Aroma Composition of Oolong Tea and Black Tea by Brewed Extraction Method and Characterizing Compounds of Darjeeling Tea Aroma†

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Volatile components of various brewed oolong teas and black teas were analyzed by gas chromatography (GC) and gas chromatography/FTIR spectrometry/mass spectrometry. The GC pattern of brewed tea extracted by dichloromethane (brewed extract) differs greatly from the GC pattern of the extract prepared by simultaneous steam distillation and extraction method (SDE). The brewed extract includes higher amounts of acids, aromatic alcohols, and monoterpenediol and lower amounts of monoterpene alcohols than the SDE extract. The Darjeeling brewed extract, which has a more complicated aroma pattern, consists of four linalool oxides, linalool, geraniol, hexanoic acid, benzyl alcohol, 2-phenylethanol, *trans*-geranic acid, (*E*)-2-hexenoic acid, (*Z*)-3-hexenoic acid, and 2,6-dimethyl-3,7-octadiene-2,6-diol. The Darjeeling SDE extract consists of seven main components which include geraniol, linalool, four linalool oxides, and methyl salicylate. The brewed extract of Chan Pin oolong (red oolong tea), which is similar in aroma to Darjeeling and is made from tea leaves infested with green flies (*Emposca flavescens*) in the same way as Darjeeling, contains very high amounts of 2,6-dimethyl-3,7-octadiene-2,6-diol. This compound appears to be the precursor of 3,7-dimethyl-1,5,7-octatrien-3-ol and to be the most important factor in Darjeeling tea flavor.

Keywords: *Aroma composition of oolong tea; Darjeeling tea aroma; brewed extraction method; black tea aroma*

INTRODUCTION

Studies of *Camellia sinensis* tea aroma have been reported by various authors (Straten and Maarse, 1989). However, GC aroma patterns of the vacuum steam volatile, steam distillation volatile (by SDE) and head space vapor have failed to reproduce real tea aroma (Kobayashi and Kawakami,1991). In a previous paper, we reported on volatile constituents of Rooibos tea as affected by extraction process (brewed extraction method and SDE method) and model experiments using the two extraction methods (Kawakami et al., 1993a,b). The direct brewed extraction method (brewed extract) was found to be more suitable for reproducing real tea aroma than the SDE method. In this work, the aroma concentrations of various oolong and black teas were prepared by brewed extraction method and analyzed by gas chromatography and gas chromatography/FTIR spectrometry/mass spectrometry. Many kinds of tea aroma concentrates were prepared by SDE and compared with those of brewed extract. The characteristics of each tea aroma, especially Darjeeling tea flavor, were also clarified.

EXPERIMENTAL PROCEDURES

Materials. Two types of oolong tea and four types of black tea were imported from various manufacturing countries.

Oolong Tea. Huang Chin Kuei is a typical Chinese flavorous oolong tea produced in Fujian Province of the People's Republic of China (1991 product) and has a sweet, green, heavy note. Another oolong tea, Chan Pin oolong (red oolong tea), has a strong sweet fruity, floral, and green flavor similar to that of muscat grape and is a typical Taiwanese oolong tea cultivated under the best quality control and no-pesticide conditions and with intentional infestation with green flies; it is manufactured under a stronger partial leaf enzymatic fermentation process close to that used for black tea.

Black Tea. Three kinds of Darjeeling tea plucked at different periods [sample A, March 15; sample B, March 30; sample C, April 12 (all 1993)] in the same garden (Pandam Tea Estate, Darjeeling, West Bengal, India) are the best quality leaf grade, FTGFOPI. Darjeeling tea is also invariably infested with green flies and has a strong heavy green note similar to that of muscat or apple. Two kinds of Sri Lanka black tea, clone DT-1 and clone 2025, produced at a tea research institute in Sri Lanka (1993 product), have a strong floral flavor and a sweet flavor, respectively. A Chinese black tea, Keemun (1991 product), imported from Anhui Province, People's Republic of China, has a heavy and sweet note.

