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THE ORIGIN OF CITRUS FLAVOR COMPONENTS—III.

A STUDY OF THE PERCENTAGE VARIATIONS IN PEEL AND LEAF OIL TERPENES DURING ONE SEASON*

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Abstract—Peel and leaf oils of "Dancy" tangerine, "Hamlin" orange, and "Marsh" grapefruit were analyzed by gas–liquid chromatography at biweekly intervals from spring until harvest. Concentrations of fourteen to sixteen main components in each oil were approximated from the peak heights on the chromatograms. The greatest changes were found in the peel oils of the orange and the tangerine where oxygenated constituents, particularly linalool, decreased markedly in relative concentration as the fruit matured, while the (+)-limonene concentration exhibited a corresponding increase. A relationship between (+)-limonene concentration and linalool concentration was shown graphically. A possible mechanism for linalool biosynthesis was suggested.

INTRODUCTION

THE first two papers in this series reported the results of chemical analyses of volatile oils from eleven cultivars of citrus leaves,[1] and petals, pistils, and stamens of three cultivars of citrus blossoms.[2] These papers increased our knowledge of the chemistry of volatile oils from active, growing tissues of the tree. The present paper extends this work to the changes in composition of the volatile oils from leaves between the first flush of growth in March and the time of fruit harvest in the winter, and from peel between fruit set in April and winter harvest. The same three cultivars studied previously, "Dancy" tangerine (*Citrus reticulata* Blanco), "Hamlin" orange (*C. sinensis* (Linn.) Osbeck), and "Marsh" grapefruit (*C. paradisi* MacF.),[2] were used.

RESULTS

In each oil the percentage compositions of the fourteen to sixteen major components were approximated from their peak heights on the gas chromatograms. Values were given to two significant figures where feasible. Although collections were made twice monthly, the results were tabulated at only monthly intervals except in cases where large monthly changes took place. In the latter instances the figures for the intervening 2 weeks were inserted to give a more accurate picture.

The data for "Dancy" tangerine, "Hamlin" orange, and "Marsh" grapefruit peel oils, respectively, are shown in Tables 1–3. A graphical illustration of the relationship between (+)-limonene and linalool in tangerine peel oil is shown in Fig. 1.

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[1] J. A. ATTAWAY, A. P. PIERINGER and L. J. BARABAS, *Phytochem.* 5, 141 (1966).
[2] J. A. ATTAWAY, A. P. PIERINGER and L. J. BARABAS, *Phytochem.* 5, 1273 (1966).

TABLE 1. PERCENTAGE COMPOSITION OF TANGERINE PEEL OIL FROM APRIL TO JANUARY

Compound	April 26	May 24	June 7	June 21	July 19	Aug. 2	Aug. 17	Sept. 13	Oct. 11	Nov. 8	Dec. 6	Jan. 3
α-Pinene + α-thujene	0·72	0·48	0·62	0·70	0·57	0·72	0·95	0·90	0·60	1·0	0·36	0·54
β-Pinene + sabinene	0·76	0·50	0·56	0·47	0·40	0·47	0·44	0·42	0·20	0·48	0·30	0·39
Myrcene	0·46	0·53	0·72	0·89	0·82	1·4	1·5	1·5	1·4	1·6	1·0	1·2
α-Terpinene	0·18	0·17	0·16	0·12	0·10	0·14	0·10	0·05	0·06	0·04	0·03	0·08
(+)-Limonene	20	31	51	64	65	79	84	87	85	86	85	87
Ocimene	1·4	0·46	0·56	0·51	0·18	0·23	0·16	0·10	0·09	0·20	0·06	0·08
γ-Terpinene + octanal	7·8	4·9	6·2	6·8	4·9	4·2	5·5	3·9	4·0	4·9	3·7	3·9
Terpinolene	0·44	0·36	0·39	0·39	0·29	0·29	0·24	0·17	0·23	0·44	0·27	0·31
Linalool	62	56	36	23	25	11	6·0	5·0	6·3	4·1	6·6	4·2
Terpinen-4-ol + thymyl methyl ether	0·86	0·95	0·66	0·58	0·47	0·36	0·28	0·17	0·34	0·44	0·39	0·27
α-Terpineol	1·0	1·7	1·2	1·0	0·86	0·75	0·52	0·42	1·1	0·32	1·2	0·89
Geranial	0·12	0·17	0·16	0·19	0·14	0·10	0·12	0·10	0·23	0·04	0·33	0·31
Geraniol	0·20	0·19	0·16	0·08	0·07	0·07	0·04	0·07	0·09	0·04	0·03	0·04
Thymol	3·4	2·1	1·3	0·55	0·43	0·16	0·12	0·10	0·23	0·20	0·30	0·23

