Phytochemistry, 1975, Vol. 14, pp. 1777–1782. Pergamon Press. Printed in England.

SATURATED AND MONO-UNSATURATED LONG-CHAIN HYDROCARBON PROFILES FROM MANDARIN JUICE SACS

HAROLD E. NORDBY and STEVEN NAGY

Citrus and Subtropical Products Laboratory,* Winter Haven, FL 33880, U.S.A.

(*Received* 21 *October* 1974)

Key Word Index—*Citrus nobilis*; *Citrus deliciosa*; Rutaceae; mandarin cultivars; long-chain hydrocarbon profiles; chemotaxonomy.

Abstract—The long-chain saturated and mono-unsaturated hydrocarbon content of the juice sacs of five mandarin cultivars (Mediterranean, Honey, Wilking, Kinnow, King) were examined. Normal homologues accounted for more than 47% of the saturated and more than 75% of the monoene hydrocarbons. In the saturated fraction the major hydrocarbon was n-C_{25} but in the monoene fraction n-C_{25} predominated in Kinnow and King while C_{29} predominated in Mediterranean, Honey and Wilking. All five cultivars could be differentiated from each other and from other citrus species by their hydrocarbon patterns. The noticeably high normal/iso ratios of saturated C_{23} and C_{25} hydrocarbons previously shown to be characteristic of mandarin species, *Citrus unshiu* and *C. reticulata*, were also found in *C. nobilis* and *C. deliciosa*.

INTRODUCTION

The taxonomic classification of citrus species has been the subject of considerable controversy, but within the past two decades, chemo-systematic studies on secondary plant constituents has become an important tool in defining and differentiating the complex genus. Previous investigations on citrus fatty acids [1–6], sterols [7,8] and long-chain hydrocarbons [9–16] indicate that lipids may be reliable markers in the chemotaxonomy of citrus fruits.

Mandarin fruit appear to be the most complex and least definable of the genus, *Citrus* and Hodgson [17] has divided them into four species, viz. *nobilis*, *reticulata*, *deliciosa* and *unshiu*. In a preliminary study [16] of the species, *unshiu*, six satsuma cultivars, viz. Silverhill, Owari, Foley, Sugiyama, Nobilis and Kuwano Wase were investigated for their long-chain hydrocarbon patterns. With the exception of Kuwano Wase, all satsuma cultivars had similar hydrocarbon patterns. The hydrocarbon pattern of Kuwano Wase was similar to that of Dancy tangerine [13] (*C. reticulata*), the most common American mandarin. Two mandarin species, *nobilis* and *deliciosa* and three of their progeny were investigated in the present study. The seed parent, King, was imported to Florida from South Vietnam in 1882 as budwood and as seedlings. Although classified as a mandarin, King is thought to be a natural tangor of unknown parentage [17]. The other parent, Mediterranean, originated in Italy around 1810 and was believed to have developed as a chance seedling from a Chinese mandarin. The three hybrids resulting from the King × Mediterranean cross were developed at the University of California Citrus Research Center. These three hybrids, viz. Kinnow, Honey and Wilking, are of commercial importance. The present study was undertaken to determine if hybrids formed by crossing two mandarin species *C. nobilis* cv King and *C. deliciosa* cv Mediterranean would show hydrocarbon patterns typical of their parents and of mandarin species in general.

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

Table 1. Hydrocarbon concentrations of mandarin juice sacs (mg/20 g dry wt)

Cultivar	Total	Hydrocarbon Saturated	Monoene	Ratio sat. mono.
Mediterranean	3·4 ± 0·2	2·8 ± 0·2	0·6 ± 0·1	4·7
Honey	2·5 ± 0·2	1·9 ± 0·2	0·6 ± 0·1	3·2
Wilking	4·5 ± 0·2	4·0 ± 0·2	0·5 ± 0·1	8·0
Kinnow	3·7 ± 0·1	3·2 ± 0·1	0·2 ± 0·1	16·0
King	4·2 ± 0·1	4·0 ± 0·1	0·2 ± 0·1	20·0

RESULTS AND DISCUSSIONS

The range of total hydrocarbons (2·5 to 4·5 mg/20 g juice sac powder) (Table 1) is consistent with the ranges found in other citrus [11,13–16] except limes [12]. All five cultivars contain saturated hydrocarbons within the 1·9 to 4 mg range. One parent (Mediterranean) and two hybrids (Honey and Wilking) however, have monoene contents (0·5–0·6 mg) 2–3 times that of the other parent, King, and Kinnow hybrid. This difference results in the Kinnow and King cultivars having saturated to monoene ratios higher than those of the other three cultivars and of other reported mandarins [10,13,16].

