Phytochemistry, 1975, Vol. 14, pp. 183–187. Pergamon Press. Printed in England.

SATURATED AND MONO-UNSATURATED LONG-CHAIN HYDROCARBON PROFILES FROM *CITRUS UNSHIU* JUICE SACS

HAROLD E. NORDBY and STEVEN NAGY

Citrus and Subtropical Products Laboratory*, Winter Haven, Florida, U.S.A.

(*Received* 14 *May* 1974)

Key Word Index—*Citrus unshiu*, *Rutaceae* satsuma mandarin cultivars; long-chain hydrocarbon profiles; chemotaxonomy.

Abstract—Six cultivars of satsuma mandarin—Kawano Wase, Owari, Silverhill, Foley, Sugiyama and Nobilis—were examined for content of saturated and mono-unsaturated long-chain hydrocarbons in their juice sacs. Linear hydrocarbons accounted for more than 53% of the saturates and more than 87% of the monoenes. In the saturated fraction the major linear hydrocarbon was C_{25} while in the monoene fraction C_{25} and C_{29} predominated. The ratios of linear/*iso* C_{23} and C_{25} saturates were greater than in other citrus previously investigated. Kawano Wase had profiles quite different from the other cultivars. Foley and Owari can be differentiated from the other cultivars by their linear monoene profiles. Long-chain hydrocarbon profiles intrinsic to the mandarin species, *C. unshiu*, were compared with profiles of non-mandarin species.

INTRODUCTION

Taxonomically mandarins are one of the most complex citrus fruits. They have been defined as 'including all highly pigmented oranges having a peel which is easily detached from the juice segments' [1]. The Japanese citriculturist, Tanaka, recognized 36 species of mandarins [2] while the American systematist, Hodgson, divided the mandarins into four species [1], viz *nobilis*, *deliciosa*, *reticulata* and *unshiu*.

This study is concerned with satsuma mandarin cultivars, viz Owari, Kawano Wase, Silverhill, Foley, Sugiyama and Nobilis belonging to the species, *Citrus unshiu*. The Owari satsuma is of unknown origin but is believed to have originated in Japan and was introduced into the United States more than 50 years ago [1]. Kawano Wase is a budsport of Owari imported from Japan in 1921. Silverhill, a nucellar seedling of Owari, introduced about 1931 [1], is the most commercially promising satsuma in the United States. Foley, another variation of Owari, was propagated in Southern Alabama in the early 1900's [3]. Sugiyama a bud variation of Owari [1] was imported from Australia. Nobilis, imported to the United States in 1934 from the U.S.S.R. has unknown parentage.

In recent years we have carried out extensive chemotaxonomic studies on citrus lipids. These investigations have included the profiles of fatty acids [4–9], sterols [10,11] and long-chain hydrocarbons [12–18]. Because each citrus species appears to have its own intrinsic pattern of long-chain hydrocarbons we have studied these most extensively. The present study was undertaken to determine if specific hydrocarbon patterns in *Citrus unshiu* might be useful in distinguishing this mandarin species from other citrus species.

RESULTS AND DISCUSSION

Table 1 shows the yields of total juice sac lipids for the six satsuma mandarins. The total hydrocarbon fraction represents 1·6–2·1% of the total juice sac lipids. A breakdown of the saturated and mono-unsaturated hydrocarbons shows that these two groups represent 1·3–1·8% and 0·2–0·5% of the total lipid content, respectively. Ratios of saturated to mono-unsaturated hydrocarbons ranged from 2·9 (Kawano Wase) to 9·7 (Foley). The 2·9 ratio for Kawano Wase is similar to ratios for Dancy (3·1) [16] and Honey (2·9) [19] mandarins of the species *reticulata*.

* One of the laboratories of the Southern Region, Agricultural Research Service, U.S. Department of Agriculture.