TABLE 2. PERCENTAGE COMPOSITION OF ORANGE PEEL OIL FROM MAY TO DECEMBER

Compound	May 10	May 24	June 7	July 6	Aug. 5	Aug. 30	Sept. 27	Oct. 26	Nov. 22	Dec. 20
α-Pinene	0·14	0·51	0·43	0·37	0·60	0·68	0·62	0·69	0·45	0·48
Sabinene	0·73	1·75	1·30	0·93	0·86	0·64	0·53	0·56	0·59	0·64
Myrcene	0·69	1·58	1·59	1·26	1·55	1·46	1·50	1·76	1·60	1·41
α-Terpinene	0·14	0·23	0·15	0·10	0·04	—	0·04	0·04	0·03	—
(+)-Limonene	52	82	87	90	94	95	95	95	94	95
γ-Terpinene	0·74	1·36	0·62	0·51	—	—	—	—	—	—
Octanal	0·62	1·3	0·82	0·79	0·79	0·79	0·62	0·73	1·57	0·80
Terpinolene	0·21	0·10	0·19	0·10	—	0·04	0·09	0·09	—	—
Citronellal	0·55	0·17	0·10	0·10	—	—	—	—	—	—
Linalool	27	7·0	5·3	3·8	0·90	0·72	0·75	0·60	1·1	1·1
Terpinen-4-ol	5·5	1·2	0·58	0·37	0·14	0·07	0·04	0·04	0·07	0·06
α-Terpineol	4·3	1·1	0·77	0·70	0·26	0·15	0·09	0·13	0·21	0·19
Geranial	3·5	1·0	0·53	0·56	0·21	0·15	0·09	0·13	0·24	0·23
Geraniol	0·52	0·06	0·05	0·05	—	—	0·04	—	—	—

TABLE 3. PERCENTAGE COMPOSITION OF GRAPEFRUIT PEEL OIL FROM APRIL TO JANUARY

Compound	April 26	May 10	May 24	June 7	July 6	Aug. 3	Aug. 31	Sept. 28	Oct. 25	Nov. 22	Jan. 3
α-Pinene	0·29	0·25	0·39	0·37	0·40	0·61	0·52	0·56	0·68	0·57	0·64
Sabinene	1·5	1·4	1·1	0·93	0·71	0·65	0·88	0·91	0·49	0·41	0·68
Myrcene	1·2	1·1	1·3	1·3	1·5	1·5	1·4	1·6	1·6	1·5	1·9
α-Terpinene	0·06	0·09	0·07	0·06	0·03	0·03	0·03	0·05	0·06	—	—
(+)-Limonene	89	83	88	86	93	93	92	92	93	93	93
Ocimene	2·7	1·8	0·82	0·70	0·55	0·48	0·48	0·43	0·49	0·41	0·36
Octanal	0·70	0·92	0·95	1·4	0·77	0·87	1·3	0·82	1·5	0·95	0·56
t-Linalool oxide	0·81	1·8	0·59	0·56	0·03	0·13	0·12	0·09	0·06	0·03	0·03
c-Linalool oxide	0·35	0·78	0·29	0·25	0·03	0·09	0·08	0·05	—	0·03	0·03
Linalool	1·6	3·1	1·5	2·2	0·74	0·53	0·52	0·47	0·49	0·41	0·36
Octanol	—	—	0·33	0·42	0·40	0·48	0·56	0·56	0·68	0·54	0·76
Terpinen-4-ol	0·70	2·4	0·39	2·2	0·18	0·09	0·12	0·05	—	0·08	0·08
α-Terpineol	0·46	1·7	0·75	1·1	0·37	0·26	0·24	0·17	0·18	0·14	0·16
Geranial	0·41	1·1	0·42	0·75	0·21	0·13	0·16	0·12	0·12	0·14	0·20

