The direct effect of the free fatty acids themselves on flavor and aroma would depend on the kind of acids liberated. The metabolic liberation of fatty acids to supply energy in the depleted live pig may produce a pattern of acids different from that produced by autolysis in post-mortem fat. Relation of fat acidity to depletion was evident from the following observation. In the 14 back fat samples having acid numbers above 0.40, the liver glycogen values in milligrams per gram were distributed as follows: 7 at 2 or lower; 2 at 4 to 8; 3 at 12; and 2 at 25. In the 16 samples having acid numbers below 0.25, the liver glycogen values were: 13 at 41 to 97; 1 at 28; and 2 at 12 to 14.

A scattergram for fat flavor and acid numbers after a 24-week frozen storage (Figure 2) gave a strikingly different pattern from that shown in Figure 1. At that time, traces of peroxide had appeared in only four samples, but the flavor had deteriorated so far that 81% of the points were located in the region comparable to area *D* (Figure 1). Free fatty acid had increased 50 to 100% in most samples. Many acid numbers were still below the high values found in fresh samples, but corresponding flavor scores had dropped as much as 5 points in a total range of 7. Therefore, it was evident that relation of free fatty acid to flavor was not dependent on quantity alone.

Wheat Ration for Hogs

In a project on relation of pork quality to ration based respectively on soybeans, corn, sorghum grain, and wheat, acidity of back fat varied with the ration (*2*). Average acid numbers for back fat with ration were: soybean, 0.15; sorghum grain, 0.21; corn, 0.38; and wheat, 0.50. A scattergram of fat flavor scores *vs.* acid numbers is shown in Figure 3 and is treated as in Figure 1.

The distribution of points is similar in the two diagrams. The boundaries of the two blank areas, *D*, intersected within 0.1 score unit and 0.03 acid unit of each other. All but one of the soybean points fell in area *A*. All of the sorghum grain points fell in area *B*. All of the wheat points fell in area *C*. The corn points were distributed among areas *A*, *B*, and *C*. Fat flavor scores for the wheat ration average 6.7; sorghum grain, 6.2; corn, 5.9; and soybean, 5.7. (Highest possible score, 7.) Lean flavor for wheat ration was also highest, 6.9, and lowest for soybeans, 5.4.

The variation in fat acidity in this case attended a variation in feed rather than in stress and depletion, as in the previous case. Nevertheless in both cases, the acid changes were brought about by normal in vivo metabolic processes and were attended by similar flavor responses. In this case, as in the previous one, post-mortem changes in the stored fat were attended by increase in acidity and lower flavor scores. However, flavor of fat from wheat ration maintained its superior position over those from other rations during storage.

Deep-fat Frying

In a third project, variation in fat acidity was brought about by external treatment, not metabolic and not autolytic (*4*). The flavor of French fried potatoes prepared in fresh, neutral, hydrogenated vegetable fat was scored only average. But with repeated use of the fat, the flavor score of the potatoes and acidity of the fat increased simultaneously. The flavor reached a maximum (superior) rating when the acid number of the fat reached approximately 5.5 with 16 frying periods of the same fat (Figure 4). Each frying period included the following conditions: 3 hours of preheating at 93° C., one quarter hour to raise to 185° C., 2 hours at 185° C., and $2^3/_4$ hours at 93° C. Thereafter occurred a rapid increase in fat acidity and a rapid decline in desirability of potato flavor with repeated use of the frying fat.

Reactions, other than hydrolysis, probably occurred in the fat as indicated by the deepening color of the fat and fading color of the potatoes, which may have affected the flavor. But significantly, an increase in flavor score was at least coincidental with, and not inhibited by, an increase in free fatty acid up to a certain point. It would not be justifiable to assume that this point of acidity was the same in the fat absorbed by the potato as it was in the frying fat. The potato slice is essentially an aqueous system, and may be expected to adsorb preferentially the free fatty acid molecules through their hydrophyllic free carboxyl groups. This behavior would tend to increase the percentage of free fatty acid in the fat held by the potato even above that in the frying fat.

