

# Identification of Potent Flavor Compounds Formed in an Aqueous Lemon Oil/Citric Acid Emulsion

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The flavor compounds of a fresh and a stored (30 days at 37 °C) emulsion of lemon oil in an aqueous citric acid solution were analyzed by high-resolution gas chromatography and eluate sniffing. Application of this technique on stepwise-diluted extracts of the volatile compounds allowed the determination of a new parameter, the FD factor, which reveals the most intense flavor compounds of the extract. On the basis of their high FD factors, geranial, neral, and linalool were identified as the most intense volatile flavor compounds of the fresh lemon oil. Lower FD factors were found for myrcene, limonene,  $\gamma$ -terpinene, octanal, nonanal, citronellal, 2(*E*)-nonenal, and decanal. After storage, neral, geranial, and citronellal were no longer sensorially detectable. On the other hand, *p*-methylacetophenone, *p*-cresol, fenchyl alcohol, *p*-cymene, and 1-terpinen-4-ol were formed during storage and showed high FD factors.

It is well-known that lemon oil is an unstable food flavoring, which deteriorates in the presence of acids and by autoxidation reactions. The most significant flavor compound among the more than 130 volatiles found in lemon oil (Van Straten and Maarse, 1983) is citral, which consists of the geometric isomers neral and geranial (Ikeda et al., 1962; Lund and Bryan, 1976). In addition, limonene, myrcene, octanal, and  $\gamma$ -terpinene among others contribute with high aroma values to the flavor of lemon oil (Drawert and Christoph, 1984). The aroma value (Rothe and Thomas, 1963) also called the "flavor unit" (FU) by Guadagni et al. (1966) of a compound is defined as the ratio of its concentration in a flavor extract to its odor threshold.

At low pH values, the concentration of citral decreases rapidly by a series of cyclization and oxidation reactions that have been studied in detail (Baines et al., 1970; Clark et al., 1977; Mc Hale et al., 1979; Kimura et al., 1981-1983; Peacock and Kuneman, 1985).  $\beta$ -Pinene, a main component of lemon oil, was also found to be unstable under these conditions (Kimura et al., 1984). Among the numerous products formed by the acid-catalyzed degradation of citral and  $\beta$ -pinene, the compounds listed in Table I have been postulated to be responsible for off-odors in deteriorated lemon oil containing products. However, these assumptions must be proven by the combined application of instrumental and sensorial analytical methods, since potent off-odor compounds formed during storage are of interest as indicator substances for the quality assessment of lemon-flavored products.

In order to detect such compounds, the volatiles isolated from fresh cold-pressed lemon oil and from an aged emulsion of the same lemon oil in aqueous citric acid were analyzed by high-resolution gas chromatography (HRGC) and eluate sniffing. Application of this technique to stepwise-diluted extracts of the volatile compounds allowed the determination of their flavor dilution (FD) factors (Schieberle and Grosch, 1987; Schmid and Grosch, 1986; Ullrich and Grosch, 1987). [The term "dilution value" (*D* value) previously used by Ullrich and Grosch (1987) is now denominated "flavor dilution factor" to distinguish it from the "decimal reduction value" (*D* value) used in microbiology and enzymology.] The compounds with the highest FD factors are the most intense odor compounds of an aroma extract since the FD factor of a compound is proportional to its aroma value (Ullrich and Grosch, 1987).

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**Table I. Postulated Lemon Off-Odor Substances Formed from Lemon Oil under Acidic Conditions**

compound	reference
<i>p</i> -cymene	Iwata et al. (1968)
$\alpha$ , <i>p</i> -dimethylstyrene	Iwata and Yamamoto (1978)
geranic acid	Kimura et al. (1981)
piperitenone	Kimura et al. (1981)
2-hydroxy- <i>p</i> -cymene	Kimura et al. (1981)
<i>p</i> -cymen-8-ol	Kimura et al. (1983)
fenchyl alcohol	Kimura et al. (1984)

## EXPERIMENTAL SECTION

**Lemon Oil.** Cold-pressed lemon oil (*Citrus limon*) from Sicilia/Italy was stored in sealed flasks at -60 °C under nitrogen before use.