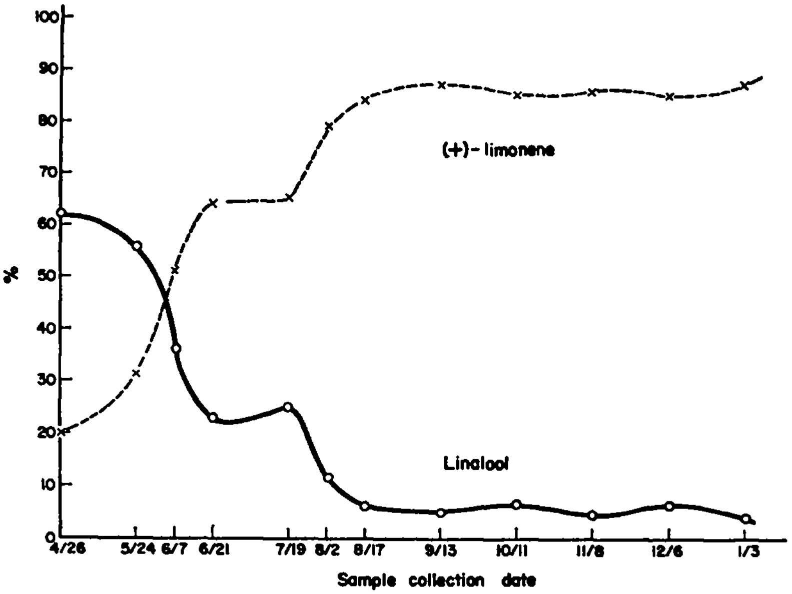

FIG. 1. RELATIVE PERCENTAGES OF (+)-LIMONENE AND LINALOOL IN TANGERINE PEEL OIL DURING 1965–66 SEASON.

The data for the tangerine, the orange, and the grapefruit leaf oils, respectively, are shown in Tables 4–6. The variations during the year in the concentrations of linalool, thymyl methyl ether, and total terpenes in tangerine leaf oil are shown graphically in Fig. 2.