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LEMON OIL COMPOSITION

Isolation and Identification of Aldehydes in Cold-Pressed Lemon Oil

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IT IS GENERALLY assumed that aldehydes are responsible for the characteristic aroma of lemon oil. The part played by individual aldehydes in this respect is not well established, although it is generally recognized that citral is the most important component contributing to the aroma of lemon oil. One of the objective measures of the quality of lemon oil is the determination of the total aldehyde content calculated as citral. However, the nature and relative amounts of individual aldehydes must be determined before any correlation can be made between organoleptic properties and composition. Recently, a specific method of analysis for citral was developed (*15*), and analyses of citral and total aldehyde content of lemon oils have shown notable variations in the ratios of citral to total aldehydes (*16*). These ratios were found to vary from 0.60 to 0.80. Changes in individual aldehydes other than citral have not been determined because specific methods of analysis were not available. This investigation was made to isolate and identify individual aldehydes in lemon oils as a basis for further studies on changes in their composition or amounts as related to changes in organoleptic and other quality factors.

Poore (*12*) reported the presence of

An investigation of the aldehydes of lemon oil has established the presence of octanal, nonanal, decanal, and undecanal by both infrared spectra and preparation of solid derivatives. Heptanal was identified by melting point and infrared spectrum of its 2,4-dinitrophenylhydrazone. Citronellal, neral, and geranial were identified by melting points and mixed melting points of their 2,4-dinitrophenylhydrazones. Dodecanal, tridecanal, tetradecanal, pentadecanal, hexadecanal, and heptadecanal were tentatively identified by gas chromatography using a plot of log retention time *vs.* number of carbon atoms for five column packings. Retention times on four stationary phases indicated the presence of hexanal.

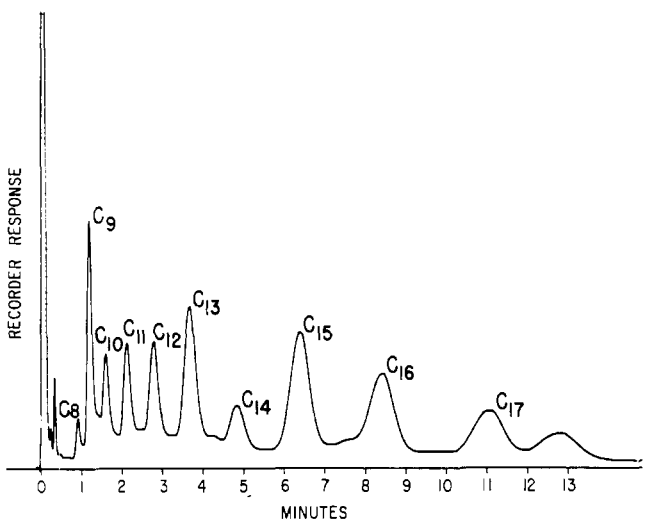

Figure 1. Chromatogram of fraction C on 1/4 inch × 10 feet diethylene glycol succinate column, at 190° C. and 85 ml. of helium per minute, 15 pounds inlet pressure

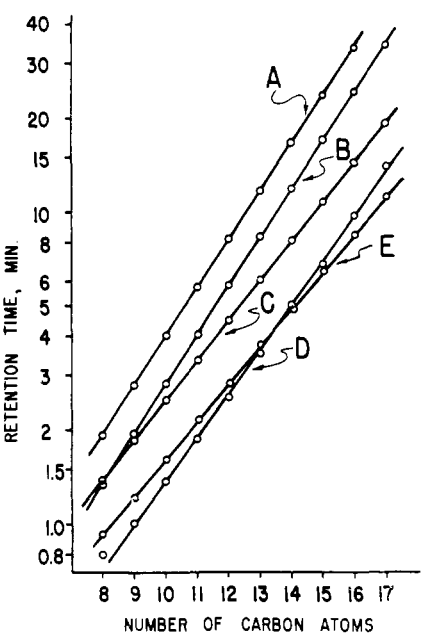

Figure 2. Plot of logarithm retention time *vs.* numbers of carbon atoms

A. Column, 1/4 inch × 10 feet, Ucon polar at 199° C. and 110 ml. of helium per minute, 15 pounds inlet pressure
B. Column, 1/4 inch × 5 feet, silicone GS SF-96 at 196° C. and 50 ml. of helium per minute, 10 pounds inlet pressure
C. Column, 1/4 inch × 10 feet, butanediol succinate at 202° C. and 70 ml. of helium per minute, 20 pounds inlet pressure
D. Column, 1/4 inch × 4 feet, Silastic 132 at 200° C. and 37 ml. of helium per minute, 5 pounds inlet pressure
E. Column, 1/4 inch × 10 feet, diethylene glycol succinate at 196° C. and 85 ml. of helium per minute, 15 pounds inlet pressure