**Chemicals.** Myrcene, *d*-limonene, *p*-cymene,  $\gamma$ -terpinene, (+)-1-terpinen-4-ol, (*E*)-2-nonenal, *p*-methylacetophenone, fenchyl alcohol, ( $\pm$ )-linalool, and citral were from Aldrich (Steinheim, Germany); *p*-cresol was from Merck (Darmstadt, Germany). Octanal, nonanal, and decanal were from Fluka (Buchs, Switzerland), citronellal and xanthan gum were from Serva (Heidelberg, Germany), 3-decanol was from ICN (Heidelberg, Germany), and Lamegin was from Gruenau (Illertissen, Germany). The solvents were purified as done by Schieberle and Grosch (1983). Silica gel 60 (Merck, Darmstadt, Germany) was treated with HCl and deactivated with 7% (w/w) water according to Esterbauer (1968).

**Storage Experiments.** Two emulsified samples were prepared, each consisting of 500 mg of the lemon oil, 200 mg of the emulsifier Lamegin, 60 mg of xanthan gum, and 9.24 g of a 5% (w/v) aqueous citric acid solution. One sample was immediately suspended in 500 mL of 0.05 M phosphate buffer (pH 7.0) and distilled as described below. The second emulsion was stored in a sealed vessel (50-mL total volume) at 37 °C for 30 days and then suspended in 500 mL of the phosphate buffer for distillation.

**Isolation of Volatiles by Simultaneous Distillation/Extraction (SDE).** The suspensions of the lemon oils were distilled and continuously extracted for 2 h with 100 mL of pentane-diethyl ether (2:1, v/v) in the apparatus designed by Nickerson and Likens (1966). The extracts obtained were dried over Na<sub>2</sub>SO<sub>4</sub> and after concentration on a Vigreux column (50 × 1 cm) filled to a total volume of 5 mL.

**Column Chromatography.** One milliliter of each of the above extracts was fractionated at 10-12 °C on a water-cooled column (30 × 1.6 cm) packed with a slurry of silica gel in pentane. The elution was performed with

**Table II. HPLC Separation<sup>a</sup> of Fraction C Obtained by Column Chromatography of Fresh Lemon Oil**

subfraction	elution range, mL
I	12.2–15.0
II	18.0–21.0
III	28.4–31.6
IV	31.7–35.0

<sup>a</sup> Elution was performed with pentane–diethyl ether (95:5 v/v), flow rate 2.0 mL/min.

**Table III. HPLC Separation of Fractions C<sup>a</sup> and D<sup>a</sup> Obtained by Column Chromatography of Stored Lemon Oil**

fraction	pentane/diethyl ether ratio (v/v)	elution range, mL
C (16)	95:5	16.3–18.9
D (17)	90:10	20.0–31.0

<sup>a</sup> Elution was performed at a flow rate of 2.5 mL/min.

100 mL of pentane (fraction A), followed by 100 mL of 95:5 pentane–diethyl ether (v/v; fraction B), 100 mL of 9:1 pentane–diethyl ether (v/v; fraction C), and finally 100 mL of 8:2 pentane–diethyl ether (v/v; fraction D).

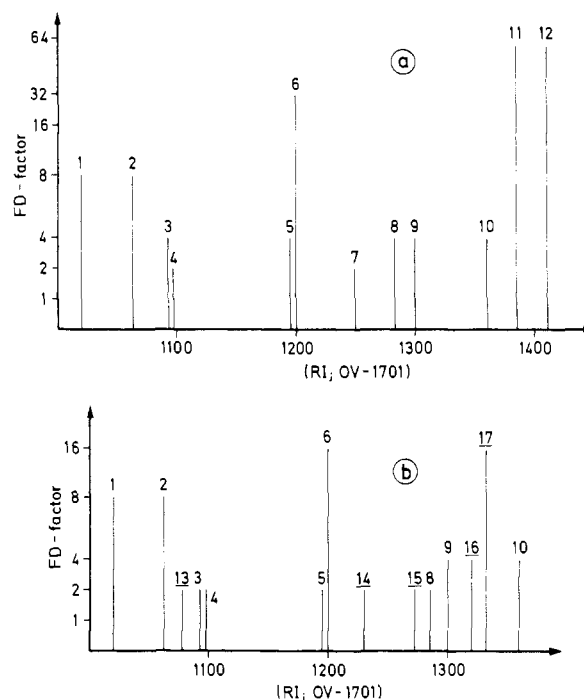
**High-Performance Liquid Chromatography (HPLC).** Fractions B–D from column chromatography of the fresh lemon oil and fractions C and D from the stored lemon oil were concentrated to 2 mL on a Vigreux column (50 × 1 cm) and further concentrated by microdistillation in the apparatus described by Bemelmans (1979). Further separation of each fraction was performed on a 50 × 0.46 cm stainless steel column filled with 5- $\mu$ m Shandon Hypersil (Shandon Products, Astmoor, GB) with use of the pentane–diethyl ether mixtures detailed in Tables II and III as elution media. The effluent of the column, which was coupled to an HPLC pump 110 B (Beckman, Muenchen, Germany) was monitored at 220 nm with a UV detector (SPD 2 A; Shimadzu, Duesseldorf, Germany) and fractionated as detailed in Tables II and III. To obtain enough material for mass spectrometry, the subfractions of about 20 runs were collected.