Sample Preparations. The aroma concentrate was prepared by two extraction methods: the brewed extraction method and the SDE method.

Brewed Extraction Method. One hundred grams of powdered sample from each tea (Huang Chin Kuei, Chan Pin oolong, three kinds of Darjeeling tea, clone DT-1, clone 2025, and Keemum) was brewed by 700 mL of deionized boiling water for 10 min. After filtration, the filtrate was saturated with sodium chloride and was extracted by using 200 mL of dichloromethane. The extract was dried over anhydrous sodium sulfate for 12 h. After sodium sulfate was filtrated out, the solvent was removed carefully by using a Kuderna-Danish evaporative concentrator.

SDE Method. Another 100 g of powdered sample (Huang Chin Kuei, three kinds of Darjeeling tea, clone DT-1, and clone 2025) was placed in a 1000 mL flask with 500 mL of deionized boiling water. The steam distillate was simultaneously ex-

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† Part of the present investigation has been presented orally at the Symposium of the Agricultural Chemical Society of Japan, Sendai, April 1993 (Kawakami et al., 1993b), and Tokyo, April 1994 (Kawakami et al., 1994).

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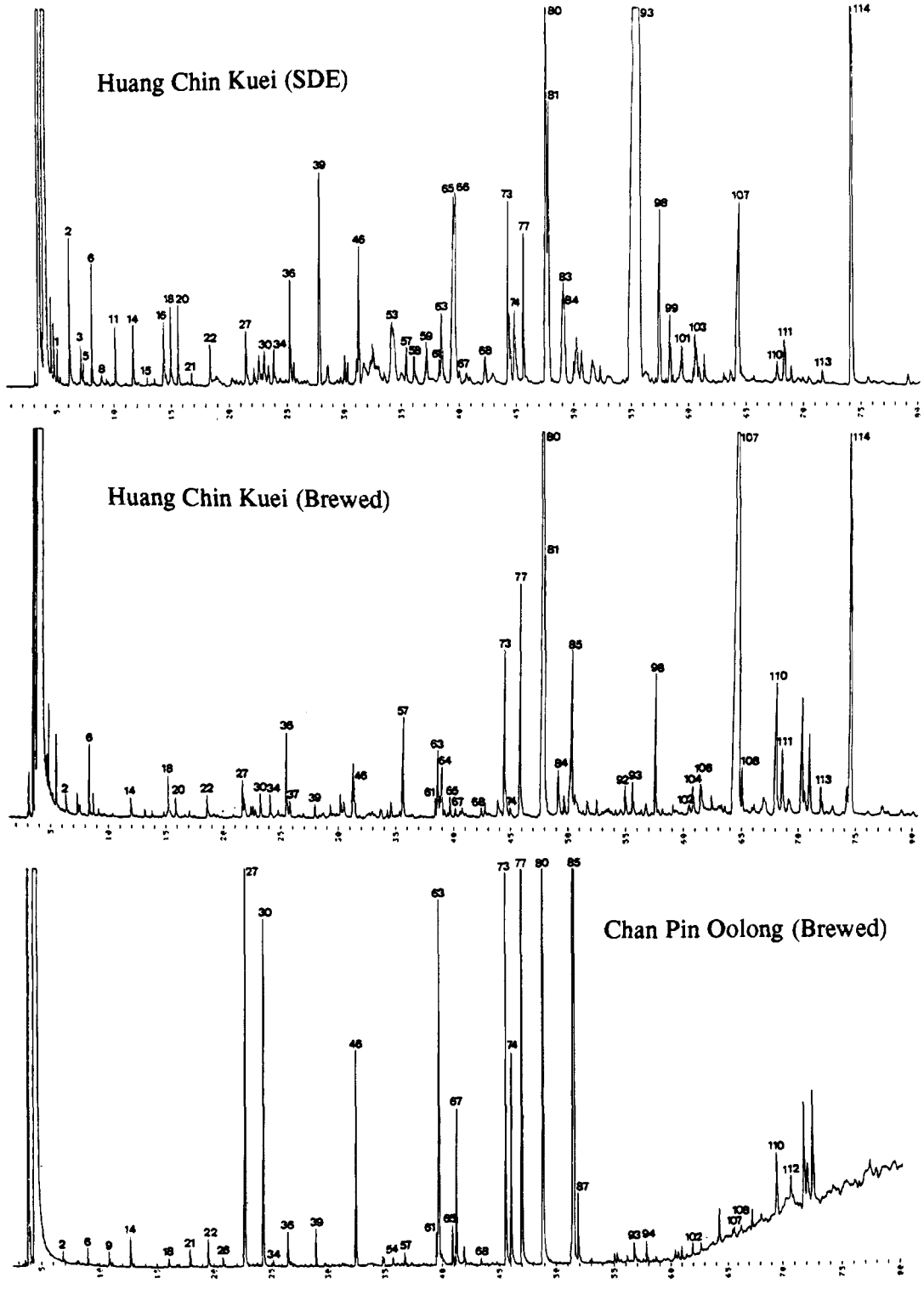

