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HYDROCARBONS FROM EPICUTICULAR WAXES OF *CITRUS* PEELS

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Abstract—The hydrocarbon fraction of the epicuticular wax layer of peel from 5 cultivars of *Citrus* was monitored at various stages of fruit development. GLC analysis showed substantial amounts of C_{20}–C_{25} alkanes and alkenes in immature fruit, whereas C_{27}–C_{33} alkanes and alkenes predominated in mature fruit. Mature fruit from 48 other cultivars in 8 *Citrus* taxa were also examined and the ratios of alkanes showed significant differences among the taxa. Monoenes were analyzed for 28 of the cultivars; their profiles were similar to those of the alkanes. The major alkanes were C_{31}, C_{29}, C_{27}, and C_{33} while the major alkenes were C_{30}, C_{28}, and C_{26}. The sequences of taxa in this study corresponded to that formerly determined on the basis of the hydrocarbon profiles of leaf wax.

INTRODUCTION

While investigating the lipid compositions of different *Citrus* juices [1–4], we observed noticeable differences between some lipid components. Further investigations [5, 6] suggested that hydrocarbons might be one of the most promising chemotaxonomic markers. In a preliminary study we found alkanes and alkenes present not only in the juice sacs but also in the leaves, peel, stem and seeds [7]. In leaves [8], peel [8] and juice sacs [9, 10] of *Citrus*, hydrocarbons are present in the epicuticular wax and can be readily obtained free from interfering substances such as essential oils and flavonoids [11]. In extensive studies of alkanes in *Citrus* leaves [2] and of the use of alkane profiles for the differentiation of nucellar from zygotic seedling trees [13], immature and mature leaves were found to differ in their alkane profiles. In a study with 3 orange cultivars, juice sac hydrocarbon profiles were also observed to change with fruit maturation [14]. Recent studies on *Citrus* wax composition [8, 15, 16, 17] have been concerned with the protective function of the wax. The purposes of the present investigation were to determine whether peel hydrocarbon profiles change with fruit maturation and whether they, like the hydrocarbon profiles of juice sac and leaf, are characteristic for each *Citrus* species.

RESULTS AND DISCUSSION

Fruit of various sizes from 1.3 to 4.2 cm mean diameters were collected from 5 citrus cultivars at 3 juvenile periods and at maturity. The composition of alkanes in the epicuticular wax of these samples is presented in Table 1. As the fruit matured the relative amounts of C_{20}–C_{24} declined for all 5 cultivars from a level of 14–31 % for the least mature fruit to a level of 2.1–11.4 % for mature fruit. The relative percentage of C_{20}–C_{24} alkanes in the mature fruit ranged from 11.4 % for Dancy mandarin to 2.1 % for Persian lime. The next higher alkane, C_{25}, declined just as dramatically over these maturation periods with a maximum decrease of 25 percentage points for Valencia and a minimum decrease of 5.7 percentage points for Persian lime. The relative percentages of C_{25}–C_{28} alkanes for the least mature fruit varied considerably among the 5 cultivars, ranging from 28.8 % for Persian lime to 71.9 % for Dancy mandarin. The values for the mature fruit of these two cultivars were 4.0 and 30.2 %, respectively. Over the maturation period, the largest increase in odd-carbon alkane was 47.1 percentage points (for C_{31} in Persian lime); the largest decrease in even-carbon alkane was 5.7 percentage points (for C_{26} in Duncan grapefruit). The least mature Dancy, Pineapple, Valencia and Duncan had quite similar profiles, with C_{25} or C_{27} as the major alkane. Persian lime had C_{31} as its major alkane during this juvenile period. At maturity, however, Dancy mandarin had a profile different from the profiles of the 4 other cultivars. The ratio of the major alkanes-C_{27}, C_{29}, and C_{31} was *ca* 1:2:1, for Dancy and *ca* 1:2:4 for the two orange cultivars and Duncan grapefruit. With Persian limes the relative percentages of only 3 alkanes from C_{20}–C_{33} were greater for the mature fruit than for the least mature. The percentages of these 3 alkanes in mature fruit were: C_{31}, 62.4 %; C_{32}, 5.7 %; and C_{33}, 17.5 %. Such changes from lower to higher carbon numbered alkanes upon maturation were also observed in the alkane profiles of citrus leaves [13] and juice sacs [14]. It appeared that cultivars representing these 4 citrus taxa must have both C_{25} and C_{26} peel alkane percentages less than 6 relative percent in order to be reliable as taxonomic markers.