TABLE 4. PERCENTAGE COMPOSITION OF TANGERINE LEAF OIL FROM MARCH THROUGH JANUARY

Compound	Mar. 25	April 21	May 24	June 21	July 19	Aug. 17	Sept. 13	Oct. 11	Nov. 8	Dec. 7	Jan. 3
α-Pinene + α-thujene	2·2	1·1	1·4	2·7	2·5	2·6	2·4	1·6	1·3	1·4	2·5
β-Pinene	2·4	1·4	1·6	2·6	2·4	2·3	2·5	1·8	1·6	1·7	2·5
Myrcene	0·6	0·3	0·4	0·6	0·6	0·6	0·6	0·4	0·4	0·4	0·6
α-Terpinene	0·3	0·1	0·2	0·2	0·2	0·2	0·1	0·2	0·1	0·1	0·2
(+)-Limonene	1·6	0·8	1·3	1·8	1·8	1·9	1·8	1·5	1·3	1·4	1·8
β-phellandrene	0·3	0·2	0·3	0·4	0·4	0·4	0·5	0·4	0·3	0·4	0·5
Ocimene	8·2	2·8	4·5	6·0	5·9	7·2	5·3	5·6	4·6	5·3	6·9
γ-Terpinene	10	4·3	6·9	8·8	8·6	9·5	8·4	7·8	6·1	6·9	9·2
Terpinolene	1·1	0·7	0·9	1·3	1·2	1·3	1·1	1·1	0·8	0·9	1·2
p-Isopropenyltoluene	0·6	0·9	0·9	1·0	1·0	0·9	1·5	1·1	1·0	1·0	0·9
Linalool	66	78	70	57	62	57	66	62	73	68	52
Thymyl methyl ether	1·1	3·5	4·9	5·8	7·5	10	6·2	11	6·0	7·1	16
α-Terpineol + neral	0·4	0·6	0·6	0·4	0·4	0·2	0·4	0·4	0·4	0·4	0·4
Geraniol	0·1	0·1	0·1	0·2	0·2	0·2	0·1	0·2	0·1	0·1	0·2
Thymol	3·8	4·6	5·6	6·9	4·5	4·7	2·6	4·6	2·7	4·2	4·0
TOTAL TERPENES	27	13	18	25	25	27	24	22	18	20	26

TABLE 5. PERCENTAGE COMPOSITION OF ORANGE LEAF OIL FROM MARCH THROUGH JANUARY

Compound	Mar. 12	April 12	May 10	June 7	July 7	Aug. 2	Aug. 30	Sept. 27	Oct. 25	Nov. 22	Dec. 20	Jan. 3
α-Pinene+α-thujene	1·8	1·7	1·7	2·2	2·1	1·6	2·3	2·1	2·6	2·0	2·0	2·2
Sabinene	56	53	53	52	55	53	55	57	53	58	53	55
Myrcene	3·9	4·0	3·9	4·9	4·8	5·3	6·1	5·1	5·7	4·6	5·1	4·2
Car-3-ene	2·0	5·2	5·3	6·3	5·8	5·8	7·3	6·8	7·6	6·2	8·0	7·6
α-Terpinene	1·0	1·0	0·9	1·1	1·0	0·9	1·1	0·8	1·5	0·7	0·9	0·9
(+)-Limonene	2·3	3·0	3·3	3·4	3·8	3·7	4·7	4·5	5·7	5·6	5·5	6·4
β-Phellandrene	1·1	1·5	1·8	1·5	1·4	1·4	1·7	1·4	1·8	1·4	1·7	1·4
Ocimene	6·2	6·8	6·4	9·3	9·1	9·1	8·4	9·3	8·7	8·1	9·2	8·3
γ-Terpinene	1·3	2·2	1·9	2·4	1·1	1·5	1·1	1·6	1·6	1·5	1·9	1·3
Terpinolene	0·9	1·5	1·7	2·0	1·7	1·9	1·9	1·8	2·1	1·6	2·1	2·1
Citronellal	0·2	1·3	2·3	1·9	2·4	3·6	1·4	2·0	1·0	2·3	2·4	2·0
Linalool	16	11	10	4·4	3·5	4·2	2·7	1·5	1·6	2·0	2·5	3·7
Terpinen-4-ol	5·0	3·5	2·5	3·9	3·4	4·0	1·9	1·9	3·8	1·4	1·5	1·9
α-Terpineol+neral	0·9	0·7	0·6	1·1	0·8	0·7	0·5	0·7	0·6	0·6	0·7	0·5
Geranial	0·5	2·0	3·0	1·5	1·8	2·5	1·0	1·3	0·9	1·5	1·4	1·3
TOTAL TERPENES	76	80	80	85	85	84	90	90	90	90	90	89