**Capillary Gas Chromatography (HRGC)–Mass Spectrometry (MS).** HRGC was performed with a Carlo Erba gas chromatograph, Type 4200. The thin-film capillary columns used were (i) capillary OV 1701 (30 m × 0.32 mm) glass capillary, coated with OV-1701 according to Grob and Grob (1979), and (ii) capillary SW 10 (30 m × 0.32 mm) fused silica capillary (Supelcowax 10; Supelchem, Sulzbach, Germany). The samples were applied by the on-column injection technique at 35 °C, and the temperature of the capillaries was raised by 60 °C/min to 50 °C, held 5 min isothermal, and then raised by 4 °C/min to 220 °C. The flow rate of the carrier gas helium was 2.2 mL/min. At the end of the capillary, the effluent was split 1:1 to a FID and a sniffing port (Schieberle and Grosch, 1983).

Retention data of the compounds are presented as retention indices (RI) calculated according to Halang et al. (1978).

HRGC–MS analyses were performed with a MS 8230 (Finnigan, Bremen, Germany) in tandem with the capillaries described above. Mass spectra in the electron impact mode (MS-EI) were generated at 70 eV and in the chemical ionization mode (MS-CI) at 115 eV with isobutane as reagent gas.

**Aromagrams.** After addition of 0.6 mg of 3-decanol as internal standard, aliquots (1 mL) of the pentane–diethyl ether extracts obtained by SDE of fresh or stored lemon oil were analyzed by HRGC and eluate sniffing as described by Schieberle and Grosch (1984); the sample size



**Figure 1.** Aromagrams of the volatiles isolated from fresh lemon oil (a) and from lemon oil stored in a dilute citric acid emulsion for 30 days at 37 °C (b). Numbering of the flavor compounds as in Tables IV and V.

of the extracts was adjusted on the basis of an equal level of the internal standard. The odor-active regions of the eluates were established and the aroma notes assigned.

The FD factors of the flavor compounds were determined by eluate sniffing of dilutions of the extracts obtained by stepwise addition of diethyl ether (Ullrich and Grosch, 1987). The aromagrams [plot of the FD factor of each flavor compound vs its retention index (Ullrich and Grosch, 1987)] were drawn.

As a control, 100 mg of fresh citrus oil was dissolved in 1 mL of diethyl ether, and after addition of 0.6 mg of 3-decanol, this sample was analyzed in the same way.

**Identification of Neral and Geranial.** Neral and geranial were isolated from citral by HPLC (Schieberle et al., 1988) and their mass spectra recorded. About 100  $\mu$ g of each of the aldehydes was taken up in 15 mL of methanol and treated with 2 mg of sodium borohydride. After 1 h the reaction mixtures were diluted with 30 mL of water, adjusted to pH 7.0 with diluted HCl, and extracted twice with 20 mL of diethyl ether–pentane (2:1, v/v). The organic layers were washed with 40 mL of a saturated NaCl solution and then dried over anhydrous  $\text{Na}_2\text{SO}_4$ .

## RESULTS

**Fresh Lemon Oil.** The aromagram of the volatiles isolated by simultaneous steam distillation/extraction (SDE) from fresh lemon oil (Figure 1a) showed 12 flavor compounds with FD factors of 2 and higher, of which 11 were subsequently identified. A preliminary experiment with fresh, undistilled lemon oil showed that the distillation procedure did not cause any changes in the composition of the flavor compounds or in the aromagram.