Table 2 presents the major even-numbered monoenes at the least mature and mature stage of the cultivars. The percentage of monoenes in the total hydrocarbon fraction increased with fruit maturity; e.g., monoenes constituted 10 % of the hydrocarbon fraction from limes to over 19 % of the fractions from Dancy mandarin and Duncan grapefruit. In both the juvenile and the mature

Table 1. Relative percentages of major *n*-alkanes in epicuticular waxes from peels of five *Citrus* cultivars during development

		Percent hydrocarbon									
Cultivar	Mean fruit diam. (cm)	($\Sigma C_{20}C_{24}$)	C_{25}	C_{26}	C_{27}	C_{28}	C_{29}	C_{30}	C_{31}	C_{32}	C_{33}
Dancy mandarin	1.3	14.2	23.0	7.9	37.8	3.2	7.9	1.4	3.4	0.7	0.5
	1.9	22.1	17.5	7.5	35.2	3.3	8.3	1.6	3.3	0.9	0.3
	4.2	18.8	7.6	7.2	33.8	5.7	20.1	1.8	3.8	0.7	0.5
	Mature (6.1)	11.4	3.5	2.7	20.4	3.6	36.5	3.2	15.2	1.5	2.0
Pineapple orange	1.3	25.0	21.1	5.9	24.4	2.7	10.1	1.8	6.0	2.4	0.6
	2.5	19.3	24.3	5.6	23.4	2.2	13.5	1.9	8.1	0.8	0.9
	4.0	5.3	10.2	4.0	24.0	3.2	26.5	4.1	19.3	1.6	1.8
	Mature (6.8)	4.2	3.4	2.0	11.9	2.3	22.6	3.5	39.4	2.6	8.1
Valencia orange	1.3	20.4	27.8	6.4	23.8	2.4	11.4	1.3	4.7	1.2	0.6
	3.0	5.9	13.4	4.4	26.5	2.9	23.7	3.6	16.6	1.4	1.6
	4.1	14.5	7.3	3.8	19.3	3.1	23.1	3.8	20.9	1.8	2.4
	Mature (8.1)	7.3	2.8	1.7	8.3	1.8	17.5	4.0	42.4	4.3	9.9
Duncan grapefruit	1.5	30.8	13.3	7.1	22.6	4.3	9.4	2.4	5.6	3.2	1.3
	2.1	24.7	16.8	6.9	25.7	3.5	11.0	2.4	6.2	1.8	1.0
	4.9	9.0	7.3	4.0	25.7	3.1	21.8	3.5	21.2	2.0	2.4
	Mature (9.1)	4.0	3.6	1.4	10.1	1.8	22.1	3.7	42.8	2.8	7.7
Persian lime	1.3	28.9	6.3	5.4	12.1	5.0	13.3	4.1	15.3	5.3	4.3
	2.2	22.0	4.8	3.9	8.5	3.8	13.6	5.3	26.7	5.8	5.6
	3.9	7.6	3.2	3.3	8.0	5.3	17.5	5.3	34.3	5.3	10.2
	Mature (4.2)	2.1	0.6	0.4	2.4	0.6	5.9	2.4	62.4	5.7	17.5

Table 2. Relative percentages of major even-numbered *n*-alkenes in epicuticular waxes from peels of five *Citrus* cultivars at juvenile and mature stages