octanal, nonanal, and citral in domestic lemon oils. Guenther (*4*) in reviewing the extensive early work on Italian lemon oils reported the presence of octanal, nonanal, decanal, dodecanal, and citral. The presence of citronellal was reported, but not confirmed. Recently, Calvarano (*2*) verified some of these findings using paper chromatography. Bernhard (*1*) reported the presence of a series of saturated, normal, aliphatic aldehydes, from C_6 to C_{12} inclusive, in domestic lemon oils by comparison of retention times using gas chromatography. Kung, Bambara, and Perkins (*7*) and Liberti and Cartoni (*8*) using gas chromatography demonstrated the separation of citral in lemon oil into neral and geranial.

A preliminary distillation was made in the isolation and identification procedure for determining the major aldehydes of lemon oil in order to remove high boiling compounds and dissolved solids. This separation was made to maintain the pot temperature as low as possible in the subsequent vacuum fractional distillation. Silicic acid chromatography was used to separate terpene hydrocarbons from oxygenated compounds as described by Kirchner and Miller (*5*). The oxygenated compounds were fractionally vacuum distilled, and the fractions were analyzed by gas chromatography. Fractions were combined according to the normal, saturated aldehydes as indicated by their retention times. Since composite fractions contained compounds other than aldehydes, further separation was made using silicic acid column chromatography. The normal saturated aldehydes have higher R_f values on chromatostrips than a number of other groups of organic compounds (*10*). It was therefore reasonable to assume that the aldehydes would be among the first compounds eluted from a silicic acid column using hexane–ethyl acetate as a developing solvent.

Equipment and Material

Gas Chromatographic Apparatus. A commercial unit equipped with a four-filament thermal conductivity detector; 5-mv. recorder with 2-second pen response, chart speed 40 inches per hour.

Columns. One quarter inch × 10 feet and 1/2 inch × 5 feet (preparative) Ucon polar; 1/4 inch × 10 feet butanediol succinate (Craig polyester succinate); 1/4 inch × 10 feet diethylene glycol succinate; 1/4 inch × 5 feet silicone GE SF-96. The above columns were of stainless steel with 25% w./w. of liquid phase on 42- to 60-mesh C-22 firebrick (Wilkens Instrument, Inc.) and 1/4 inch × 4 feet (copper) Dow Corning Silastic 132, 25% w./w. on 60- to 80-mesh C-22 firebrick.

Fraction Collector. Two hundred and forty tubes (18 × 150 mm.), time actuated.

Experimental

Major Aldehydes. A sample of cold-pressed lemon oil (4430 grams) was rapidly distilled in a Claisen flask at 1

Table I. Distillation of Oxygenated Compounds of Lemon Oil

Fraction No.	Weight, Grams	Boiling Range, °C	Pressure, Mm. Hg
1	1.0	52–58	18
2	1.7	58–60	18
3	2.9	60–64	20
4	4.7	64–65	20
5	6.8	65	20
6	3.7	65	21
7	5.0	65–68	21
8	2.9	68–73	21
9	2.2	73–77	21
10	0.7	77	21
11	3.3	76–79	16
12	2.6	79–82	15
13	2.2	79–82	12
14	3.9	82	12
15	3.9	82	12
16	2.5	82–84	12
17	1.4	84–86	10
18	3.1	86–87	9
19	4.5	87	9
20	4.5	87–90	9
21	5.7	90	9
22	10.8	90	9
23	6.7	80	8
24	5.9	81–82	8
25	34.4	82–90	8
26	8.3	90–98	8
27	1.1	84–86	5
28	2.3	86–88	5
29	5.4	88	5
30	2.1	88	5
31	5.9	88	5
32	4.9	78	2.5
33	4.2	60	1
Residue	16.6		

