5/5/64	6/16/64
Oil yield, lb/ton fruit	2.25	2.38	1.25	1.33	1.23	0.50
Specific gravity, 25° C/25° C	0.8538	0.8518	0.8517	0.8514	0.8515	0.8531
Refractive index, $n^{20}D$	1.4765	1.4761	1.4760	1.4759	1.4761	1.4764
Refractive index, 10% distillate, $n^{20}D$	1.4714	1.4716	1.4717	1.4718	1.4720	
Difference	0.0051	0.0045	0.0043	0.0041	0.0041	
Optical rotation, $\alpha[^{25}D]$	+92.77	+92.97	+93.17	+93.57	+93.13	+92.17
Optical rotation, 10% distillate, $\alpha[^{25}D]$	+97.64	+97.84	+98.24	+98.64	+98.55	
Difference	+4.87	+4.87	+5.07	+5.07	+5.42	
Aldehyde content, %	1.43	1.62	1.80	1.76	1.79	1.61
Evaporation residue, %	7.89	6.87	7.48	7.25	7.26	8.42
Acid no.	1.33	1.14	1.54	1.46	1.40	1.66
Free acid, %	0.34	0.29	0.39	0.38	0.36	0.43
Ester no. before acetylation	10.88	9.97	9.34	8.51	9.36	9.62
% Ester before acetylation	3.80	3.48	3.26	2.97	3.27	3.36
Ester no. after acetylation	15.45	13.33	13.17	13.24	13.08	12.65
% Ester after acetylation	5.40	4.66	4.60	4.63	4.57	4.42
Free alcohol, %	1.26	0.93	1.06	1.30	1.03	0.83
Total alcohol, %	4.24	3.66	3.62	3.64	3.59	3.48
Nootkatone, %	0.0065	0.285	0.503	0.693	0.750	0.810

[a] From Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

ketone with a carbon skeletal structure identical to valencene, is found in expressed grapefruit oil. It is the present feeling in the flavor industry that good grapefruit flavor is related to the nootkatone content of expressed grapefruit oil.

Duncan grapefruit were processed throughout the 1963–1964 season from the date the fruit first reached legal maturity and at 6-week intervals thereafter. Samples were stored in sealed amber bottles under nitrogen at 4° C until the last oil sample was aged for 6 months. A progressive increase in nootkatone content of expressed grapefruit oil from 0.065 to 0.810% was observed as the fruit became more mature (Table IX). The nootkatone content of oil could probably be used as a measure of fruit maturity. However, the yield of oil recovered from the fruit diminished from approximately 2.30 lb to 0.50 lb per ton of fruit as the fruit ripened. There is a likelihood that a compromise can be made between yield and good flavor quality in order to make oil production economically feasible. If the percent nootkatone, a ketone, is deducted from the aldehyde content, a gradual decrease occurs in the aldehyde content which is also characteristic of orange with advancing maturity. However, nootkatone increases at a faster rate than the aldehyde decreases, so the net effect is an overall increase in total carbonyl content. Majority opinion of a panel of experts preferred samples containing the highest aldehyde content in which the nootkatone concentration varied between 0.50 and 0.70%. Therefore, it would seem advisable to manufacture grapefruit oils during the months of February, March, and April to obtain oils with the best odor and flavor characteristics.

Curing Florida Grapefruit Oils. Commercial practices described by Kesterson and Hendrickson (1963) and Kesterson *et al.* (1965a,b) have shown that expressed grapefruit oils should be carefully aged for 6 to 12 months to develop their full, rich-bodied, distinctive grapefruit character. Results obtained in their studies showed the values for specific gravity, refractive index, optical rotation, evaporation residue, aldehyde, acid number, percent free acid, ester number before acetylation, percent ester before acetylation, and percent total alcohol remained relatively constant, while the values for ester number after acetylation, percent ester after acetylation, and percent free alcohol decreased with aging or curing of the oil. These changes were associated with a decrease in percent free alcohol which was primarily related to the disappearance of linalool. When grapefruit oils are freshly prepared they possess an orange-like bynote that disappears on storage. The most striking change in the chemical composition of samples during storage or aging was the complete loss of linalool by the end of 12 months. This change is very likely responsible for the disappearance of the orange-like character. This study was concerned only with the oxygenated components since they were thought to contribute most to the flavor of an oil. However, since many changes

Table X. Chemical Composition of Cold-Pressed Grapefruit Oil[a]