TABLE 6. PERCENTAGE COMPOSITION OF GRAPEFRUIT LEAF OIL FROM MARCH TO JANUARY

Compound	Mar. 16	April 12	May 10	June 8	July 6	Aug. 2	Aug. 30	Sept. 27	Oct. 26	Nov. 11	Dec. 6	Jan. 3
α-Pinene	1·8	1·6	1·7	2·0	2·7	2·2	1·9	2·2	2·7	2·5	2·1	1·9
Sabinene	44	49	44	53	57	58	48	57	51	42	43	59
Myrcene	2·8	3·1	2·7	4·2	4·0	3·7	3·2	3·6	3·9	4·1	3·1	4·3
Car-3-ene	0·2	0·2	0·1	0·1	0·1	0·1	0·1	0·1	0·1	0·2	0·1	0·2
α-Terpinene	1·8	1·0	1·1	0·9	1·2	1·0	0·8	1·0	1·3	1·1	0·7	0·7
(+)-Limonene	1·8	2·0	1·6	2·4	2·7	2·2	2·9	2·5	3·3	2·9	2·8	2·9
β-Phellandrene	1·1	1·2	2·0	1·8	1·6	1·3	1·0	1·2	1·3	1·8	0·7	1·1
Ocimene	8·1	9·5	8·1	12	11	11	11	13	11	13	12	11
γ-Terpinene	3·8	2·5	2·6	2·3	2·4	2·2	1·8	2·4	2·7	1·7	2·3	1·2
Terpinolene	0·8	0·6	0·6	0·5	0·6	0·5	0·4	0·6	0·6	0·7	0·4	0·4
Citronellal	0·1	1·2	1·6	3·3	3·2	6·4	6·2	3·5	4·4	11	12	5·1
Linalool	23	24	18	14	9·1	5·9	15	7·3	9·8	11	13	7·1
Terpinen-4-ol	8·2	1·4	14	2·0	2·8	1·8	2·2	3·9	4·3	2·3	3·3	2·2
α-Terpineol+neral	1·2	0·6	0·9	0·6	0·4	0·5	1·2	0·8	1·0	1·2	1·0	0·8
Geranial	0·1	0·7	0·8	0·7	0·6	0·6	3·3	0·7	1·4	1·5	2·0	1·0
TOTAL TERPENES	66	70	63	79	83	82	71	84	78	70	66	82

DISCUSSION

The peel oils of all three cultivars showed a large decrease in the percentage of oxygenated components, particularly linalool, during the course of the season. This was accompanied by a corresponding increase in the percentage of terpene hydrocarbons, particularly (+)-limonene. The change was most noticeable in tangerine peel oil where linalool decreased from 62 per cent on April 26 to 4 per cent on January 3, while (+)-limonene increased from 21 to 87 per cent during this same time interval. In the "Hamlin" orange peel oil, linalool decreased from 27 to 1·1 per cent as limonene increased from 52 to 95 per cent, while in grapefruit peel