Figure 1. Gas chromatograms of the oolong tea extracts.

tracted with 50 mL of dichloromethane for 1 h by using a modified Likens–Nickerson apparatus (SDE method; Schultz et al., 1977). The extract was dried over anhydrous sodium sulfate for 12 h. After removal of sodium sulfate and solvent, aroma concentrate was obtained as described for the brewed extraction. Samples prepared by both extraction methods were then analyzed by gas chromatography (GC) and gas chromatography/IR spectrometry/mass spectrometry (GC/FTIR/MS).

Instrumental Methods. A Hitachi Model G-3000 gas chromatograph equipped with an FID and a 50m × 0.25 mm Carbowax 20M fused silica capillary column was used. The peak area was integrated by using a Hitachi D-2500 integrator. The oven temperature was held at 60 °C for 4 min and then programmed to 180 °C at 2 °C/min. The helium carrier gas flow rate was 30 cm/s. The injector and detector temperatures were 200 and 210 °C, respectively. A Hewlett-Packard 5972 mass spectrometer and a 5965B FTIR spectrometer interfaced to a Hewlett-Packard 5890 gas chromatograph were used for MS and IR identification. The GC conditions were as follows: column I, 60 m × 0.25 mm Carbowax 20M fused silica capillary column (held at 60 °C for 4 min and then programmed to 180 °C at 2 °C/min); column II, 25 m × 0.25 mm HP-5 column (held at 70 °C for 4 min and then programmed to 200 °C at 2 °C/min); and column III, CP-cyclodex column (held at 60 °C for 8 min and then programmed to 200 °C at 1 °C/min) for the use