Terpenes

α-pinene
sabinene
β-myrcene
d-limonene
α-terpinene
γ-terpinene
β-ocimene
α,β-cubebene
α,β-copaene
β-elemene
carophyllene
?
α,β-humulene
cadinene
?
Δ-cadinene
$C_{15}H_{24}$
auraptene

Aldehydes

heptanal
octanal
nonanal
citronellal
decanal
undecanal
dodecanal
citral { genanial / neral

Phenols

o-phenylphenol

Acids

acetic acid
caprylic acid
capric acid

Alcohols

methylheptenol
linalool
octanol
nonanol
decanol
α-terpineol
nerol
geraniol
nerolidol
elemol

Triterpenoids

β-sitosterol
citrostadienol
campesterol
stigmasterol
cycloartenol
24-methylene-cycloartenol
24-methylenelophenol

Esters

octyl acetate
linalyl acetate
nonyl acetate
geranyl acetate
decyl acetate
neryl acetate
citronellyl acetate
geranyl butyrate

Oxides

trans-linalool oxide
cis-linalool oxide

Alcohols

trans-2-8-*p*-menthadien-1-ol
cis-2-8-*p*-menthadien-1-ol
citronellol
trans-carveol
cis-carveol
dodecanol
1-8-*p*-menthadien-9-ol
8-*p*-menthene-1,2-diol

Coumarins and psoralens

bergamottin
7-geranyloxycoumarin
osthol
limettin (citroptene)
bergapten
bergaptol
7-methoxy-8-(2-formyl-2-methylpropyl)coumarin
7-[(6,7-dihydroxy-3,7-dimethyl-2-octenyl)oxy]coumarin
5-[(3,6-dimethyl-6-formyl-2-heptenyl)oxy]psoralen
umbelliferone

Ketones

nootkatone
methylheptenone
carvone

[a] Taken from Kesterson and Hendrickson (1971). Reprinted with permission of University of Florida Institute of Food and Agricultural Sciences.

have been shown to occur which were not anticipated, it is quite likely that the terpene fractions undergo similar changes that could contribute to flavor.

In light of changes encountered in oils at 4° C storage, samples of grapefruit oil were also stored at 15 and 26° C. Based on gas chromatographic analyses of terpeneless oils, almost the identical changes occurred as at 4° C storage, except that at the higher storage temperatures the changes were greatly accelerated and linalool completely disappeared in 6 months at 26° C. If the linalool content was used as a criterion to determine the proper curing procedure for expressed grapefruit oil, it would follow that the most ideal storage temperature lies somewhere between 15 and 26° C and most probably in the range of 18 to 21° C. At this temperature the holding time could be reduced to 6 months or half of that time at 4° C. Prior to the curing period a dewaxing at −23 to −1° C takes place.

Chemical Composition of Grapefruit Oil. Grapefruit oils have been as extensively analyzed as orange oils. Table X from the work of Kesterson and Hendrickson (1971) provides a summary list of chemical components identified by several referenced researchers. In all, nearly 80 flavor components have been identified out of more than 100 components detected largely by gas chromatographic techniques.

Other Florida Citrus Essential Oils. Discussion to this point has been concerned with physicochemical characteristics of orange and grapefruit oils which constitute the major production in the industry. Lesser volumes of essential oils such as lime, lemon, and the mandarin type citrus oils, tangerine, Murcott, tangelo, and Temple are also produced in Florida. Most of the physicochemical properties of these oils have been reported by Kesterson and Hendrickson (1958, 1960, 1969, 1971). Also, in their latter publication, a bulletin on Florida Citrus Oils, the methods employed in recovery of oil from these fruit varieties and cultivars, are reviewed in light of yields of oil, the effects of equipment on physical properties, and relationships between yield and quality. Many factors affecting quality in these several oils relate similarly to the more significant considerations affecting quality of orange and grapefruit oils discussed earlier. Of these several essential oils, the chemical composition of lemon oil relative to flavor components has been explored extensively by Bernhard (1960), Slater (1960), Ikeda and Spitler (1964), Hunter and Moshonas (1966), and MacLeod *et al.* (1966). Hunter and Moshonas (1966) determined the chemical composition of tangerine oil and lime oils. Slater (1961) and Strickler and Kovats (1966) also reported on the chemical composition of lime oil. The composition of tangelo oil was studied by Kesterson and Hendrickson (1969, 1971).