Table 1. Total lipid and hydrocarbon concentrations of satsuma juice sacs (mg/20 g dry wt)

Cultivar	Total lipid	Hydrocarbon fraction				
		Total	Saturated	Monoene	Ratio Sat./mono.
Kawano Wase	188 ± 7	3·9 ± 0·1	2·9 ± 0·1	1·0 ± 0·1	2·9
Silverhill	242 ± 11	4·9 ± 0·2	4·3 ± 0·2	0·6 ± 0·1	7·2
Owari	217 ± 4	4·2 ± 0·1	3·7 ± 0·1	0·5 ± 0·1	7·4
Foley	187 ± 8	3·2 ± 0·2	2·9 ± 0·2	0·3 ± 0·1	9·7
Sugiyama	246 ± 14	5·0 ± 0·2	4·3 ± 0·2	0·7 ± 0·1	6·1
Nobilis	170 ± 22	2·7 ± 0·1	2·2 ± 0·1	0·5 ± 0·1	4·4

Table 2 shows the saturated long-chain hydrocarbons in the six satsumas according to their linear, *iso*- and *anteiso*-structures. The average percentage of saturated linear hydrocarbons is 57·8%; the minimum was 53%. This high percentage of linear hydrocarbons was observed previously in three lemon cultivars [17], sour limes [15] and Dancy mandarin [16]. Kawano Wase with a value of 67·2% is similar to that observed for Dancy mandarin (65·3%) [16]. Odd-numbered hydrocarbons account for most of the linear saturates; C_{23} and C_{25} were the most prominent as observed previously in other citrus fruits. The percentages of linear C_{23} saturated hydrocarbon ranges from 11·9% (Foley) to 14·6% (Sugiyama); the same ranges have been reported for grapefruit [14] and tangors [12]. The range of linear C_{25} in satsumas is much broader, i.e. 18·1% in Nobilis to 26·2% in Kawano Wase. Only Dancy mandarin (19·5%) [16] and the Temple × Kinnow mandarin hybrid (18·5%) [12] had been reported to have such high percentages. Apparently a high percentage of linear saturated C_{25} may be a good index of mandarin parentage. Previously we reported that the linear saturated C_{23}/C_{25} ratios showed ranges which were intrinsic for each citrus species [13,15]. These ranges were: *C. sinensis* (sweet oranges, 1·05–1·34), *C. paradisi* (grapefruit, 0·85–0·94), *C. limon* (lemons, 0·30–0·47), *C. aurantifolia* and *C. latifolia* (sour limes, 0·38–0·39) and *C. reticulata* (mandarins, 0·62–0·69). In this current study satsuma C_{23}/C_{25} ratios varied from 0·50 (Kawano Wase) to 0·81 (Sugiyama) with an overall average of 0·66. If the two mandarin species, *reticulata* and

Table 2. Saturated long-chain hydrocarbon profiles of satsuma juice sac (wt%)