Table 2 shows the composition of the *n*- and *br*-saturated hydrocarbons in the five mandarins. The mean percentage of *n*-hydrocarbons is 57·2%, similar to that reported for the satsuma mandarins [16] and is in the high percentage range found in lemons [14], sour limes [12] and Dancy tangerine [13]. All cultivars, except King, comprise at least 56% *n*-hydrocarbons. The King, with a *n*-hydrocarbon content of 47%, resembles that of sweet oranges [9,10,15] and may be a reflection of King's tangor character. The three mandarin hybrids have contents intermediate between their parents, King and Mediterranean. The Mediterranean mandarin has a composition (62·3%) similar to that of Dancy tangerine [13].

As observed in other citrus fruits, odd-numbered hydrocarbons account for the majority of *n*-saturated hydrocarbons, with C_{23} and C_{25}

Table 2. Saturated hydrocarbon

Cultivar	Carbon Odd-numbered 21	23	25	27	29	31	33	35
Normal								
1. Mediterranean	0·44	16·18	20·55	6·79	2·04	0·82	*	I
2. Honey	0·46	14·86	18·84	6·32	1·82	0·88	t	t
3. Wilking	0·33	16·26	21·12	4·80	1·18	0·24	0·07	
4. Kinnow	0·38	14·04	20·99	5·22	1·39	1·21	0·35	t
5. King	0·32	11·87	18·25	3·84	0·82	0·67	0·23	0·08
Mean	0·39	14·64	19·95	5·39	1·45	0·76	0·13	0·02
Iso-branched								
1. Mediterranean	0·04	5·65	7·85	2·80	0·75	0·19	t	
2. Honey	t	6·44	9·47	3·65	1·24	0·21		
3. Wilking	t	4·86	13·41	4·20	0·85	0·36		
4. Kinnow	0·07	3·56	9·73	4·45	1·16	0·44	t	
5. King	t	6·26	12·25	4·97	0·95	0·33	0·06	t
Mean	0·02	5·35	10·54	4·01	0·99	0·31	0·01	
Anteiso-branched								
1. Mediterranean	t	t	1·50	0·69	0·21	0·04		
2. Honey	t	t	1·35	0·56	0·29	0·09		
3. Wilking	t	t	1·49	0·63	0·07	t		
4. Kinnow	t	t	1·45	0·91	0·20	0·11	t	
5. King	t	t	1·83	1·02	0·24	0·13	0·04	t
Mean	t	t	1·52	0·76	0·20	0·07	t	

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

being dominant. The content of n-C_{23} hydrocarbon ranges from 11·9 (King) to 16·3 (Wilking) but in four of the five cultivars (King excluded) shows a narrower range of 14·0 to 16·3%. This high proportion has only been observed in sweet oranges [9,15] and tangors [13]. The content of n-C_{25} hydrocarbon ranges from 18·2 (King) to 21·1% (Wilking) and these amounts have only been previously observed in satsuma mandarins [16], Dancy tangerine [13], and a Temple × Kinnow mandarin hybrid [9]. This evidence verifies a previous postulate [16] that a high percentage of n-C_{25} strongly indicates mandarin parentage. Previously these authors suggested that n-C_{23}/n-C_{25} ratios may be used to differentiate between citrus species [10, 12]. The unique n-C_{23}/n-C_{25} range for *reticulata* [10] and *unshiu* [16] is also observed for *nobilis* and *deliciosa*. The ratios of Mediterranean (0·79), Honey (0·79) and Wilking (0·77) are different from those of King (0·65) and Kinnow (0·67), but all values are within the mandarin range. Even-numbered n-hydrocarbons range from 11·2 to 17·6% and the major homologues in all five mandarins are C_{24} and C_{26}. These compounds as well as the other minor hydrocarbons occur in the same relative proportions as those reported for satsumas [16].