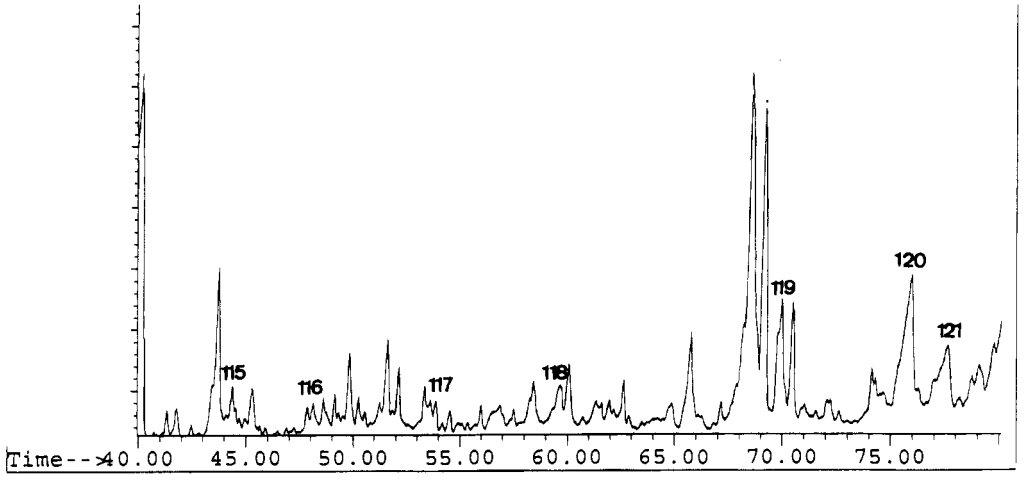

Figure 2. Gas chromatogram of the Chan Pin oolong tea extract analyzed by HP-5 column.

of diastereometric analysis. MS ion source temperature was 200 °C, and electron energy was 70 eV. The MS data and IR data were analyzed by a HP Chem Station system. The GC Kovats index (KI), IR spectrum, and MS fragmentation pattern of each component were compared to those of the authentic compound as reported in the literature.

RESULTS AND DISCUSSION

The aroma of the SDE extract emitted more greenish odor than that of each tea infusion, while the aroma of the brewed extract bore strong resemblance with that of each tea infusion. Gas chromatograms of two types of oolong tea are shown in Figure 1, and the gas chromatogram of Chan Pin oolong analyzed by HP-5 column is shown in Figure 2. The identified compounds and relative quantities of each compound, as calculated by the peak area, are listed in Table 1. Fifty-two compounds were identified from the brewed extract and 100 compounds from the SDE extract of Huang Chin Kuei, respectively. Forty-nine compounds were identified from the brewed extract of Chan Pin oolong by the Carbowax 20M column, and seven extra compounds by the HP-5 column. Of these, six ester compounds in Huang Chin Kuei extract, including hexyl hexanoate, 2-phenylethyl isobutyrate, 2-phenylethyl butyrate, 2-phenylethyl isovalerate, 2-phenylethyl benzoate, and 2-phenylethyl hexanoate, and five compounds in Chan Pin oolong extract, including dimethylmaleic anhydride, methylethylmaleimide, 2-indolinone, homovanillic acid, and coniferil alcohol, were newly identified as *C. sinensis* tea flavor (Straten and Maarse,1989). From the comparison of the brewed extract with the SDE extract of Huang Chin Kuei, the SDE extract shows thermal effects during the distillation and extraction process. Terpene alcohols such as nerolidol and linalool and terpenes such as α-farnesene, both of which have been reported previously as the main oolong tea flavor (Nobumoto et al., 1990), were found only in small amounts in brewed extract. The concentrate of brewed extract consists of high levels of jasmine lactone, 2-phenylethanol, and indole. Other important components in brewed extract were benzyl alcohol, benzyl cyanide, methyl jasmonate, 2-phenylethyl benzoate, hexanoic acid, and 2,6-dimethyl-3,7-octadiene-2,6-diol. Amounts of lactone compounds such as jasmine lactone, dihydroactinidiolide, 4-hexanolide, 5-decanolide, 4-butanolide, 4-nonanolide, and 2-hexen-4-olide were found to have decreased by the SDE process, as noted in previous studies (Kawakami et al., 1993a,b), and these lactones appear to be easily hydrolyzed to hydroxy acids by SDE.

The brewed extract from Chan Pin oolong consists of high levels of 2,6-dimethyl-3,7-octadiene-2,6-diol, 2-phenylethanol, benzyl alcohol, linalool oxides I, II, and III, and hexanoic acid. Other important components were geraniol and 3,7-dimethyl-1,5,7-octatrien-3-ol. The largest quantity component in Chan Pin oolong tea extract, 2,6-dimethyl-3,7-octadiene-2,6-diol, seemed to be the precursor of 3,7-dimethyl-1,5,7-octatrien-3-ol. The latter seemed to be formed easily from the former by dehydration during the SDE process. The amount of the latter compound has been reported to increase during the firing process of green tea manufacturing (Hara and Kubota,1984), and the latter has also been reported as the thermal induction of the former in juices of muscat grapes (Williams et al., 1980).