			%*					
Cultivar	Maturity Mean fruit diam. (cm)	% Monoene of total hydrocarbon	C_{24}	C_{26}	C_{28}	C_{30}	C_{32}	Σ†
Dancy mandarin	1.3	1.0	12.0	50.0	10.0	—‡	—	72.0
	Mature (6.1)	19.7	3.9	44.3	22.0	2.6	—	72.8
Pineapple orange	1.3	3.7	5.7	33.8	9.3	13.1	—	61.9
	Mature (6.8)	15.6	3.0	17.6	19.8	28.0	10.9	79.3
Valencia orange	1.3	4.4	5.4	31.6	18.9	17.2	—	73.1
	Mature (8.1)	17.0	2.9	22.6	21.2	21.1	6.8	74.6
Duncan grapefruit	1.5	3.9	7.2	57.9	17.5	—	—	82.6
	Mature (9.1)	19.4	6.1	16.8	20.0	25.4	7.5	75.8
Persian lime	1.3	4.2	—	54.0	14.7	31.3	—	100.0
	Mature (4.2)	10.1	1.7	9.9	28.4	28.2	5.8	74.0

*Relative percentage of the $C_{20} \rightarrow C_{33}$ alkenes.
†Sum of the percentages of the 5 major even-numbered alkenes.
‡Less than 0.1 relative percent.

stages *ca* 70% of the linear monoenes consisted of the 5 C_{24}–C_{32} even-numbered alkanes. Of the remaining 30%, C_{20}–C_{25} alkenes were dominant in the juvenile stage and C_{27}–C_{33} were dominant in the mature stage. As observed for alkanes, the relative amounts of lower alkenes (C_{24} and C_{26}) decreased while those of the higher alkenes (C_{28}, C_{30}, C_{32}) increased during maturation. In 3 of the 5 mature cultivars (exceptions being Valencia orange and Persian lime), the two major monoenes were one carbon atom shorter than the two major alkanes within the same cultivar.

Table 3 is a compilation of the mean percentages of peel alkanes in 8 *Citrus* taxa. These taxa include the following 49 cultivars: (1) Mandarins (*C. reticulata*, *C. unshu* and hybrids), Dancy*, Tong Hung Kat, Sugiyama*, Kwano Wase, Kara, Honey*, and Ponkan*. (2) Tangors (*C. sinensis*? × *C. reticulata*?), Ortanique, Temple*, Murcott*, and King*. (3) Tangelos (*C. paradisi* *C. reticulata*), San Jacinto, Orlando*, Minneola*, Seminole*, Sunshine and Sampson. (4) Sweet Oranges (*C. sinensis*), Parson Brown*, Pineapple*, Shamouti*, Jaffa, Lue Gim Gong, Pope Summer and Valencia*. (5) Grapefruit (*C. paradisi*), Hudson, Duncan*, Inman, McCarty*, Ruby Red*, Thompson*, Pink* and March seedless. (6) Lemons (*C. limon*), Lisbon*, Sicilian*, Frost Eureka*, Ross Eureka, Carrigill, Villafranka*, Avant*, Monroe*, Gallagin*, Bearss and Harvey. (7) Citrons (*C. medica*), Indian, Sandfield and Corsican. (8) Limes (*C. aurantifolia*), Key*, Persian*, and Columbian Sweet*. Asterisks denote that mono-unsaturated hydrocarbons (alkenes)

Table 3. Mean relative percentages of C_{25}–C_{33} *n*-alkanes from epicuticular waxes from peels of mature *Citrus*

No.	Taxa	No. cultivars analyzed	C_{25}	C_{26}	C_{27}	C_{28}	C_{29}	C_{30}	C_{31}	C_{32}	C_{33}
1	Mandarins	7	5.3	5.1	26.3	4.7*	34.3	3.1	16.4*	1.5	2.3
2	Tangors	4	6.3	3.5	17.3	2.5	25.4	3.2	34.0	2.0	5.8
3	Tangelos	6	3.6	2.6	10.7	2.6	23.6	4.2	42.3	3.0	7.4
4	Sweet oranges	7	3.5	1.9	11.3	2.0	24.9†	3.5	42.1	2.6	8.2
5	Grapefruit	8	4.8*	2.2	12.0†	2.3†	18.1†	3.9†	43.8*	3.1	9.8†
6	Lemons	11	3.1	1.7	5.3	4.3	35.8†	5.8	35.6†	3.0†	5.4†
7	Citrons	3	1.5	1.3†	3.6†	3.7†	14.2	5.5†	49.4†	7.3*	13.5
8	Limes	3	0.9	0.3	2.3	0.6	9.8	2.3	62.7	4.6	16.5