The separation of the extract containing the volatiles by column chromatography on silica gel gave four fractions designated A–D. HRGC combined with eluate sniffing showed that the flavor compounds were located in fractions A, C, and D (Table IV). Myrcene, limonene, and  $\gamma$ -terpinene were found in fraction A; linalool was the only flavor compound in fraction D. Fraction C was further separated by HPLC into four subfractions (Table II). As

Table IV. Important Volatile Flavor Compounds of Fresh Lemon Oil

peak no. <sup>a</sup>	compound	fraction <sup>b</sup>	subfraction <sup>c</sup>	RI		odor description
				OV-1701	SW 10	
1	myrcene <sup>d</sup>	A		1020	1145	ethereal, soapy
2	limonene <sup>d,e</sup>	A		1056	1178	ethereal, citrus-like
3	$\gamma$ -terpinene <sup>d</sup>	A		1089	1223	gasoline-like, ethereal
4	octanal <sup>d</sup>	C	I	1096	1273	soapy
5	nonanal <sup>d</sup>	C	I	1195	1378	soapy, floral
6	linalool <sup>d,e</sup>	D		1202	1537	floral
7	citronellal <sup>d</sup>	C	I	1246	1450	fatty
8	( <i>E</i> )-2-nonenal <sup>d</sup>	C	II	1282	1517	green, fatty
9	decanal <sup>d</sup>	C	I	1303	1484	soapy, floral
10	unknown	C	II	1360	1637 <sup>f</sup>	fatty, nut-like
11	neral	C	III	1383	1667	citrus-like, fatty
12	geranial	C	IV	1416	1715	citrus-like, pungent

<sup>a</sup> Peak in the aromagram (Figure 1a). <sup>b</sup> Fraction obtained by column chromatography on silica gel. <sup>c</sup> Subfraction obtained by HPLC (Table II). <sup>d</sup> The compound was identified by comparison to reference substances of the following parameters: RI on two capillaries of different polarity (OV 1701, SW 10), mass spectra obtained by MS-EI and MS-CI and odor quality. <sup>e</sup> The stereochemistry was not investigated. <sup>f</sup> The RI was determined on a fused silica capillary (25 m  $\times$  0.32 mm) CP Wax 52 (Chrompack; Müllheim, Germany).

listed in Table IV, the flavor compounds of this fraction were found in subfractions I (octanal, nonanal, citronellal, decanal), II ((*E*)-2-nonenal and a compound of unknown structure), III (neral), and IV (geranial).

The identification experiments (Table IV) together with the aromagram (Figure 1a) revealed that neral and geranial followed by linalool were the most potent aroma compounds of the fresh lemon oil.

With the exception of citronellal, (*E*)-2-nonenal, and decanal, the compounds listed in Table IV belong to the flavor compounds that, according to Drawert and Christoph (1984), contribute with high aroma values to the flavor of lemon oil. These authors found in addition relatively high aroma values for 1,8-cineole and ethyl hexanoate, both of which did not appear in the aromagram of the fresh lemon oil (Figure 1a). One reason for the differences might be that Drawert and Christoph (1984) used a different technique (headspace) for the isolation of the volatiles.

**Aged Lemon Oil.** The aromagram shown in Figure 1b was obtained after storage of the lemon oil emulsion for 30 days at 37 °C. A comparison of this aromagram with that of the fresh oil (Figure 1a) indicates that the concentrations of neral, geranial, and citronellal were so far decreased during storage that they did not appear in the aromagram of the aged lemon oil. As opposed to this, the FD factors of linalool,  $\gamma$ -terpinene, nonanal, and (*E*)-2-nonenal deviated only slightly from those found in the aromagram of the fresh lemon oil, and the FD factors of the resting flavor compounds did not change during storage of the lemon oil. However, the comparison of both aromagrams (Figure 1a,b) showed the appearance of five additional aroma compounds (peaks 13–17 in Figure 1b) in the stored lemon oil.

These flavor compounds were enriched by column chromatography on silica gel and in the cases of peaks 16 and 17 in addition by HPLC (Table III).

Table V lists the results of the identification experiments. Of the compounds formed during storage of the lemon oil emulsion, compound of peak 17 showed the highest FD factor (Figure 1b) and was identified as *p*-methylacetophenone. Its odor was described as "bitter almond-like".