Gas chromatograms of Darjeeling SDE extract and four types of brewed black tea extracts are shown in Figure 3. The identified compounds and relative quantities of each compound, as calculated by the peak area, are listed in Table 2. Thirty-eight compounds were identified from the Darjeeling brewed extract and 52 compounds from the Darjeeling SDE extract. The Darjeeling brewed extract included higher amounts of acids, aromatic alcohols, and monoterpenediol and lower amounts of monoterpene alcohols than the SDE extract. The main components in the SDE extract were geraniol, linalool, linalool oxides II, I, and IV, and methyl salicylate, while the main components in the brewed extract were more complicated, including linalool oxide II, geraniol, linalool oxide IV, *trans*-geranic acid, linalool, (*E*)-2-hexenoic acid, benzyl alcohol, hexanoic acid, linalool oxide I, 2-phenylethanol, (*Z*)-3-hexenoic acid, and 2,6-dimethyl-3,7-octadiene-2,6-diol, in order of quantity. Terpene alcohols such as geraniol and linalool, which have been reported previously as the main Darjeeling tea flavor, decreased in amount in the brewed extract, similar to the result obtained for oolong tea. These terpene alcohols appeared to be formed by the SDE process from the precursors, glycosidic derivatives, namely primeverosides (Guo et al., 1994; Moon et al., 1994). 2,6-Dimethyl-3,7-octadiene-2,6-diol in Darjeeling tea brewed extracts was also found in larger quantities than in other black tea brewed extracts. 2,6-Dimethyl-3,7-octadiene-2,6-diol and the dehydration compound, 3,7-dimethyl-1,5,7-octatrien-3-ol, are characteristic factors common in both samples, Darjeeling tea and Chan Pin oolong tea, made from tea leaves infested with green flies (*E. flavescens*). These two compounds appeared to be related to Darjeeling's characteristic muscat flavor. It has been reported that the SDE extract of Taiwanese

Table 1. Composition of the Aroma Extracts of Chinese Oolong Teas, Ogonkei and Red Oolong