About 25% of the saturated hydrocarbon fraction is composed of *iso*-branched homologues and the proportion ranges from 19·6% in Mediterranean to 28·2% in King; the three hybrids fall between these values. From 86 to 88% of the *iso*-hydrocarbons are odd-numbered and C_{23}, C_{25}, and C_{27} together comprise *ca* 94%. In contrast to the n-C_{23}, n-C_{25} and n-C_{27} homologues, the *iso*-homologues of the King parent are higher in relative percentage than the Mediterranean parent. *Iso*-C_{23},-C_{25},-C_{27}, and -C_{29} may be used to differentiate these mandarins from sweet oranges [9,10,15], grapefruit [11], lemons [14], limes [12], tangors [9] and tangelos [13] but cannot differentiate between different mandarin species.

The King and Mediterranean cultivars and their hybrids vary slightly in their contents of saturated *anteiso*-branched hydrocarbons. King with over 9% *anteiso*-C_{26} has the highest percentage reported for this hydrocarbon in citrus fruits. In contrast to *iso*-hydrocarbons where odd-numbered compounds predominate, *anteiso*-branched paraffins show a predominance of even-numbered

profiles of mandarin juice sac (wt. %)

number			Even-numbered							
20	22	24	26	28	30	32	34	Total odd no.	Total even no.	Total
0·21	2·19	7·43	4·06	1·17	0·35	0·11		46·79	15·52	62·31
0·20	1·72	6·17	3·46	1·02	0·41	0·13	t	43·18	13·11	56·29
0·24	1·51	8·00	3·71	0·82	0·28	0·18	t	44·00	14·74	58·74
0·36	2·03	8·58	5·02	1·06	0·42	0·14	t	43·58	17·61	61·19
0·13	1·49	5·94	2·64	0·44	0·35	0·27	t	36·08	11·26	47·34
0·23	1·79	7·22	3·78	0·90	0·36	0·17	t	42·73	14·45	57·18
t	0·43	0·96	0·57	0·26	0·14	t		17·28	2·36	19·64
t	0·08	1·43	0·82	0·35	0·14	0·02		21·01	2·84	23·85
t	t	2·16	1·17	0·15	0·05	t		23·68	3·53	27·21
t	0·05	1·50	0·99	0·38	0·12	0·03		19·41	3·07	22·48
t	0·09	1·73	1·03	0·32	0·13	0·08	t	24·82	3·38	28·20
t	0·13	1·56	0·92	0·29	0·12	0·03		21·24	3·04	24·28
t	0·06	6·55	6·27	2·12	0·61	t		2·44	15·61	18·05
t	0·03	6·80	7·06	2·80	0·81	0·07		2·29	17·57	19·86
t	0·14	3·98	5·81	1·47	0·46	t		2·19	11·86	14·05
t	t	3·82	6·18	2·79	0·66	0·21		2·67	13·66	16·33
t	0·03	6·91	9·22	3·81	0·94	0·29	t	3·26	21·20	24·46
t	0·05	5·61	6·91	2·60	0·70	0·11		2·53	15·98	18·53

Table 3. Mono-unsaturated long-chain hydrocarbon

								Carbon
				Odd-numbered				
Cultivar	21	23	25	27	29	31	33	35
Normal								
1. Mediterranean	0·12	2·51	15·59	12·61	30·36	25·18	0·71	
2. Honey	0·21	2·91	17·16	15·39	26·11	19·88	t	
3. Wilking	0·11	1·42	14·74	15·05	28·09	20·80	t	
4. Kinnow	0·47	2·49	31·73	19·48	5·76	2·03	t	
5. King	0·15	4·37	32·79	22·14	4·52	1·57	t	
Mean of Linear Nos. 1, 2, 3	0·15	2·28	15·83	14·35	28·19	21·95	0·24	
Mean of Linear Nos. 4, 5	0·31	3·43	32·26	20·81	5·14	1·80	t	
Iso-branched								
1. Mediterranean	t	0·37	1·47	0·81	0·12	0·20	t	
2. Honey	0·15	0·51	1·89	1·38	0·73	0·38		
3. Wilking	t	0·28	3·31	1·97	0·54	0·73	t	
4. Kinnow	0·51	0·66	6·88	4·81	1·55	1·22		
5. King	t	1·16	6·53	4·04	0·87	0·62		
Mean of *Iso* Nos. 1, 2, 3	0·05	0·39	2·22	1·39	0·46	0·44	t	
Mean of *Iso* Nos. 4, 5	0·25	0·91	6·71	4·43	1·21	0·92		
Anteiso-branched								
1. Mediterranean	t	0·20	0·29	0·16	0·05	0·14	t	
2. Honey	t	0·19	0·09	t	t	t		
3. Wilking	t	0·14	0·21	0·30	0·05	t	t	
4. Kinnow	t	0·43	0·87	0·68	t	t		
5. King	t	0·19	0·81	0·61	0·12	0·13		
Mean of *Anteiso* Nos. 1, 2, 3	t	0·18	0·20	0·15	0·03	0·05	t	
Mean of *Anteiso* Nos. 4, 5	t	0·31	0·84	0·65	0·06	0·07		