Cultivar	Carbon number: Odd-numbered 21	23	25	27	29	31	33	35	Even-numbered 20	22	24	26	28	30	32	34	Total odd no.	Total even no.	Total
Linear																			
Kawano Wase	0·30	13·11	26·18	9·27	2·46	0·85	0·08	tr*	0·08	1·24	6·34	5·13	1·37	0·63	0·15	tr	52·25	14·94	67·19
Silverhill	0·44	14·23	19·47	4·94	1·87	1·54	0·29	tr	0·10	1·58	6·09	3·01	0·69	0·52	0·30	0·06	42·78	12·35	55·13
Owari	0·40	14·10	21·83	5·18	1·39	0·69	0·12	tr	0·30	1·41	6·41	3·39	0·73	0·45	0·28	tr	43·71	12·97	56·68
Foley	0·32	11·90	23·47	6·08	2·13	1·35	0·23	tr	0·10	1·03	6·93	3·77	0·88	0·58	0·27	tr	45·48	13·56	59·04
Sugiyama	0·57	14·63	18·11	4·70	1·58	1·01	0·16	tr	0·12	1·69	6·38	3·16	0·63	0·36	0·15	tr	40·76	12·49	53·25
Nobilis	0·57	13·16	18·05	6·10	2·18	0·80	0·07	tr	0·19	1·82	6·53	3·99	1·28	0·64	0·14	tr	40·93	14·59	55·52
Avg. Linear	0·43	13·52	21·19	6·05	1·94	1·04	0·16	tr	0·15	1·46	6·45	3·74	0·93	0·53	0·22	tr	44·32	13·48	57·80
Iso-branched																			
Kawano Wase	tr	4·13	8·66	4·30	1·02	0·20	tr	†	tr	0·28	0·93	0·51	0·38	0·18	0·04		18·31	2·32	20·63
Silverhill	0·04	6·64	9·48	3·23	0·86	0·21	tr		0·06	0·20	1·62	0·69	0·24	0·11	tr	tr	20·46	2·92	23·38
Owari	0·04	7·59	10·65	4·20	1·01	0·33	tr		tr	0·13	1·35	0·80	0·31	0·16	0·03		23·82	2·78	26·60
Foley	tr	6·21	12·37	3·67	0·88	0·28	tr		tr	0·02	1·89	1·16	0·34	0·06			23·41	3·47	26·88
Sugiyama	0·02	7·94	9·35	3·11	0·81	0·17	tr		0·01	0·04	1·71	0·90	0·22	0·15	0·03		21·40	3·06	24·46
Nobilis	0·02	7·14	12·02	4·23	1·10	0·23	tr		tr	0·07	1·31	1·17	0·59	0·20	tr		24·74	3·34	28·08
Avg. *Iso*	0·02	6·61	10·42	3·79	0·95	0·24	tr		0·01	0·12	1·47	0·87	0·34	0·14	0·02		22·02	2·98	25·01
Anteiso-branched																			
Kawano Wase	tr	tr	0·76	0·36	0·30	0·02	tr		tr	tr	3·53	4·87	1·80	0·44	0·10		1·44	10·74	12·18
Silverhill	tr	tr	1·49	0·71	0·18	0·07	tr		0·01	0·09	7·99	7·91	2·37	0·48	0·19	tr	2·45	19·04	21·49
Owari	tr	tr	1·70	0·90	0·24	0·08	tr		tr	0·07	5·94	5·51	1·78	0·38	0·12		2·92	13·80	16·72
Foley	tr	tr	1·18	0·50	0·09	tr			tr	0·01	5·44	5·30	1·27	0·29	tr		1·77	12·31	14·08
Sugiyama	tr	tr	1·72	0·92	0·21	0·06	tr		tr	0·04	8·69	7·64	2·29	0·61	0·11		2·91	19·38	22·29
Nobilis	tr	tr	0·40	0·75	0·34	0·03	tr		tr	0·04	6·00	6·17	1·95	0·61	0·11		1·52	14·88	16·40
Avg. *Anteiso*	tr	tr	1·21	0·69	0·23	0·04	tr		tr	0·04	6·26	6·23	1·91	0·47	0·11		2·17	15·03	17·19

* Trace, less than 0·01%.
† Not detected under GLC parameters but may be present below 0·001%.

unshiu, are combined the C_{23}/C_{25} ratio range of 0·50–0·81 would still be different from other citrus species. Totals of even-numbered linear hydrocarbons ranged from 12·4 to 14·9% with C_{24} and C_{26} paraffins predominating.

About one-fourth of all satsuma saturates have an *iso*-branched structure consisting of 87–89% odd- and 11–13% even-numbered paraffins. The major *iso*-branched paraffins are C_{23}, C_{25} and C_{27}; collectively these three comprise *ca* 84% of all *iso*-branched paraffins. Kawano Wase appears different from the other five satsumas by showing lower percentages for *iso*-C_{23}, -C_{24}, -C_{25} and -C_{26}. The percentages observed for odd-numbered, *iso*-C_{23}, -C_{25}, -C_{27}, and -C_{29} can be used to differentiate satsuma mandarins from sweet oranges [12,13,18], grapefruit [14], lemons [17], limes [15], tangors [12] and tangelos [16].