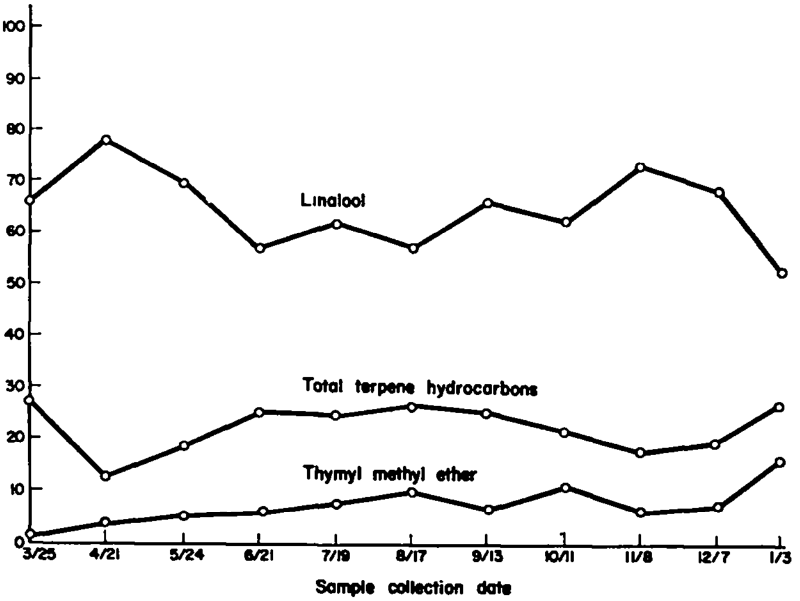

FIG. 2. RELATIVE PERCENTAGES OF LINALOOL, TOTAL TERPENE HYDROCARBONS, AND THYMYL METHYL ETHER IN TANGERINE LEAF OILS DURING 1965–66 SEASON.

oil the percentages of linalool and linalool oxides decreased from a total of 5·7 per cent on May 10 to 0·4 per cent on January 3, as limonene increased from 89 to 93 per cent. The inverse relationship between the linalool and limonene percentages in tangerine peel oil is well illustrated in Fig. 1. The limonene curve is seen to increase as the linalool curve decreases from April 26 to June 21 where a valley in the linalool curve coincides with a plateau in the limonene curve. Slight maxima for limonene on September 13, November 8, and January 3 coincide with equally slight minima for linalool on those dates. Trends were not so evident among the other terpenes. The percentages of total terpenes other than limonene held constant or decreased as the limonene percentage increased.

Consistent trends were not so evident in the leaf oils. Most compounds showed peaks and valleys in their percentage curves, but were at about the same percentage in January as they had been in March. Generally, fluctuations in the concentration of linalool were accompanied by opposite fluctuations in the concentration of total terpenes. This is illustrated for tangerine leaf oil in Fig. 2, wherein the linalool curve varied from 66 per cent to a maximum of 78 per cent and then decreased to a minimum of 52 per cent while total terpenes were 27, 13 and 26 per cent on the corresponding dates. This again indicates a possible relationship between linalool and the total terpenes. It is also evident from Fig. 2 that the percentage of thymyl methyl ether increased gradually during the course of the season and that after August 17 a connection existed between the peaks and valleys of its curve and those of the linalool curve.

In "Hamlin" orange leaf oil linalool decreased from 16 to 3·7 per cent for a net change of $-12·3$ per cent while the total terpenes increased from 76 to 89 per cent for a net change of $+13$ per cent. The individual compounds contributing the most to this gain were car-3-ene and (+)-limonene, while sabinene, the major component in the oil, held constant.

In grapefruit leaf oil the total oxygenated components decreased in concentration throughout the year as the total terpene hydrocarbons increased, with the exception of citronellal which increased from 0·1 per cent to a maximum in December of 12 per cent. Linalool oxide occurs in all grapefruit leaf oils and is no longer thought to be an artifact as suggested earlier.[1]

THEORETICAL CONSIDERATIONS

In the first paper of this series[1] a possible mechanism was presented for the formation of citrus leaf oil terpenes from a common precursor, linalool. It was based on the known chemistry of linalool and its role in citrus essential oils. Actually, the hypothesis that linalool might be the parent compound of the plant terpenes was not new, as it had been suggested as early as 1937 by Ganaparti[3] and more recently by Fujita[4] who made an exhaustive study of some thirty-nine compounds and 412 plant species before drawing this conclusion.

The results of this paper provide additional indirect evidence to support this hypothesis as both the tabular data and the graphs suggest a direct connection between the linalool concentration and the concentrations of related terpenes. However, in recent years the study