mm. of Hg pressure, and the distillate was collected until the flask temperature rose to 100° C. The yield was 4240 grams of distillate and 170 grams of residue. Terpene hydrocarbons in the distillate were separated from oxygenated compounds by chromatography on a 6-inch diameter column of powdered silicic acid (1250 grams) (*5*). Silicic acid was activated prior to use by heating at 180° C. for 24 hours in an air-circulating oven. The column was packed with a suspension of activated silicic acid in two volumes of redistilled hexane (*9*). Hydrocarbons were eluted from the column with hexane. Samples of eluate were tested at regular intervals on chromatostrips using fluorescein-bromine spray reagent for detecting unsaturated compounds (*6*). When terpene hydrocarbons were eluted, the developing solvent was changed from hexane to ethyl acetate (5 liters) followed by absolute ethanol (2 liters) to remove oxygenated compounds. The combined ethyl acetate–ethanol eluates were concentrated in a rotating vacuum evaporator, and the residual mixture (192 grams) was distilled at reduced pressure through a 1.5 × 95 cm. spinning brush column. Thirty-three fractions were collected (Table I), analyzed by gas chromatography (1/4-inch Ucon column, 175° C., 20 pounds helium inlet pressure), and composites made accordingly.

Octanal. Fractions 1, 2, 3, and 4 were combined and chromatographed on a silicic acid column (5.5 × 35 cm.) using hexane, followed by 0.5% (v./v.) of ethyl acetate in hexane to elute aldehydes. The eluate was collected in 15-ml. fractions. Each fraction was tested on chromatostrips, and zones were detected by spraying with fluorescein-bromine and 2,4-dinitrophenylhydrazine reagents. Fractions indicating presence of aldehyde constituents were combined, and developing solvents were removed under vacuum in a rotating flash evaporator. A portion of the residue was converted to the 2,4-dinitrophenylhydrazone (2,4-DNPH) as described by Shriner and Fuson (*13*). The product, recrystallized from absolute ethanol, melted at 107° to 108° C. The mixed melting point with the 2,4-DNPH of an authentic sample of octanal was not depressed.

Analysis, calculated for $C_{14}H_{20}N_4O_4$: C, 54.5%; H, 6.54%; N, 18.2%. Found: C, 54.6%; H, 6.53%; N, 17.9%.

The methone derivative was prepared as described by Shriner and Fuson (*13*), recrystallized from aqueous ethanol, and had a m.p. at 90° C. The mixed melting point with an authentic methone derivative of octanal was 89° to 90° C.

Analysis, calculated for $C_{24}H_{38}O_4$: C, 73.8%; H, 9.81%. Found: C, 73.6%; H, 9.73%.

Another portion of the residue was gas chromatographed on a preparative scale, Ucon polar column (160° C., 20 pounds helium inlet pressure). The material having the retention time of octanal was collected and analyzed by infrared spectrophotometry. The spectrum was identical with that of commercial octanal purified in a similar manner.

Gas chromatographic analysis of fractions 5 to 8 indicated the presence of both octanal and nonanal. No further investigations were made on these fractions.

Nonanal. Fractions 9, 10, 11, and 12 were combined and chromatographed on silicic acid, and eluted fractions were tested for aldehydes as described for octanal. Eluates were also tested by gas chromatography. The 2,4-DNPH was prepared from fractions containing the largest quantity of nonanal, and recrystallized from absolute ethanol, m.p. 105° to 107° C. The mixed melting point with an authentic 2,4-DNPH derivative of nonanal was not depressed.

Analysis, calculated for $C_{15}H_{22}N_4O_4$: C, 55.9%: H, 6.88%; N, 17.4%. Found: C, 56.0%; H, 6.89%; N, 17.0%.

The methone derivative was prepared and recyrstallized from aqueous ethanol, m.p. 87° C. The mixed melting point with an authentic methone derivative of nonanal was not depressed.

Analysis, calculated for $C_{25}H_{40}O_4$: C, 74.2%; H, 9.96%. Found: C, 74.2%; H, 9.85%.

The sample of nonanal was purified by gas chromatography for infrared analysis as described for octanal. The infrared spectrum was identical with that of an authentic sample of nonanal purified by gas chromatography.