Satsumas vary considerably in their saturated *anteiso*-branched paraffin content with a range from 12·2 (Kawano Wase) to 22·3% (Sugiyama). This difference is due primarily to the differing levels of C_{24} and C_{26}. In contrast to *iso*-branched paraffins where odd-numbered structures predominated, *anteiso*-branched paraffins show a predominance of even-numbered structures. The *anteiso* group shows relatively consistent percentages for even- (83–91%) and odd-numbered (9–17%) structures. The three major *anteiso*-branched paraffins are C_{24}, C_{26} and C_{28}. The *anteiso* C_{24}/C_{26} ratio for the six satsumas range from 0·72 (Kawano Wase) to 1·14 (Sugiyama) with an overall average of 0·99. This range may be useful in distinguishing satsuma mandarins from sweet oranges (1·21–1·76) [12,13,18] lemons (0·40–0·72) [17], sour limes (0·37–0·50) [15] and tangelos (1·44–1·61) [16] but does not appear capable of differentiating satsumas from grapefruit (0·93–1·17) [14].

Scrutiny of the ratios of linear C_{23} and C_{25} to *iso*-C_{23} and -C_{25} reveals some interesting comparative data. The average ratios of linear C_{23}/*iso*-C_{23} and linear C_{25}/*iso*-C_{25} are 2·05 and 2·03, respectively. With the exception of Nobilis with a linear C_{25}/*iso*-C_{25} ratio of 1·50, all satsumas have ratios greater than any of the other citrus species previously examined except Dancy mandarin.

Table 3 shows the mono-unsaturated hydrocarbons present in satsuma juice sacs. Linear alkenes account for 87·6 (Foley and Nobilis) to 97·4% (Kawano Wase) of these hydrocarbons. This range is 10% higher than that of other citrus species except grapefruit (87·6–95·0) [14] and Dancy mandarin (98·0) [16]. From 89 to 94% of these linear hydrocarbons are odd-numbered, the major compounds being in the C_{23} to C_{33} range. The average percentages for the six major odd-numbered linear monoenes, i.e. C_{23}, C_{25}, C_{27}, C_{29}, C_{31}, C_{33} are 7,

Table 3. Mono-unsaturated long-chain hydrocarbon profiles of satsuma juice sacs (wt%)

	Carbon number																		
	Odd-numbered								Even-numbered										
Cultivar	21	23	25	27	29	31	33	35	20	22	24	26	28	30	32	34	Total odd no.	Total even no.	Total
Linear																			
Kawano Wase	0·20	1·78	9·89	11·67	32·76	33·21	2·13	tr	0·18	0·32	0·54	0·96	0·99	2·02	0·76	tr	91·64	5·77	97·41
Silverhill	0·03	8·52	24·40	17·97	18·27	12·33	1·38	tr	tr	0·39	2·26	2·14	1·17	1·00	0·19	tr	82·90	7·15	90·05
Owari	0·08	11·13	23·45	18·26	16·27	14·58	1·63	tr	tr	0·37	1·69	2·15	1·17	1·74	0·45	tr	85·40	7·57	92·97
Foley	0·15	3·36	26·74	20·11	22·24	5·27	0·93	tr	0·35	0·48	2·49	2·60	1·45	0·88	0·25	tr	78·80	8·50	87·30
Sugiyama	0·05	9·01	19·24	15·38	22·26	18·28	1·97	tr	0·09	0·35	1·58	1·73	0·98	1·13	0·34	tr	86·19	6·20	92·39
Nobilis	0·12	8·19	20·77	13·64	19·18	17·14	1·02	tr	0·41	0·86	2·57	2·04	1·78	1·08	0·51	tr	80·06	9·25	89·31
Avg. Linear	0·11	7·00	20·75	16·17	21·83	16·80	1·51	tr	0·17	0·46	1·86	1·94	1·26	1·31	0·42	tr	84·13	7·41	91·57
Iso-branched																			
Kawano Wase	tr	0·15	0·64	0·39	0·11	tr	—	—	0·06	tr	0·06	0·07	0·01	tr	—	—	1·29	0·20	1·49
Silverhill	tr	0·76	2·28	1·15	0·50	0·17	0·07	—	tr	tr	0·18	0·21	tr	tr	—	—	4·93	0·39	5·32
Owari	tr	0·60	1·53	1·01	0·37	0·35	tr	—	tr	tr	tr	tr	tr	tr	tr	—	3·86	tr	3·86
Foley	0·11	0·20	3·95	1·79	0·55	0·51	tr	—	0·08	tr	0·15	0·33	tr	tr	tr	—	7·11	0·56	7·67
Sugiyama	0·02	0·61	1·65	0·81	0·35	0·10	tr	—	0·03	0·01	0·15	tr	tr	tr	tr		3·54	0·19	3·73
Nobilis	0·04	0·56	2·49	2·05	0·23	0·75	tr	—	0·11	0·07	0·19	tr	tr	tr	tr		6·12	0·37	6·49
Avg. *Iso*	0·03	0·48	2·09	1·20	0·35	0·31	tr	—	0·05	0·01	0·12	0·10	tr	tr	tr		4·48	0·29	4·76
Anteiso-branched																			
Kawano Wase	tr	0·06	0·12	0·13	tr	tr	—	—	0·04	0·05	0·14	0·45	0·08	tr	0·03	—	0·31	0·79	1·10
Silverhill	tr	0·07	0·42	0·11	tr	tr	—	—	tr	tr	1·17	2·10	0·54	0·13	0·09		0·60	4·03	4·63
Owari	tr	0·20	0·38	0·21	0·06	tr		—	tr	0·03	0·54	1·29	0·36	0·06	0·04	—	0·85	2·32	3·17
Foley	tr	0·29	0·46	tr	tr	tr	tr	—	0·08	0·16	0·44	2·99	0·49	0·12	tr		0·75	4·28	5·03
Sugiyama	0·01	0·12	0·33	0·17	tr	tr		—	0·03	0·03	0·90	1·71	0·40	0·13	0·05		0·63	3·25	3·88
Nobilis	0·08	0·18	0·43	tr	tr	tr	tr	—	0·19	0·11	1·00	1·09	0·74	0·38	tr		0·69	3·51	4·20
Avg. *Anteiso*	0·01	0·15	0·36	0·10	tr	tr	—		0·06	0·06	0·70	1·61	0·44	0·14	0·04	—	0·64	3·03	3·67