The other additional flavor compounds of the stored lemon oil were found to be *p*-cresol, fenchyl alcohol, *p*-cymene, and 1-terpinen-4-ol (Table V).

## DISCUSSION

The results revealed that the decrease in the concentration of citral together with the formation of *p*-

Table V. Flavor Compounds Formed during Storage (37 °C, 30 Days) of Lemon Oil in the Presence of Citric Acid

peak no. <sup>a</sup>	compound	fraction <sup>b</sup>	RI		odor description
			OV-1701	SW 10	
13	<i>p</i> -cymene	A	1076	1245	gasoline-like
14	fenchyl alcohol	D	1230	1574	camphor-like
15	1-terpinen-4-ol	D	1273	1591	terpene-like, musty
16	<i>p</i> -cresol	D	1321	2067	phenolic
17	<i>p</i> -methylacetophenone	C	1332	1759	bitter almond-like

<sup>a</sup> Peak in the aromagram (Figure 1b). <sup>b</sup> Fraction obtained by column chromatography on silica gel.

methylacetophenone, *p*-cresol, fenchyl alcohol, *p*-cymene, and 1-terpinen-4-ol characterize the deterioration of lemon oil in the presence of citric acid. Comparison of these results with previous findings (cf. Table I) shows that *p*-methylacetophenone and *p*-cresol are reported for the first time as compounds contributing to the off-odor of stored lemon oil.

The decrease of neral and geranial during storage of mixtures containing citral or lemon oil in addition to citric acid agrees with the findings of Kimura et al. (1981, 1984) and of Peacock and Kuneman (1985).

1-Terpinen-4-ol has previously been reported to be a constituent of fresh lemon oil (Shaw, 1979; Staroscik and Wilson, 1982). Its presence in the aromagram of aged lemon oil is due to its large increase during storage (Schieberle et al., 1983). A similar increase of 1-terpinen-4-ol was found by Kimura et al. (1984) during storage of an alcoholic lemon beverage.

Citral emulsified in an aqueous citric acid solution leads to *p*-methylacetophenone and *p*-cresol during storage (Schieberle et al., 1988). A plausible mechanism for the formation of *p*-methylacetophenone may be derived from the model experiments of Kimura et al. (1983) and Peacock and Kuneman (1985). They suggest that geranial in acidic media rapidly isomerizes into neral, which in turn cyclizes, leading to the formation of a mixture consisting of *p*-menthadien-8-ols and *p*-menthadien-4-ol. The *p*-menthadien-8-ols are converted by dehydration, redox, and disproportionation reactions to *p*-cymene and  $\alpha$ ,*p*-dimethylstyrene. That the latter compound does not appear in the aromagram of the stored lemon oil could be explained by its odor threshold (665–2660 ng/L of air) being too high (Schieberle et al., 1988). However, oxidation of the  $\Delta^8$  double bond of  $\alpha$ ,*p*-dimethylstyrene might explain the formation of *p*-methylacetophenone whose odor threshold (2.7–10.8 ng/L of air), like that of *p*-cresol (0.3–1.0 ng/L of air), is relatively low (Schieberle et al., 1988).

The mechanism for the formation of fenchyl alcohol from  $\beta$ -pinene was proposed by Kimura et al. (1984). The reaction steps leading to formation of *p*-cresol are still unclear.

The results reported here suggest that *p*-methylacetophenone, *p*-cresol, *p*-cymene, and fenchyl alcohol could be useful as indicator substances for the assessment of the deterioration of lemon oil flavorings in acidic foods.

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**Registry No.** Neral, 106-26-3; geranial, 141-27-5; *d*-limonene, 5989-27-5; myrcene, 123-35-3; octanal, 124-13-0;  $\gamma$ -terpinene, 99-85-4; *p*-cymene, 99-87-6; (+)-1-terpinen-4-ol, 2438-10-0; (*E*)-2-nonenal, 18829-56-6; *p*-methylacetophenone, 122-00-9; fenchyl alcohol, 2217-02-9; ( $\pm$ )-linalool, 22564-99-4; *p*-cresol, 106-44-5; nonanal, 124-19-6; decanal, 112-31-2; citronellal, 106-23-0; citric acid, 77-92-9.

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