Fraction 13 was not analyzed because of the number of components indicated by gas chromatographic analysis.

Citronellal. Distillation fraction 14 appeared by gas chromatographic analysis to contain citronellal. This material was converted to the water-soluble Girard T derivative, extracted with hexane to remove noncarbonyl components, and regenerated with aqueous formaldehyde (*17*). The regenerated citronellal was analyzed by gas chromatography and found to be contaminated with nonanal and decanal. The citronellal was separated by gas chromatography on the preparative scale gas chromatographic column as described for the infrared analysis of octanal. The infrared spectrum was found to be identical with that obtained for an authentic sample of citronellal which had been purified by gas chromatography.

The 2,4-DNPH derivative was prepared and purified by chromatography on a Celite–silicic acid column (*3*) using 0.5% (v./v.) of ethyl ether in hexane as developing solvent. One major and two minor yellow bands were recovered by elution. Eluate containing the major band was evaporated and the product 2,4-DNPH was recrystallized from absolute ethanol, m.p. 79° to 79.5° C. Mixed melting point with the 2,4-DNPH of an authentic sample of citronellal purified in the same manner was not depressed.

No further investigation was made on fraction 15.

Decanal. Distillation fractions 16 through 19 were combined and chromatographed on silicic acid as described for octanal. Fractions containing the largest amount of decanal by gas chromatographic analysis were combined, and developing solvent was removed. The 2,4-DNPH was prepared and recrystallized from absolute ethanol, m.p. 105° to 106° C. The mixed melting point with the 2,4-DNPH derivative of an authentic decanal was not depressed.

Analysis, calculated for $C_{16}H_{24}N_4O_4$: C, 57.1%; H, 7.19%; N, 16.6%. Found: C, 57.0%; H, 7.14%; N, 16.3%.

The methone derivative was prepared and recrystallized from aqueous ethanol, m.p. 90° C. The mixed melting point with methone derivatives of known decanal was 89° to 90° C.

Analysis, calculated for $C_{26}H_{42}O_4$: C, 74.6%; H, 10.1%. Found: C, 74.1%; H, 9.9%.

The decanal fraction was purified by gas chromatography for infrared analysis as described previously for octanal. The infrared spectrum was virtually identical with that of an authentic decanal purified in the same manner with the exception of one minor band at 8.05 microns. This band was eliminated by passing the material a second time through the preparative scale gas chromatography column.

Citral. The only carbonyl compounds found in fractions higher than 19 were neral and geranial. The aldehydes were detected by gas chromatography and by 2,4-DNPH reagent. Fraction 26 contained the largest quantity of neral and geranial as analyzed by gas chromatography. Distillation fraction 26 was treated with Girard T reagent (*17*). The 2,4-DNPH's were prepared from the regenerated material and chromatographed on a Celite–silicic acid column (*3*). Two major bands appeared and were eluted from the column using 0.5% (v./v.) of ethyl ether in hexane; the faster moving band was bright red-orange, the slower band yellow-orange.

Geranial. The eluate containing the slower moving yellow-orange zone was evaporated to dryness, and the residue was crystallized from absolute ethanol, m.p. 127° C.

Analysis, calculated for $C_{16}H_{20}N_4O_4$: C, 57.8%; H, 6.06%; N, 16.9%. Found: C, 57.9%; H, 6.00%; N, 16.8%.

An authentic sample of citral containing 95% of geranial (determined by gas chromatographic analysis) was converted to the 2,4-DNPH derivative and chromatographed on Celite–silicic acid as described above. Only one band was visible on the column. This band was eluted, and the developing solvent was evaporated. The product was recrystallized twice from ethanol to yield orange-red needles, m.p. 126° to 128° C.; the mixed melting point with material from the slower moving band of lemon oil was not depressed.

Neral. Eluate containing the faster moving 2,4-DNPH derivative from lemon oil was evaporated to dryness, and the residue was recrystallized twice from absolute ethanol. The product, glistening red platelets, melted at 122° to 123° C.

Analysis, calculated for $C_{16}H_{20}N_4O_4$: C, 57.8%; H, 6.06%; N, 16.9%. Found: C, 57.8%; H, 6.04%; N, 16.6%.