*Significantly different (analysis of variance) from the value directly below at 5% level.
†Significantly different (analysis of variance) from the value directly below at 1% level.

were also obtained from these cultivars. Categories (1) and (2) were less exclusive than in the leaf study [12] and included only cultivars with mandarin parentage and no known *C. paradisi* parentage. Only first generation tangelos were included in this study; thus, complications involved in back-crosses with a heterozygous hybrid, such as *C. paradisi*, were avoided. A number of cultivar selections from the same species were included in the orange, grapefruit and lemon species categories due to the unavailablility at the time of more cultivars. Columbian Sweet lime was included under limes since its leaf [12], juice [19] and peel alkane profiles were very similar to those for the acid limes, *C. aurantifolia*. Since the percentages of the C_{20}–C_{24} alkanes fluctuated in developing fruit (see Table 1), we selected the C_{25}–C_{33} alkanes as the most reliable for chemosystematic evaluation. Therefore, the percentages of the C_{25}–C_{33} alkanes were normalized so that their sum equalled 100%.

Generally, the 8 taxa were arranged in the sequence presented for the leaf alkane study [12], with some modifications. Satsumas were included with the mandarins, while sour oranges and pummelos were not included in this study. The percentages of the C_{25}–C_{29} alkanes tended to decrease with increasing taxon number; those of the C_{30} alkanes were about the same for all taxa; and those of the C_{31}–C_{33} alkanes generally increased with taxon number.

Singh and Nath [19] suggested the sequential alignment of 31 *Citrus* taxa on the basis of morphological characteristics. Thus, the 31 *Citrus* taxa were positioned between the genus *Poncirus* and the genus *Fortunella*. Nine of 31 *Citrus* taxa were ranked in the following relative order. (1) *C. paradisi*, grapefruit; (2) *C. medica*, citrons; (3) *C. limon*, lemons; (4) *C. sinensis*, sweet oranges; (5) *C. rugulosa*, tangelos; (6) *C. nobilis*, tangors; (7) *C. reticulata*, mandarins; (8) *C. limittoides*, sweet limes, and (9) *C. aurantifolia*, Key lime. Our alignment of the *Citrus* taxa is essentially the reverse of Singh's and Nath's [19] but with 3 exceptions, viz. (i) all limes were included under *C. aurantifolia* and placed after *C. medica* as taxon No. 8, (ii) *C. paradisi* was placed between *C. sinensis* and *C. limon* as taxon no. 5, and (iii) tangelos and tangors were not given species status as Singh and Nath accorded them (*C. noblis* and *C. rugulosa*, respectively) but they were placed sequentially between *C. reticulata* and *C. sinensis*. Our sequence correlates partially with Singh's and Nath's species sequence except for the placing of grapefruit and limes.

The reliability of these relative percentages in showing differences between any two *Citrus* taxa adjacent to each other in the sequence was determined by analysis of variance. In Table 3, any value with one asterisk is significantly different at the 5% confidence level from the one directly below; any value with a dagger is significantly different at the 1% level from the one directly below. Relative percentages of C_{28} and C_{31} in mandarins were significantly different from those of tangors at the 5% level. Two relatively similar taxa, oranges and grapefruit [12], were significantly different in their C_{29} values at the 1% level. Grapefruit were different from lemons, lemons from citrons and citrons from limes in the percentages of 7, 4 and 6 alkanes respectively.