21, 16, 22, 17 and 1·5%, respectively. Some cultivars deviate considerably from these averages, e.g. C_{23} for Kawano Wase and Foley show one-half the average C_{23} value (7%) while Owari shows twice this average.

Iso-branched monoenes comprise between 1·4 and 7·7% of the total monoene fraction. Of this group, odd-numbered *iso* structures comprise 87–100% while even-numbered *iso* monoenes ranged from a trace to 13%. The Owari satsuma is noticeably different from other satsumas in showing predominately odd-numbered *iso* monoenes and only a trace of even-numbered *iso* structures. The two major *iso* monoenes in all satsumas are C_{25} and C_{27}. *Anteiso*-branched monoenes comprise between 1·10 and 5·03% of the total monoene fraction. The percentage distribution of *anteiso* monoenes is essentially the reverse of the *iso*-monoenes, i.e. even-numbered > odd-numbered. Even-numbered *anteiso* monoenes ranged from 72 (Kawano Wase) to 87% (Silverhill) and odd-numbered *anteiso* monoenes from 13 to 28%. The four major anteiso monoenes are C_{24}, C_{25}, C_{26} and C_{28} with C_{26} predominating.

Satsuma long-chain hydrocarbons can generally be distinguished from those of non-mandarin citrus species. However, cultivars within the *unshiu* species except for Kawano Wase are not easily differentiated. Foley and Owari can be identified from their linear monoene profiles. Nobilis has the only low saturated linear/*iso* C_{25} ratio (1·50%). Profiles of Silverhill and Sugiyama do not have any differentiating characteristics. The long-chain hydrocarbon profiles of Kawano Wase resembles those previously published for Dancy mandarin [16]. The question as to whether long-chain hydrocarbon patterns of *C. unshiu* can be used to distinguish this mandarin species from other mandarin species is still to be answered.