compounds (84–87%). The three major *anteiso*-hydrocarbons are C_{24}, C_{26} and C_{28}. The *anteiso* C_{24}/C_{26} ratios for the five mandarins range from 0·62 (Kinnow) to 1·04 (Mediterranean) and parallels those observed for satsumas [16].

Examination of the ratios of *n*-C_{23} and *n*-C_{25} to *iso*-C_{23} and *iso*-C_{25} reveals that, like the satsumas, these mandarin ratios are relatively higher than those of other citrus. The mean ratios of *n*-C_{23}/*iso*-C_{23} and *n*-C_{25}/*iso*-C_{25} are 2·87 and 1·97, respectively. Three of the five mandarin cultivars have ratios greater than those of any non-mandarin species previously examined.

The mono-unsaturated hydrocarbons present in mandarin juice sacs are shown in Table 3. *n*-Alkenes account for 74·9 (King) to 93·9% (Mediterranean) of these hydrocarbons and the patterns can be further divided into two groups. Mediterranean and its hybrids, Honey and Wilking, have mean *n*-alkene percentages of 90·9% and King and Kinnow have 75·0%. The *n*-alkene profiles of King and Kinnow are very similar, but they differ markedly from every other citrus species in having high values for C_{25} and C_{27} (32·3 and 20·8% respectively) and low values for the remaining alkenes. Mediterranean, Honey and Wilking have *n*-alkene profiles similar to those of Dancy tangerine [13] with high percentages of C_{29} and C_{31}. Of these three cultivars, Mediterranean has the highest proportion of these two alkenes and we suspect, from profiles of other mandarins [18], that the lower values for Honey and Wilking are due to a hybridization dilution effect.

Iso-branched monoenes comprise between 3·3 and 17·1% of the total monoene fraction and Mediterranean, Wilking and Honey can be differentiated from King and Kinnow by the percentage of these monoenes. King and Kinnow contain *ca* three times the amount of *iso*-alkenes as the other three cultivars and results from a greater content *iso*-C_{25} and *iso*-C_{27}.

Anteiso-branched monoenes comprise between profiles of mandarin juice sacs (wt. %)

number										
			Even-numbered					Total	Total	
20	22	24	26	28	30	32	34	odd. no.	even no.	Total
0·34	0·44	1·30	1·64	1·13	1·77	0·21		87·08	6·83	93·91
0·60	0·69	1·64	2·15	1·97	1·91	0·19		81·66	9·15	90·81
0·56	0·40	1·12	1·90	1·56	1·89	0·37		80·21	7·80	88·01
1·58	1·67	3·26	3·72	1·68	0·72	0·45		61·96	13·08	75·04
0·47	0·46	2·75	3·94	1·39	0·36	t		65·54	9·37	74·91
0·50	0·51	1·35	1·90	1·55	1·86	0·26		82·98	7·93	90·91
1·03	1·07	3·01	3·83	1·54	0·54	0·23		63·75	11·23	74·98
0·12	t	0·08	0·13	t	0·03	t		2·97	0·36	3·33
0·14	0·22	0·11	0·06	t	t	t		5·04	0·53	5·57
0·16	0·05	0·29	0·18	0·11	0·06	t		6·83	0·85	7·68
t	0·25	0·45	0·53	0·25	t	t		15·63	1·48	17·11
0·18	t	0·28	0·40	0·14	0·05	t		13·22	1·05	14·27
0·14	0·09	0·16	0·12	0·04	0·03	t		4·95	0·58	5·53
0·09	0·13	0·37	0·47	0·20	0·03	t		14·43	1·27	15·69
0·08	0·21	0·40	0·79	0·32	0·12	t		0·84	1·92	2·76
0·14	t	0·69	1·29	0·88	0·34	t		0·28	3·34	3·62
0·10	0·16	0·72	1·64	0·81	0·18	t		0·70	3·61	4·31
0·36	0·45	0·66	3·12	1·28	t	t		1·98	5·87	7·85
0·08	t	1·79	5·26	1·54	0·29	t		1·86	8·96	10·82
0·11	0·12	0·60	1·24	0·67	0·21	t		0·61	2·96	3·56
0·22	0·23	1·23	4·19	1·41	0·15	t		1·92	7·42	9·34