			GC peak area, %						GC peak area, %		
			Ogonkei		red oolong				Ogonkei		red oolong
PN	KI[a]	compound	SDE	brewed	brewed	PN	KI[a]	compound	SDE	brewed	brewed
1	991	1-penten-3-one	0.11	*b*	*b*	60	1685	dimethylmaleic anhydride[c]	*b*	*b*	0.07
2	1064	hexanal	0.56	0.09	0.10	61	1689	valeric acid	0.15	0.08	0.44
3	1100	(*E*)-3-penten-2-one	0.20	*b*	*b*	62	1689	naphthalene	0.12	*b*	*b*
4	1104	(*E*)-2-pentenal	0.07	*b*	*b*	63	1698	linalool oxide III (*trans*, pyranoid)	0.94	0.87	6.35
5	1110	4-methyl-3-penten-2-one	0.12	0.09	0.05						
6	1139	1-penten-3-ol	0.56	0.51	0.20	64	1700	2-hexen-4-olide	*b*	0.33	0.08
7	1158	2-heptanone	0.04	*b*	*b*	65	1727	methyl salicylate	1.17	0.71	0.61
8	1160	heptanal	0.08	*b*	0.05	66	1735	α-farnesene	2.63	0.33	*b*
9	1174	3-methylbutanol	*b*	*b*	0.25	67	1739	linalool oxide IV (*cis*, pyranoid)	0.17	0.30	2.57
10	1184	limonene	0.04	*b*	*b*						
11	1192	(*E*)-2-hexenal	0.32	*b*	0.09	68	1766	nerol	0.43	0.08	0.12
12	1215	2-pentylfuran	0.02	*b*	*b*	69	1774	α-damascone	0.38	*b*	*b*
13	1218	(*Z*)-4-heptenal	0.02	*b*	*b*	70	1777	(*E*)-2,(*E*)-4-decadienal	0.10	*b*	*b*
14	1231	pentanol	0.44	0.24	0.45	71	1788	2-phenylethyl acetate	0.38	*b*	*b*
15	1278	octanal	0.05	*b*	*b*	72	1792	(*E*)-β-damascenone	0.10	*b*	*b*
16	1292	perillen	0.55	*b*	*b*	73	1807	hexanoic acid	1.46	2.63	5.96
17	1296	(*Z*)-2-pentenol	0.12	*b*	*b*	74	1812	geraniol	0.61	0.61	3.61
18	1302	(*E*)-2-pentenol	0.63	0.59	0.16	75	1818	α-ionone	0.59	*b*	*b*
19	1308	2,5-dimethylpyrazine	0.04	*b*	*b*	76	1820	geranylacetone	0.59	0.15	*b*
20	1320	6-methyl-5-hepten-2-one	0.55	0.26	0.07	77	1833	benzyl alcohol	1.40	3.68	11.71
21	1332	hexanol	0.12	*b*	0.32	78	1836	*N*-ethylsuccinimide	*b*	0.22	0.20
22	1361	(*Z*)-3-hexanol	0.32	0.36	0.47	79	1850	2-phenylethyl isobutyrate[c]	0.03	*b*	*b*
23	1364	(*Z*)-3-hexenyl isobutyrate	0.05	*b*	*b*	80	1863	2-phenylethanol	5.67	17.21	17.28
24	1368	nonanal	0.14	*b*	*b*	81	1871	benzyl cyanide	2.74	3.61	*b*
25	1380	(*E*)-3-hexenol	*b*	*b*	0.04	82	1887	heptanoic acid	*b*	*b*	0.20
26	1386	(*E*)-2-hexenol	*b*	*b*	0.18	83	1889	β-ionone	1.12	*b*	*b*
27	1415	linalool oxide I (*trans*, furanoid)	0.45	0.48	7.09	84	1891	*cis*-jasmone	1.12	1.13	*b*
						85	1907	2,6-dimethyl-3,7-octadiene-2,6-diol	0.26	2.40	18.40
28	1418	1-octen-3-ol	0.10	*b*	0.03						
29	1429	(*Z*)-3-hexenyl butyrate	0.37	*b*	0.08	86	1915	2-phenylethyl butyrate[c]	0.54	0.13	*b*
30	1440	linalool oxide II (*cis*, furanoid)	0.39	0.38	5.56	87	1922	(*E*)-2-hexenoic acid	*b*	*b*	1.33
						88	1925	2-acetylpyrrole	*b*	0.64	*b*