Stability and Quality in Citrus Oils. Quality of citrus peel oils is largely determined by the yield of oil obtained, which in turn is related to the types of processing equipment used. Studies conducted by Kesterson and McDuff (1949) showed that a direct correlation existed between evaporation residue and yield of oil and that an oil with a high value for evapora-

tion residue will be more stable toward oxidation than an oil with a low value for evaporation residue. Sanitation and bacteriological contamination, discussed by Murdock and Hunter (1970), are factors that have an influence on oil quality. Antioxidants, studied by Kesterson and McDuff (1949), were shown to have a direct influence on the oxidative stability of citrus oils. Proper storage and handling of citrus oils are most important in maintaining maximum flavor and odor qualities.

Spectrophotometric Methods for Evaluation and Identification of Oils. Examination of the physicochemical properties of cold-pressed orange oils has revealed differences in the ultraviolet spectra and in the evaporative residue of the oils which reflect the geographical source of the oil and the method by which it was obtained. These tests, taken together, offer a quick and accurate means by which the method of extraction and geographical origin may be determined.

Recent studies conducted by Kesterson *et al.* (1970) and reviewed by Kesterson and Hendrickson (1971) show some very distinct advantages in the use of luminescence techniques. Spectrophotofluorescence (SPF) can be employed for both identification and classification of citrus oils. By use of SPF, varieties within a species may be identified, which is not possible with the uv absorption method. However, Kesterson *et al.* (1959) employing the method of Sale (1953) have shown that uv absorption data for cold-pressed orange and grapefruit oils were significantly different to make a distinction between these two types of oil. Commercial Florida and California orange oil can also be separated since CD values for California oil rarely exceed a value of 0.20, while for Florida oils they are always greater than 0.20. On the basis of SPF, Florida, California, and Arizona commercial orange and grapefruit oils were found to be sufficiently different from one another to allow identification.

In conclusion, it has been shown that the evaluation of citrus essential oils requires a number of analytical considerations, as well as the ultimate criterion of organoleptic properties of the individual type or variety of oil required for a particular flavoring application. It has been shown that fruit variety, degree of maturity, seasonal variations, storage of fruit prior to extraction, rainfall, budwood, rootstock, method of extraction, and yield of oil were factors which affected the chemical and physical properties of expressed citrus oils.