2·8 and 10·8% of the total monoene fraction. As with the *iso*-monoenes, King and Kinnow again show *ca* three times the amount of *anteiso*-branched alkenes as the other three cultivars. The monoene profiles of Mediterranean, Wilking and Honey are quite similar to those of satsuma mandarins. King and Kinnow have the same general monoene profile but show substantially more *anteiso*-C_{24},-C_{25},-C_{26}-C_{27} and C_{-28}.

Long-chain hydrocarbon profiles of *nobilis* and *deliciosa* can be distinguished from non-mandarin citrus species. King (*nobilis*) has hydrocarbon profiles distinct from any other citrus species studied. The hybrid, Kinnow, has a monoene profile similar to its King parent but its saturated hydrocarbon profile is distinct from King. The two hybrids, Honey and Wilking, have similar hydrocarbon profiles, but can be distinguished by their respective *n*-C_{23}/*iso*-C_{23} and *n*-C_{25}/*iso*-C_{25} ratios. The Mediterranean mandarin can be distinguished from its closely related hybrids, Honey and Wilking, by its higher proportion of *n*-C_{29} and *n*-C_{31} monoenes.

The data in Tables 2 and 3 indicate that the relative percentages of mandarin long-chain hydrocarbons generally occur at the same level of either parent or between them. This is strongly supported by the monoene profiles (Table 3) and in particular the percentage of C_{23}, C_{25}, C_{27}, C_{29} and C_{31}. Knowing that hybrids resulting from a cross of two mandarin species show general mandarin hydrocarbon profiles, the question arose as to what effect a cross of a mandarin hybrid with another citrus species would have on these profiles. To this effect, the saturated long-chain hydrocarbon profiles were studied in a hybrid formed by crossing a Temple orange (*C. temple*) with the Kinnow mandarin (*C. nobilis* × *C. deliciosa*). The Temple orange, although given separate species classification by Tanaka [17], is thought to be a natural tangor (mandarin × orange hybrid).

Table 4. Comparative saturated long-chain hydrocarbon data of Temple, Temple × Kinnow and Kinnow cultivars

Relationships	Temple*	Temple × Kinnow*	Kinnow
% n-C_{25}	16·77	18·53	20·99
% Iso-C_{23}	10·37	6·39	3·56
n-C_{23}/n-C_{25}	1·05	0·61	0·67
Iso-C_{23}/iso-C_{25}	0·66	0·37	0·37
n-C_{23}/iso-C_{23}	1·69	1·78	3·94
n-C_{25}/iso-C_{23}	1·06	1·06	2·16
$Anteiso$-C_{24}/$anteiso$-C_{26}	1·14	0·80	0·61

* Values calculated from Ref. [9].

Table 4 is a compilation of information which we believe indicates a hybridization dilution effect of mandarin characteristics in the resulting Temple × Kinnow hybrid. A decrease is observed in *iso*-C_{23} from 10·4% (Temple) to 6·4% (Temple × Kinnow) to 3·6% (Kinnow) while an increase is shown for n-C_{25} from 16·8–18·5–21·0% for these cultivars, respectively. The n-C_{23}/n-C_{25} and *iso*-C_{23}*iso*-C_{25} ratios, previously shown to be important in differentiating citrus species [10], fall within the mandarin range for the Kinnow and Temple × Kinnow hybrid.

Methods for the chemical differentiation of citrus fruit are required to understand more fully how dominant qualities are inbred into hybrids. In citrus breeding, mandarins are crossed extensively with oranges, grapefruit and other mandarins. This has been brought about by the acute need for the development of hybrids which possess good consumer acceptance, good handling and processing qualities and an economically continuous processing season.

EXPERIMENTAL

Isolation and purification of mandarin juice sac lipids. Mature Mediterranean mandarins were obtained from the Budwood Register, Winter Haven, Florida. The Honey, Wilking, Kinnow and King cvs were from Whitmore Foundation Farm (U.S. Horticultural Research Laboratory, U.S.D.A., Orlando, Florida). The fruit were cut in half and the intact juice sacs (vesicles) separated from core, peel, seeds and carpellary membrane with a citrus spoon. The juice sacs were freeze-dried (H_2O less than 4%) and stored at −18° until extracted. Lipids were extracted from 20 g of juice sac powder with $CHCl_3$ and MeOH by a method previously described for total juice sac powder [19]. Quadruplicate extractions run on single batches of fruit from each cv were purified on Sephadex columns [19].