Full comprehension of the differences between pairs of alkane profiles required examination of the ratios between the various alkanes. Eight of the 36 possible C_{25}–C_{33} alkane ratios showing the greatest differences among the profiles of these eight *Citrus* taxa are presented in Table 4. The ratios are for mature fruit. Changes

Table 4. Mean ratio of *n*-alkanes in epicuticular waxes from peel of mature *Citrus*

	Hydrocarbon ratios							
Taxa	C_{30}/C_{26}	C_{31}/C_{26}	C_{32}/C_{26}	C_{33}/C_{26}	C_{31}/C_{27}	C_{33}/C_{27}	C_{32}/C_{28}	C_{31}/C_{29}
Mandarins	0.7	4.1	0.4	0.8	0.7	0.2	0.3*	0.6
Tangors	1.0	12.1	0.6*	1.9	3.1	0.5	0.8	1.5*
Tangelos	1.6	27.4	1.2	4.1	4.0	0.7	1.2	1.8
Sweet oranges	1.8	25.0	1.4	4.9	4.0	0.7	1.3	1.8†
Grapefruit	1.7*	20.4	1.3	4.5	3.6	0.8	1.4	2.4†
Lemons	3.4	25.8	1.8†	3.8†	6.7	1.0†	0.7†	1.0†
Citrons	4.2	38.9	5.6*	10.7*	13.7†	3.8†	2.0†	3.5
Limes	7.7	239.4	15.3	59.8	27.3	7.2	7.7	6.4

*Significantly different from the value directly below at 5% level.
†Significantly different from the value directly below at 1% level.

in the ratios of the higher alkanes to the lower alkanes during the maturation period emphasized the increase in the relative amounts of the longer alkanes with increasing fruit maturity (cf. Table 1). Each of the 8 ratios increased in magnitude as the fruit matured. Thus, only the ratios for mature fruit were reliable in *Citrus* taxa differentiation. All the ratios tended to increase with increase in taxon number. Mandarins were significantly different from tangors (5%), in their C_{32}/C_{28} ratios. The C_{31}/C_{29} ratio clearly differentiated sweet oranges from grapefruit, grapefruit from lemons, and lemons from citrons. These 8 select ratios showed that the alkane profiles of lemons, citrons and limes were very different from those of the other taxa.

Alkenes were detected in the epicuticular wax from the peels of some *Citrus*; the maximum level was 19.7% (Dancy mandarin) of the total hydrocarbon fraction. We examined the alkene content for 28 of 48 cultivars. Because most were *n*-alkenes (less than 3% were branched), only C_{20}–C_{33} homologues were tabulated.

In comparing GLC R_ts of alkenes from peels and from juice sacs, we noted that those from peel were consistently longer. After hydrogenation, both homologous series had the same R_t. The different R_ts of peel alkenes might be due to isomeric or positional differences of the double bond in the chain but these postulations were not confirmed. Table 5 gives the mean C_{20}–C_{33} alkene values for 28 of the 49 cultivars examined. The ratio of even to odd numbered alkenes was *ca* 4 : 1. This was in contrast to that observed for citrus juice sacs where odd-numbered alkenes predominated [5, 6]. Three alkenes, C_{26}, C_{28}, and C_{30} accounted for 84% of the even-numbered monoenes. The ratio of these 3 alkenes, however, was different for each of the 7 taxa. The relative percentage of C_{26} varied from 44% for mandarins to 8–9% for lemons–limes. The relative percentages of C_{23}, C_{24}, C_{25} and C_{27} tended to decrease while those of C_{28}–C_{33} tended to increase with increase in taxon number. The chain lengths of the 3 major alkenes were one C atom shorter than those of the major alkanes. As a percentage, however, they did not relate 1 to 1, e.g. for limes—32% C_{30} alkene, 63% C_{31} alkane.

Analysis of variance for these alkenes showed that the greatest differences were between grapefruit–lemons and between lemons–limes. Less significant (5% level) differences were observed between tangelos–sweet oranges and sweet oranges–grapefruit. The number of significant differences between taxa distinguishable by alkene values was less than that distinguishable by alkane values.