31	1440	(*Z*)-3-hexenyl isovalerate	0.23	*b*	*b*	89	1927	2-phenylethyl isovalerate[c]	0.51	*b*	*b*
32	1448	(*Z*)-3-hexenyl 2-methylbutyrate	0.11	*b*	*b*	90	1954	5,6-epoxy-β-ionone	0.60	0.30	0.12
						91	1956	phenol	0.19	0.21	*b*
33	1453	2-ethylhexan-1-ol	0.07	*b*	*b*	92	1989	4-nonanolide	0.08	0.59	*b*
34	1456	(*E*)-2,(*E*)-4-heptadienal	0.37	0.30	0.09	93	1991	nerolidol	38.10	0.70	0.49
35	1463	2-acetylfuran	0.11	*b*	*b*	94	2013	octanoic acid	0.16	0.36	0.30
36	1482	benzaldehyde	0.77	1.08	0.56	95	2025	*o*-cresol	0.07	*b*	*b*
37	1484	(*E*)-3,(*Z*)-5-octadien-2-one	0.22	0.30	*b*	96	2028	hexyl benzoate	0.04	*b*	*b*
38	1509	(*E*)-2-nonenal	0.07	*b*	*b*	97	2031	*m,p*-cresol	0.07	*b*	*b*
39	1522	linalool	2.19	0.16	0.57	98	2057	2-phenylethyl benzoate[c]	1.83	2.29	*b*
40	1534	octanol	0.21	*b*	*b*	99	2065	(*Z*)-3-hexenyl benzoate	0.75	*b*	*b*
41	1539	(*E*)-3,(*E*)-5-octadien-2-one	0.27	*b*	*b*	100	2081	(*E*)-2-hexenyl benzoate	0.11	*b*	*b*
42	1555	(*E*)-2,(*Z*)-6-nonadienal	0.11	*b*	*b*	101	2087	bovolide	0.70	*b*	*b*
43	1561	6-methyl-(*E*)-3,5-heptadien-2-one	0.20	*b*	*b*	102	2096	nonanoic acid	0.14	0.21	0.20
						103	2104	2-phenylethyl hexanoate[c]	0.45	*b*	*b*
44	1566	1-ethyl-2-formylpyrrole	0.27	*b*	*b*	104	2128	theaspirone	0.29	0.84	*b*
45	1570	2,6,6-trimethyl-2-hydroxycyclohexanone	*b*	0.19	0.14	105	2134	4-vinylguaiacol	0.24	*b*	*b*
						106	2136	5-decanolide	0.41	1.57	*b*
46	1573	3,7-dimethyl-1,5,7-octatrien-3-ol	1.12	0.46	3.01	107	2157	jasmine lactone	1.83	26.43	0.30
						108	2159	methylethylmaleimide[c]	*b*	1.10	0.20
47	1573	4-butanolide	*b*	0.36	0.20	109	2163	decanoic acid	*b*	*b*	0.40
48	1581	hexyl hexanoate[c]	0.26	*b*	*b*	110	2251	dihydroactinidiolide	0.21	1.56	1.86
49	1593	β-cyclocitral	0.26	*b*	*b*	111	2260	methyl jasmonate	0.65	3.33	*b*
50	1595	phenylacetaldehyde	0.18	0.79	*b*	112	2294	*trans*-geranic acid	*b*	*b*	1.38
51	1596	safranal	0.40	*b*	*b*	113	2325	4-vinylphenol	0.26	0.73	*b*
52	1609	acetophenone	0.72	*b*	*b*	114	2365	indole	7.88	11.93	*b*
53	1626	(*Z*)-3-hexenyl hexanoate	0.70	0.10	*b*	115		2,6-dimethyl-1,7-octadiene-3,6-diol[e]			*d*
54	1631	isovaleric acid	0.33	*b*	0.21						
55	1636	(*E*)-2-hexenyl hexanoate	0.78	*b*	*b*	116		salicylic acid			*d*
56	1655	2,6,6-trimethylcyclohex-2-ene-1,4-dione	0.47	*b*	*b*	117		vanilline			*d*
						118		2-indolinone[c]			*d*
57	1660	4-hexanolide	0.76	1.62	0.25	119		homovanillic acid[c]			*d*
58	1662	α-terpineol	0.83	*b*	*b*	120		coniferyl alcohol[c]			*d*
59	1681	(*E*)-3-hexenyl (*Z*)-3-hexenoate	0.13	*b*		121		loliolide[c]			*d*
								total, %	96.87	95.62	94.40