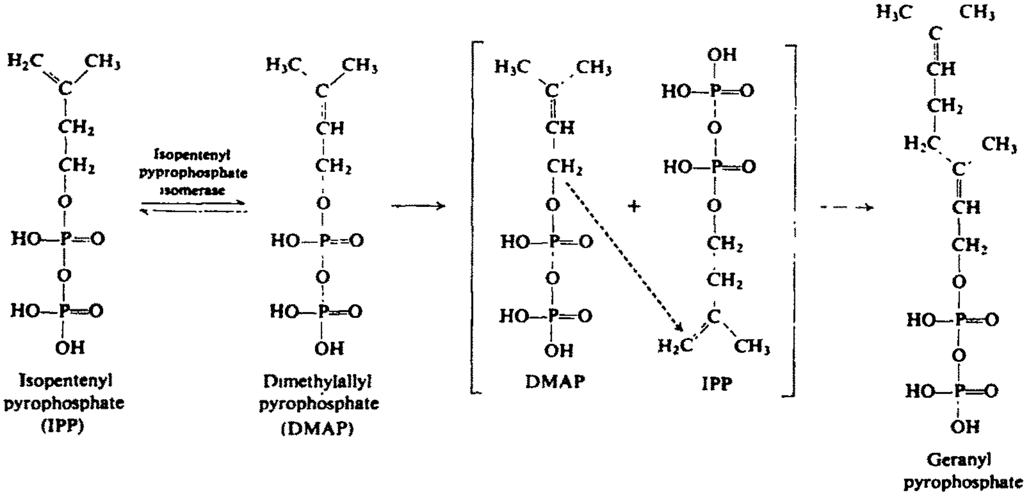

FIG. 3. THE PATHWAY FROM ISOPENTENYL PYROPHOSPHATE TO GERANYL PYROPHOSPHATE.

of the biosynthesis of terpenes has progressed from only indirect evidence to the firm establishment of one pathway for the formation of terpenes from acetate. This is the route through mevalonic acid, isopentenyl pyrophasphate, and dimethylallylpyrophosphate to geranyl and farnesyl pyrophosphates.[5] Consequently, evidence for linalool as the precursor for terpenes in higher plants must be re-evaluated in the light of these findings.

The key to the "acetate to squalene" path is the formation of isopentenyl pyrophosphate, the "active isoprene unit", from mevalonic acid followed by its isomerization and subsequent condensation with dimethylallylpyrophosphate to give geranyl pyrophosphate as shown in Fig. 3. This reaction involves alkylation of the carbon–carbon double bond by the highly reactive allyl group to give geranyl pyrophosphate. The latter, also being allylic, can react with another mole of isopentenyl pyrophosphate to give farnesyl pyrophosphate. Two

[3] K. Ganaparti, *Current Sci., India* 6, 19–20 (1937); *Chem. Abstr.* 31, 7939 (1937).
[4] Y. Fujita, *Kagaku (Kyoto)* 11, 874 (1956)—quoted in *Biochemistry of Natural Compounds*, Pergamon Press, The MacMillan Co., N.Y., pp. 651–653 (1963).
[5] H. J. Nicholas, *Biogenesis of Terpenes in Plants*, Chap. 14 of *Biogenesis of Natural Compounds* (see Ref. 4).

enzymes involved in these transformations were isopentenyl pyrophosphate isomerase,[6] found in yeast and rat tissue, which converted isopentenyl pyrophosphate to dimethylallylpyrophosphate, and farnesyl pyrophosphate synthetase,[7] from yeast extracts, which converted dimethylallylpyrophosphate to geranyl and farnesyl pyrophosphates.

The above reactions are very important steps in steroid biosynthesis in animal tissue, and may also constitute the major pathway to the terpenes in higher plants. If so, then geraniol, or geranyl pyrophosphate, is the parent compound of the monoterpenes and the large accumulation of linalool in citrus oils is due to factors independent of the pathway from mevalonate to the C_{10} terpene skeleton. However, there is also the possibility that other pathways from isopentenyl pyrophosphate to monoterpenes exist in higher plant tissue. For the purpose of our own hypothesis, which regards linalool as the parent compound, let us examine mechanistically what happens if the reaction of isopentenyl pyrophosphate with dimethylallylpyrophosphate takes place in a different manner. Instead of alkylation and loss of pyrophosphate as shown in Fig. 3, assume that dimethylallylpyrophosphate adds across the double bond followed by concerted elimination of pyrophosphate from the isopentenyl group as shown in Fig. 4. This would give linalyl pyrophosphate as the product.