The odd/even *n*-alkane ratios reported previously for citrus juice sacs [12–18] and the ratios reported herein range from 2·6 to 9·8. This range, which is distinctively low for flowering plants [20–26], is undoubtedly due to the high level of even-numbered *n*-alkanes and branched alkanes present in citrus. Generally, for higher plants the major hydrocarbons are C_{29}, C_{31} and C_{33} [20]. Citrus juice sac alkanes are unique in that C_{23} and C_{25} predominate. Recently four species of moss [27] and three species of the genus *Balanites* [28] also have been reported to contain predominant amounts of C_{23} and C_{25}. The range of odd/even alkane ratios for the moss is quite similar to those of citrus. Herbin and Robins [29] have reported that internal area of leaves contained more even-numbered alkanes than the external portion. The cuticular wax of the cranberry has an alkane pattern characteristic of plant cuticular waxes [30]; however, the major alkanes in the seed of the cranberry [31] are all even-numbered with only minor amounts of the common C_{27} and C_{29} *n*-alkanes. In citrus however, there is no difference in alkane patterns between internal parts of the fruit (juice sacs) and the external parts (peel oil) [32].

In most plants branched hydrocarbons are usually minor alkane components; however, in some e.g. tobacco, they may range up to 50% [25]. The branched alkanes are predominately odd-numbered and the even-numbered alkanes are virtually absent in tobacco. Citrus fruits are unique in having both *iso*- and *anteiso*-branched structures of both odd- and even-numbered hydrocarbons. Mono-unsaturated alkenes are found in a number of higher plants including sugar cane and roses [33]. Citrus, like these two higher plants, contain major amounts of $C_{27:1}$, $C_{29:1}$, $C_{31:1}$, $C_{33:1}$.

EXPERIMENTAL

Isolation and purification of satsuma juice sac lipids. Samples of mature Sugiyama, Silverhill, Nobilis, Owari, Foley and Kawano Wase satsuma mandarins were obtained from Whitmore Foundation Farm (U.S. Horticultural Research Laboratory, USDA, Orlando, FL). The fruit were cut in half and the intact juice sacs (vesicles) carefully separated from core, peel, seeds and carpellary membrane with the aid of a citrus spoon. The juice sacs were freeze-dried to a powder (moisture content no greater than 4%) and stored at −18° until lipids were extracted. Lipids were extracted from 20 g of juice sac powder with distilled $CHCl_3$ and MeOH by a method described for total juice sac powder [34]. Quadruplicate extractions were made on single batches of fruit from each cultivar and were purified on Sephadex columns [34].

Column and TLC. Columns, 0·9 × 30 cm, containing 10 g Merck, 70–325 mesh silica gel were washed by elution with 100 ml distilled $CHCl_3$. The total purified lipid (*ca* 140–200 mg) was dissolved in distilled $CHCl_3$ and percolated into this pre-washed column. Neutral lipids were eluted with 200 ml distilled $CHCl_3$. The neutral lipid fraction was concentrated to a small vol. and streaked on precoated silica gel G plates (20 × 20 cm, 500 μm). The plates were developed at room temp. in hexane–Et_2O (9:1). The band corresponding to the long-chain hydrocarbon and containing a portion of overlapped carotenoid esters was scraped from the plate and eluted with Et_2O. This fraction was in turn restreaked on a $AgNO_3$-impregnated silica gel G plate [4] and developed in 2% Et_2O in light petrol (30–60° boiling range), which separated the saturated and mono-unsaturated hydrocarbons leaving the carotenoid esters at the origin. These two hydrocarbon fractions were eluted with Et_2O and dry wts taken. The monene fraction was dissolved in 1 ml hexane and hydrogenated in a Parr apparatus with 10 mg 10% Pd-C catalyst under 50 lbs/in^2 at room temp. for 1 hr.

Quantitation. Hydrocarbons were analyzed by GLC with an F & M Model 7610A FID GC. The analyses were determined on a glass column (3·05 m length and 4 mm i.d.) coated with 3% SP-1000 (Supelco, Inc., Bellefonte, PA) on 100–120 mesh Gas Chrom Q. The injection port was at 265° and the detector at 275°. He flow was 80 ml/min at 40 lbs/in^2. The sample (1·5–4·5 λ representing 1–10% hydrocarbon in hexane) was injected on-column at 160° and programmed at 4°/min for 3 min, 2°/min for 19 min, then 4°/min up to 260° and finally held isothermally at 260° until the C_{35} hydrocarbon eluted from the column. Quantitative measurements were obtained with an Autolab System IV computing integrator for chromatography. Quantitative mixtures of hydrocarbon standards were prepared to verify the linear response of the FID as well as the computing integrator. Representative hydrocarbon samples from previous studies [14,16] were also injected to correlate data obtained by disc integration quantitations and by triangulation with the computing integrator data presented in this study. Each value shown in Table 2 and 3 represents the mean of 5–10 determinations. The coefficient of variation (CV) for several mean ranges (MR) showed the following: MR 0·01–0·10; CV 10–35%; MR 0·1–1·0; CV 5–10%; MR 1·0–5·0; CV 3–5%; MR above 5·0; CV less than 2%.