We compared the alkane profiles for leaf [12] and peel waxes. Both of these epicuticular waxes contain predominantly C_{20}–C_{33} *n*-alkanes. Five alkanes, C_{29}–C_{33}, accounted for over 90% of alkanes in leaf whereas 9 alkanes C_{25}–C_{33} accounted for 90% in peel. Longer chain alkanes predominated in the waxes of both mature leaf and peel. For the same sequence of *Citrus* taxa the relative percentages of alkanes in both waxes tended to follow the same trend. For example, C_{29} content decreased and C_{33} content increased with increasing taxon number.

Peel alkane profiles were shown to be potentially important as taxonomic markers. Although application of these profiles for determination of hybrid parentage was not shown, the similarity of the profiles to those present in leaf wax suggests that peel alkanes could be as definitive in chemosystematics as leaf alkanes. For this objective, however, mature fruit is necessary; immature fruit showed a higher proportion of lower alkanes than mature fruit. Alkenes, although present in the peel at a maximum of 19.7% of total hydrocarbons, had profiles characteristic of the different *Citrus* taxa. Both alkanes and alkenes showed similar trends in hydrocarbon percentages for the same sequence of *Citrus* taxa.

EXPERIMENTAL

Peel samples. For the maturation study, *ca* 10 immature fruit (1.3–4.2 cm mean diam.) were harvested on the 12th of March, April, and May, from each of 5 citrus cultivars. Also 3 fruits were picked at harvest maturity from each of the cultivars. Mature fruit for the taxonomic study were obtained from Budwood Register, Winter Haven, Florida, and from Whitmore Foundation Farm (U.S. Horticultural Research Laboratory, USDA, Orlando, Florida). Fruit in both studies were dipped for 3 min in 200 ml $CHCl_3$, and the hydrocarbons isolated from the wax extract as described previously [7, 12].

GLC. Both alkanes and alkenes were analyzed in duplicate on a 3.05 m, 4 mm i.d. glass column coated with 3% SP-1000 on Gas Chrom Q [20]. Quantitative measurements were made with a computing integrator. The coefficient of variation (CV) for two mean ranges (MR) are MR 1.0–5.0; CV 3–5%, MR above 5.0; CV less than 2%.

Table 5. Mean relative percentages of *n*-alkenes in epicuticular waxes from *Citrus* peel

No.	Taxa	No. of cultivars	Hydrocarbon C_{20}	C_{21}	C_{22}	C_{23}	C_{24}	C_{25}	C_{26}	C_{27}	C_{28}	C_{29}	C_{30}	C_{31}	C_{32}	C_{33}
1	Mandarins	4	0.1	0.5	1.8	2.0	5.7	5.9	44.0	10.6	15.5	3.9	7.7	2.0	0.3	tr†
2	Tangors	3	tr	0.3	0.8	0.8	7.7	5.1	35.8	6.8	15.2	4.0	15.9	2.2	4.9	0.4
3	Tangelos	3	0.2	0.4	2.1	2.4*	6.9	3.9	19.5	6.2	22.5	3.9	22.1	2.7*	6.8*	0.4
4	Sweet oranges	4	0.1	0.1	0.9*	0.7	4.3	2.4	19.9	4.6	19.0	6.2	23.2	6.7	10.0	1.8†
5	Grapefruit	4	0.3	0.3	1.6	1.6	7.3‡	3.2‡	17.8‡	5.7	21.1‡	7.0	23.1	4.5	6.0	0.5
6	Lemons	7	0.2	1.0	1.3	1.1	3.1	1.8	8.3	6.9‡	37.7*	9.5	20.9‡	4.6	3.4‡	0.2
7	Limes	3	tr	tr	1.7	0.9	3.4	1.1	9.0	4.2	27.5	7.0	32.2	3.8	8.8	0.4

*Significant difference from next level taxa at 5% level.
†Less than 0.1%.
‡Significant difference from next level taxa at 1% level.

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