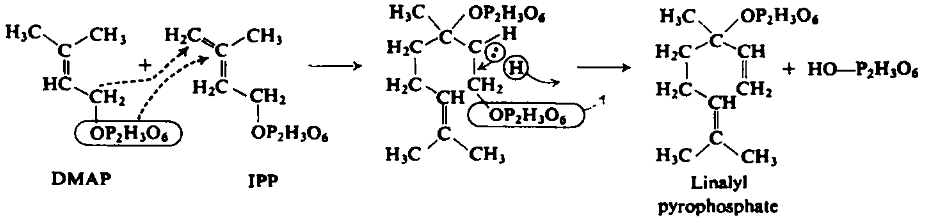

FIG. 4. A POSSIBLE ROUTE TO LINALYL PYROPHOSPHATE.

The existence of this or a similar reaction, catalyzed by an enzyme which might be called linalyl pyrophosphate synthetase, cannot be totally excluded at this time.

EXPERIMENTAL

Preparation of Samples

Fruit and leaves were collected every other week during the 1965–66 season from mature trees grown in commercial groves. Amounts of young fruit processed depended on the yield of oil from the particular species, "Dancy" tangerine giving a much better yield than "Hamlin" orange or "Marsh" grapefruit. For example, on May 10, 900 g of tangerines varying in diameter from 12–18 mm yielded 1·5 ml of oil, while on this same date 1400 g of oranges 32–50 mm in diameter gave only 0·35 ml, and 1800 g of grapefruit 38–50 mm in diameter gave 0·60 ml. The first samples were obtained by steam distillation of the whole fruit, but by June 7 the sample size was sufficiently large to permit removal of the peel for steam distillation. An attempt was made to recover oil from the pulp remaining after the peel had been removed, but yields were too small for analytical purposes. The steam distillation procedure was described earlier.[2]

Leaf collections began with the development of the spring flush. All collections throughout the year, in so far as possible, were made from this same flush. Tangerine leaves gave the

[6] B. W. AGRANOFF, H. EGGERER, U. HENNING and F. LYNEN, *J. Biol. Chem.* **235**, 326 (1960).
[7] F. LYNEN, B. W. AGRANOFF, H. EGGERER, U. HENNING and E. M. MOSLEIN, *Angew. Chem.* **71**, 657 (1959).

best yields of oil, as was cited earlier by Pieringer *et al.*[8] In a typical run 500 g of tangerine leaves yielded 0·75 ml of oil, while it required 1000 g of orange leaves and over 2000 g of grapefruit leaves to produce this quantity.

Oil Analysis by Gas–Liquid Chromatography

Gas chromatograms were prepared using the F & M Model 810 gas chromatograph fitted with a 50 ft $\times \frac{1}{8}$ in. column packed with Carbowax 20M on 60–80 mesh Gas-Chrom Z as described in the previous paper.[2] After injecting 0·15 μl samples, the column was held isothermal at 93° for 16 min and then programmed to 210° at the rate of 4° per min. It was held isothermal at 210° for an additional 20 min to elute thymol from the tangerine peel and leaf oils. An attenuation of $10^2 \times 4$ produced good peaks from the quantitatively minor components while attenuations to $10^2 \times 128$ were necessary for some of the large peaks such as (+)-limonene in the peel oils and sabinene and linalool in the leaf oils. The height of each peak was determined in recorder scale units from 0 to 100 and multiplied by an appropriate factor where necessary to correct for attenuations greater than $10^2 \times 4$. Peak heights were added to get a total value suitable for determining the percentages of each compound being measured.

[8] A. P. PIERINGER, G. J. EDWARDS and R. W. WOLFORD, *Proc. Am. Soc. Hort. Sci.* **84**, 204 (1964).