REFERENCES

1. Hodgson, R. W. (1967) *The Citrus Industry*, (Reuther, W., Webber, H. J. and Batchelor, L. D., eds.) Vol. 1, pp. 496–505, University of California Press, Berkeley, California.
2. Tanaka, T. (1954) *Jap. Soc. Prom. Sci.* 152.
3. Hearn, J., USDA, Orlando, Florida, personal communication.
4. Nordby, H. E. and Nagy, S. (1969) *Phytochemistry* **8,** 2027.
5. Nordby, H. E. and Nagy, S. (1971) *Phytochemistry* **10,** 615.
6. Nordby, H. E. and Nagy, S. (1971) *Lipids* **6,** 554.
7. Nagy, S. and Nordby, H. E. (1974) *Phytochemistry* **13,** 153.
8. Nordby, H. E. and Nagy, S. (1974) *Phytochemistry* **13,** 443.
9. Nordby, H. E. and Nagy, S. (1974) *Phytochemistry* **13,** 2215.
10. Nagy, S. and Nordby, H. E. (1971) *Lipids* **6,** 826.
11. Nordby, H. E. and Nagy, S. (1973) *J. Chromatogr.* **79,** 147.
12. Nagy, S. and Nordby, H. E. (1971) *Phytochemistry* **10,** 2763.
13. Nagy, S. and Nordby, H. E. (1972) *Lipids* **7,** 666.
14. Nagy, S. and Nordby, H. E. (1972) *Phytochemistry* **11,** 2789.
15. Nagy, S. and Nordby, H. E. (1972) *Phytochemistry* **11,** 2865.
16. Nagy, S. and Nordby, H. E. (1972) *Lipids* **7,** 722.
17. Nordby, H. E. and Nagy, S. (1972) *Phytochemistry* **11,** 3249.
18. Nagy, S. and Nordby, H. E. (1973) *Phytochemistry* **12,** 801.
19. Nordby, H. E. and Nagy, S. Unpublished data.
20. Eginton, G. and Hamilton R. J. (1963) in *Chemical Plant Taxonomy*, (Swain, T. ed.), p. 187. Academic Press, London.
21. Weete, J. D. (1972) *Phytochemistry* **11,** 1201.
22. Stransky, K., Streible, M. and Herout, V. (1967) *Czeck. Chem. Commun.* **32,** 3213.
23. Lytle, T. F. and Sever, J. R. (1973) *Phytochemistry* **12,** 623.
24. Butlari, F., Marsili, A., Morelli, I. and Pacchiani, M. (1972) *Phytochemistry* **11,** 2519.
25. Wilkinson, R. E. (1972) *Phytochemistry* **11,** 2439.
26. Holman, R. and Nichols, P. C. (1972) *Phytochemistry* **11,** 333.
27. Corrigan, D., Kloos, C., O'Connor, C. S., and Timoney, R. F. (1973) *Phytochemistry* **12,** 213.
28. Hardman, R., Wood, C. N., and Safowora, E. A. (1970) *Phytochemistry* **9,** 1087.
29. Herbin, G. A. and Robins, P. A. (1969) *Phytochemistry* **8,** 1985.
30. Croteau, R. and Fagerson, I. S. (1971) *Phytochemistry* **10,** 3239.
31. Croteau, R. and Fagerson, I. S. (1969) *Phytochemistry* **8,** 2219.
32. Hunter, G. L. K. and Brogden, W. B. (1966) *Phytochemistry* **5,** 807.
33. Kolattukudy, P. E. (1970) *Lipids* **5,** 257.
34. Nagy, S. and Nordby, H. E. (1969) *J. Agr. Food Chem.* **18,** 593.