Column and TLC. Columns, 0·9 × 30 cm containing 10 g, 70–325 mesh Si gel were washed with 100 ml $CHCl_3$. The total purified lipid (*ca* 140–200 mg), dissolved in $CHCl_3$, was percolated into the prewashed column and the neutral lipids eluted with 200 ml $CHCl_3$. The fraction was concentrated to a small vol. streaked onto precoated Si gel G plates and developed in hexane–Et_2O (9:1). The hydrocarbon band (containing a portion of carotenoids) was removed from the plate and eluted with Et_2O. The fraction was then restreaked on a $AgNO_3$–Si gel G plate [1] and developed in Et_2O–petrol 2:98. This system separated the saturated and mono-unsaturated hydrocarbons, the carotenoids remaining at the origin. The two hydrocarbon bands were removed and eluted with Et_2O and weighed. The monoene fraction was dissolved in 1 ml hexane and hydrogenated in a Parr apparatus with 10 mg 10% Pd–C catalyst (3 kg/cm²) at 20° for 1 hr.

Quantitation. Hydrocarbons were analyzed by GLC with a FID instrument using a glass column (3·05 m × 4 mm) coated with 3% SP-1000, on 100–120 mesh Gas Chrom Q. Injection port and detector were maintained at 265° and the He flow was 80 ml/min (2·6 kg/cm²). The sample 1–4 μl (representing 1–10% hydrocarbon in hexane) was injected on-column at 160°, temp programmed at 2°/min to 190°, 4°/min to 250° and held at 250° for 5 min. Quantitative measurements were made with a computing integrator and mixtures of reference hydrocarbons were prepared to verify the linear response of the FID and of the computing integrator. MS of citrus *br*-hydrocarbons were previously determined by Hunter and Brogden [20] from this laboratory. Representative hydrocarbon samples from previous studies [11,13,16] were also injected to correlate data obtained from disc integration and by triangulation with the computing integrator used in this study.

Each value shown in Tables 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. Nordby, H. E. and Nagy, S. (1969) *Phytochemistry* **8**, 2027.
2. Nordby, H. E. and Nagy, S. (1971) *Phytochemistry* **10**, 615.
3. Nordby, H. E. and Nagy, S. (1971) *Lipids* **6**, 554.
4. Nagy, S. and Nordby, H. E. (1974) *Phytochemistry* **13**, 153.
5. Nordby, H. E. and Nagy, S. (1974) *Phytochemistry* **13**, 443.
6. Nordby, H. E. and Nagy, S. (1974) *Phytochemistry*, **13**, 2215.
7. Nagy, S. and Nordby, H. E. (1971) *Lipids* **6**, 826.
8. Nordby, H. E. and Nagy, S. (1973) *J. Chromatogr.* **79**, 147.
9. Nagy, S. and Nordby, H. E. (1971) *Phytochemistry* **10**, 2763.
10. Nagy, S. and Nordby, H. E. (1972) *Lipids* **7**, 666.
11. Nagy, S. and Nordby, H. E. (1972) *Phytochemistry* **11**, 2789.
12. Nagy, S. and Nordby, H. E. (1972) *Phytochemistry* **11**, 2865.
13. Nagy, S. and Nordby, H. E. (1972) *Lipids* **7**, 722.
14. Nordby, H. E. and Nagy, S. (1972) *Phytochemistry* **11**, 3249.
15. Nagy, S. and Nordby, H. E. (1973) *Phytochemistry* **12**, 801.
16. Nordby, H. E. and Nagy, S. (1975) *Phytochemistry*, **17**, 183.
17. Hodgson, R. W. (1967) in *The Citrus Industry* (Reuther, W., Webber, H. J. and Batchelor, L. D., eds.) Vol. 1, pp. 496–527. University of California Press, Berkeley, California.
18. Nordby, H. E. and Nagy, S., *J. Am. Soc. Hort. Sci.*, in review.
19. Nagy, S. and Nordby, H. E. (1969) *J. Agric. Food Chem.* **18**, 593.
20. Hunter, G. L. K. and Brogden, W. B. (1966) *Phytochemistry* **5**, 807.