LITERATURE CITED

Association of Official Agricultural Chemists, Official and Tentative Methods of Analysis, 11th ed., Washington, D.C., 1970.
Bartholomew, E. T., Sinclair, W. B., *Plant Physiol.* **21,** 319 (1946).
Bernhard, R. A., *J. Chromatogr.* **3,** 471 (1960).
de Villiers, F. J., *Farming S. Afr.* **4,** 515 (1930).
Essential Oil Association of U.S.A., Specifications and Standards, New York, N.Y., 1965.
Flores, H., Morse, R. E., *Food Technol.* **6,** 6 (1952).
Foote, P. A., Gelpi, R. Z., *J. Amer. Pharm. Ass.* **32,** 145 (1943).
Gildemeister, E., Hoffman, F., "Die Aetherischen Oele," 3rd ed., Leipzig, Vols. I, II, and III (1928–1931).
Guenther, E., "The Essential Oils," Vols. I, II, III, IV, V, and VI., Van Nostrand, New York, N.Y., 1952.
Hendrickson, R., Kesterson, J. W., "Peel Oil Content of Citrus Fruit Cultivars," unpublished data (1970).
Hendrickson, R., Kesterson, J. W., Cohen, M., *Proc. Fla. State Hort. Soc.* in press (1970).
Hendrickson, R., Kesterson, J. W., Ting, S. V., *Proc. Fla. State Hort. Soc.* **82,** 192 (1969).
Hood, S. C., *Ind. Eng. Chem.* **8,** 709 (1916).
Hunter, G. L. K., Moshonas, M. G., *J. Food Sci.* **31,** 168 (1966).
Ikeda, R. M., Spitler, E. M., J. AGR. FOOD CHEM. **12,** 114 (1964).
Johnson, W. O., The Seasons Report of the Federal–State Frost Warning Service, U.S. Dept. of Commerce, Weather Bureau and Univ. of Fla. Agr. Expt. Sta., Lakeland, Fla., 1965.
Kenyon, E. M., Proctor, B. E., *Food Res.* **16,** 365 (1951).
Kesterson, J. W., Edwards, G. J., Hendrickson, R., *Amer. Perfum. Cosmet.* **84**(3), 41 (1969).
Kesterson, J. W., Edwards, G. J., Hendrickson, R., *Amer. Perfum. Cosmet.* **85**(10), 41, 46 (1970).
Kesterson, J. W., Hendrickson, R., *Food Technol.* **5,** 220 (1951b).
Kesterson, J. W., Hendrickson, R., *Amer. Perfum. Essent. Oil Rev.* **57**(6), 441 (1951a).
Kesterson, J. W., Hendrickson, R., "Essential Oils from Florida Citrus," Fla. Agr. Expt. Sta. Tech. Bull. 521 (1953).
Kesterson, J. W., Hendrickson, R., *Proc. Fla. State Hort. Soc.* **71,** 132 (1958).
Kesterson, J. W., Hendrickson, R., *Amer. Perfum. Aromat.* **75**(11), 35 (1960).
Kesterson, J. W., Hendrickson, R., *Amer. Perfum. Cosmet.* **78**(5), 32 (1963).
Kesterson, J. W., Hendrickson, R., *Amer. Perfum. Cosmet.* **84**(5), 51 (1969).
Kesterson, J. W., Hendrickson, R., *Citrus Ind.* **48,** 23 (1967).
Kesterson, J. W., Hendrickson, R., "Florida Citrus Oils," Univ. Fla. Inst. Food Agr. Sci. Tech. Bull. in press (1971).
Kesterson, J. W., Hendrickson, R., Edwards, G. J., *Amer. Perfum. Aromat.* **74**(4), 33, 52 (1959).
Kesterson, J. W., Hendrickson, R., Seiler, R. R., Huffman, C. E., Brent, J. A., Griffiths, J. T., *Amer. Perfum. Cosmet.* **80**(12), 29 (1965a).
Kesterson, J. W., Hendrickson, R., Seiler, R. R., Huffman, C. E., Brent, J. A., Griffiths, J. T., *Proc. Fla. State Hort. Soc.* **78,** 207 (1965b).
Kesterson, J. W., McDuff, O. R., *Amer. Perfum. Essent. Oil Rev.* **54**(10), 285 (1949).
Kirchner, J. G., Miller, J. M., *Ind. Eng. Chem.* **44,** 318 (1952).
MacLeod, W. D., McFadden, W. H., Buigues, N. M., *J. Food Sci.* **34,** 591 (1966).
Markley, K. S., Nelson, E. K., Sherman, M. S., *J. Biol. Chem.* **118,** 433 (1937).
Murdock, D. I., Hunter, G. L. K., *J. Food Sci.* **35,** 652 (1970).
Nelson, E. K., *Amer. Perfum. Essent. Oil Rev.* 347 (1934).
Nelson, E. K., Mottern, H. H., *J. Amer. Chem. Soc.* **56,** 1238 (1934).
Perry, E. J., "The Chemistry of Essential Oils and Artificial Perfumes," 2nd ed., 546 pp, Scott, Greenwood & Son, London, 1908.
Poore, H. D., "Analyses and Composition of California Lemon and Orange Oils," U.S. Dept. Agr. Tech. Bull. 241, 1–30 (1932).
Proctor, B. E., Kenyon, E. M., *Food Technol.* **3,** 387 (1949).
Sale, J. W., *J. Ass. Offic. Agr. Chem.* **36,** 112 (1953).
Slater, C. A., *Chem. Ind.* **833** (1961).
Slater, C. A., *J. Sci. Food Agr.* **3,** 257 (1960).
Strickler, Von H., Kovats, E., *Helv. Chim. Acta* **49,** 2055 (1966).
United States Pharmacopoeia, 17th revision, 1156 pp, Mack Publishing Co., Easton, Pa., 1965.
von Loesecke, H. W., Pulley, G. N., *Fruit Prod. J.* **18,** 228–230, 249, 251 (1939).
Winton, A. L., Winton, K. B., "Analysis of Foods," Wiley, New York, N.Y., Part 2, 904 (1935).
Wolford, R. W., Atkins, C. D., Dougherty, M. H., MacDowell, L. G., *Trans. Citrus Eng. Conf.* **14,** 64 (1968).
Wolford, R. W., Dougherty, M. H., Petrus, D. R., *Int. Fed. Fruit Juice Producers, Sci. Tech. Commun.* IX, 151 (1969).

Received for review May 14, 1971. Accepted August 13, 1971. Cooperative research by the Florida Department of Citrus and the University of Florida, IFAS, Agricultural Research and Education Center, Florida Agricultural Experiment Stations, Journal Series No. 3959. Presented at symposium on Chemistry of Essential Oils and Related Products, 161st ACS meeting, Los Angeles, California, March–April 1971.