A known synthetic sample of citral reported to contain 69% of neral and 31% of geranial (determined by gas chromatographic analysis) was converted to the 2,4-DNPH derivative and column chromatographed as above on Celite–silicic acid. Two zones appeared. The material in the slower moving band showed no mixed melting point depression with geranial 2,4-DNPH. The crystalline product from the faster moving red-orange zone was recrystallized from absolute ethanol, m.p. 122° to 123° C. The melting point of a mixture of the faster moving 2,4-DNPH derivative from lemon oil and the faster moving 2,4-DNPH from synthetic citral was not depressed.

Relative amounts of the major aldehydes in lemon oil were determined directly by conversion to the Girard T derivatives. These data are presented in a separate publication in which the recovery method is completely described (*14*).

Minor Aldehydes. One thousand grams of cold-pressed lemon oil were chromatographed on a silicic acid column (7.5 × 40 cm.) (*9*) and developed with hexane, followed consecutively by 0.5 and 1% (v./v.) mixtures of ethyl acetate in hexane. Eluate fractions were tested using chromatostrips and gas chromatography as previously described for octanal. Fractions containing the normal, saturated, aliphatic aldehydes with a trace of citral were combined, and the developing solvent was removed by vacuum evaporation. The residue was converted to the water-soluble Girard T derivative by refluxing for 1 hour with Girard T reagent (25 grams), IRC-50 ion exchange resin (acid form, 2.5 grams) and 2-propanol (1000 ml.). The reaction mixture was diluted with water (1500 ml.) and extracted successively with 500, 400, and 200 ml. portions of hexane to remove noncarbonyl material. To the aqueous phase was added 30 ml. of aqueous formaldehyde (36%) and 100 ml. of redistilled petroleum ether (b.p. 30° to 60° C.) to regenerate the aldehydes. The mixture was allowed to stand for 24 hours at 39° C. under nitrogen in a glass-stoppered bottle. The aqueous phase was then removed and extracted with two 150-ml. portions of petroleum ether. The three petroleum ether phases were combined, dried over anhydrous sodium sulfate, and the solvent was removed by vacuum evaporation. The residue (2.4 ml.) was gas chromatographed (0.3 ml. per pass) on the preparative scale, polar Ucon column (205° to 220° C., 30 pounds helium inlet pressure). The products were collected in three fractions: fraction *A* containing components with retention times corresponding from octanal to decanal; fraction *B* containing undecanal and dodecanal; and fraction *C* containing tridecanal and higher molecular weight aldehydes.

Undecanal. Fraction *B* was further separated on the preparative scale, polar Ucon column (180° C., 10 pounds helium inlet pressure) into fractions *B-1* and *B-2*, having retention times corresponding respectively to undecanal and dodecanal. The infrared spectrum of fraction *B-1* was found to be identical with that of a known sample of undecanal similarly purified. The 2,4-DNPH was prepared from fraction *B-1* and recrystallized from absolute ethanol, m.p. 103° to 105° C. Mixed melting point with the 2,4-DNPH derivative of an authentic sample of undecanal was 105° to 106° C.

Analysis, calculated for $C_{17}H_{26}N_4O_4$: C, 58.2%; H, 7.48%; N, 16.0%. Found: C, 58.3%; H, 7.35%; N, 15.4%.

C_{12} to C_{17} Normal, Saturated Aldehydes. The amount of dodecanal (fraction *B-2*) was too small for the preparation of derivatives or infrared analysis. Consequently, this fraction was added to fraction *C*, and the mixture was analyzed by gas chromatography. The following columns were used: Silastic No. 132, silicone oil, Ucon polar, butanediol succinate, and diethylene glycol succinate. A chromatogram of fraction *C* is shown in Figure 1. The last peak was not identified. Because of incomplete desorption, small amounts of the lower molecular weight aldehydes (C_8 to C_{11}) were also found in fraction *C*. Consequently, it was possible to plot logarithms of retention times of higher molecular weight aldehydes as an extension of the curve obtained for the identified lower members of the homologous series. Plots of logarithm retention time against number of carbon atoms for components in the combined fraction *B-2* and *C* obtained with the five stationary phases appear in Figure 2. It is concluded from this evidence that small amounts of the normal, saturated, aliphatic aldehydes C_{12} through C_{17} are present in cold-pressed lemon oil.

Hexanal and Heptanal. To obtain these low-boiling aldehydes, a 1000-ml. sample of cold-pressed lemon oil was fractionally distilled at atmospheric pressure through a 2.8 × 120 cm. heli-grid column. The material boiling below 160° C. was collected in six fractions. Fractions 1 and 2 and fractions 3 and 4 were combined, and the aldehydes present were converted to the water-soluble Girard T derivative (*17*). Noncarbonyl material was removed by extraction with hexane, and the aldehydes were regenerated with aqueous formaldehyde. The regenerated material was analyzed by gas chromatography employing the following four stationary phases: butanediol succinate (126° C., 20 pounds helium inlet pressure); Apiezon (140° C., 20 pounds helium inlet pressure); silicon (154° C., 10 pounds helium inlet pressure); Ucon preparative (154° C., 20 pounds helium inlet pressure). Three peaks had retention times identical with those obtained for authentic samples of hexanal, heptanal, and octanal.

The 2,4-DNPH of heptanal was

prepared by bubbling material separated by gas chromatography (Ucon preparative) through the 2,4-DNPH reagent (*11*). A yellow precipitate was dried, and chromatographed on a Celite–silicic acid column as described above for citronellal. The product, fine yellow needles after crystallization from absolute ethanol, melted at 105.5° to 106° C. The mixed melting point with the 2,4-DNPH derivative of an authentic sample of heptanal was 106° to 107° C. The infrared analysis of the 2,4-DNPH derivative was identical with that of known heptanal.

The amount of hexanal was too small for conversion to derivatives. Consequently, proof of identity must rest on comparison of retention time with that of authentic material as described above.

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FRUIT FLAVORS AND ODORS

Isolation and Identification of Some Volatile Carbonyl Components from Orange Essence

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An analytical procedure is described for the detection and identification of carbonyl components in trace amounts from a commercial orange essence. Volatile organic components were separated as an oil by liquid-liquid extraction of the essence and fractionated by gas-liquid chromatography. Carbonyl peaks were detected by bubbling the effluent gas through an ethanolic solution of 2,4-dinitrophenylhydrazine sulfate and identified by their dinitrophenylhydrazones. Carbonyl components identified include acetaldehyde, hexanal, hexenal (two isomers), octanal, octenal (?), furfural (?), neral, geranial, and carvone.

THE IMPORTANCE of oxygenated compounds to the flavor of citrus juices and citrus oils has been described by many investigators. Stanley (*9*) states that although the terpene hydrocarbon, *d*-limonene, is the major component of citrus oils, the oxygenated terpenes representing only about 5% of the oil provide the aroma typical of the individual fruit. Nelson and Mottern (*6*) identified *n*-decyl aldehyde and citral as components of Florida orange oil. More recently, Kirchner and Miller (*5*) reported the presence of acetaldehyde, acetone (trace), furfural (trace), hexanal, octanal, decanal, 2-dodecenal (?), citronellal, three C_{15} carbonyls, and carvone in fresh California Valencia orange juice. The volatiles were removed from the orange juice by low temperature distillation. The lower the pressure and the lower the subsequent distillation temperature, the less was the alteration in the typical aroma of the juice.

Workers in many fields have found the 2,4-dinitrophenylhydrazones to be useful derivatives in the identification of aldehydes and ketones. Ellis, Gaddis, and Currie (*2*) reported a rapid paper chromatographic method for the separation and tentative identification of saturated aldehyde dinitrophenylhydrazones. Gaddis and Ellis (*3*) extended this method to include the unsaturated aldehyde derivatives. Ross (*7*) and Jones, Holmes, and Seligman (*4*) studied the infrared spectra of dinitrophenylhydrazones and found them very useful for the identification of the parent carbonyl compounds. The latter also found that the position of the N—H stretching band, usually found near 3.05 microns, could be used to determine whether the parent compound was an aldehyde or a ketone.

Apparatus and Reagents

Gas chromatographic separations were carried out on an F & M Model 202 programmed temperature gas chromatograph equipped with a thermistor-type detector. A 6-foot column, 0.25-inch o.d., packed with 30 weight %