[a] Kovats index on Carbowax 20M. [b] Not detected. [c] Newly identified compounds as tea aroma. [d] Identified on HP-5 column. [e] Tentatively identified.

Pom Fon oolong tea, also made from tea leaves injured with green flies, contained 10.14% 3,7-dimethyl-1,5,7-octatrien-3-ol (Takami et al., 1990). 2,6-Dimethyl-3,7-octadiene-2,6-diol seemed to be formed by an abnormal biosynthesis pathway caused by infestation of green flies. To see the relation between these two components, the ratio of *R* and *S* configuration of these compounds analyzed by using a CP-cyclodex column is shown in

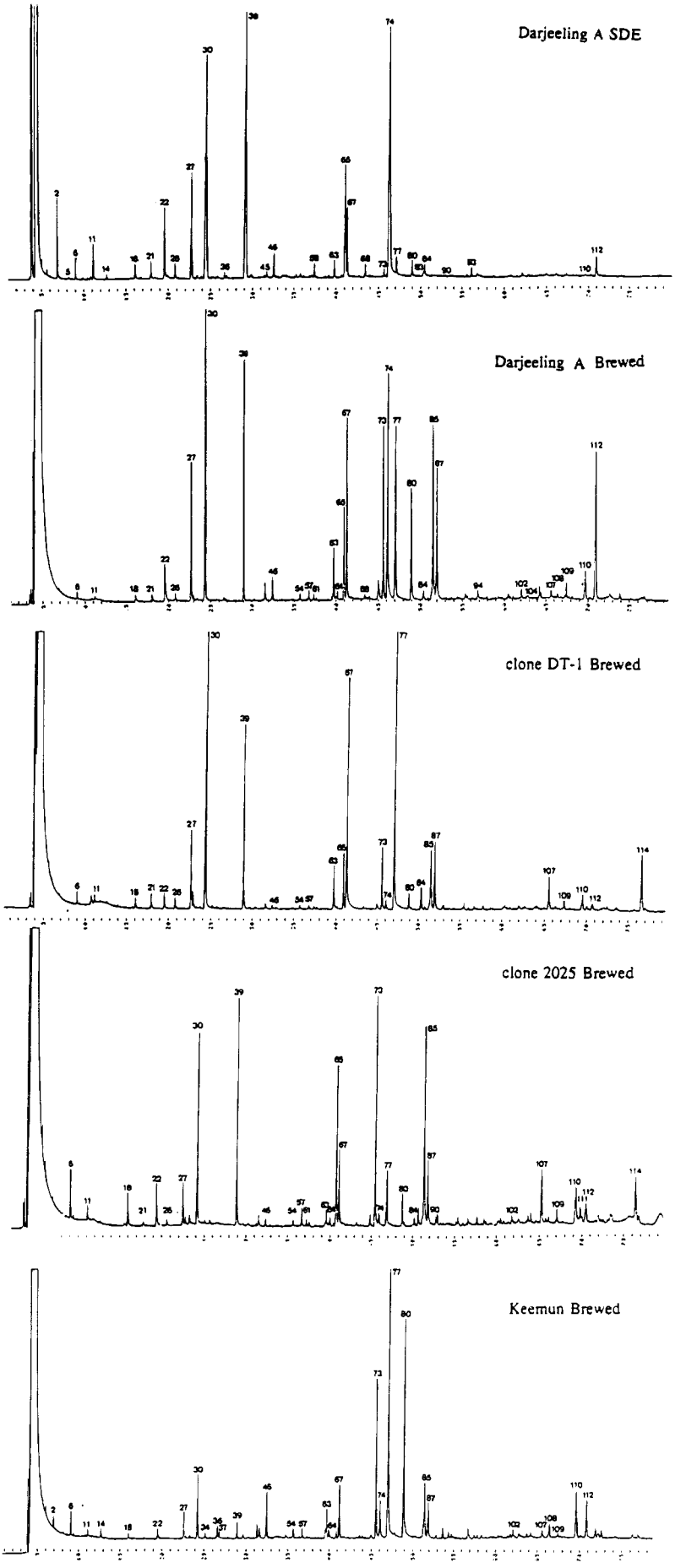

Figure 3. Gas chromatograms of black tea extracts.

Table 2. Composition of Aroma Extracts of Black Tea, Darjeeling, Clone DT-1 (C-DT-1), Clone 2025 (C-2025), and Keemun

			GC peak area, %								
			Darjeeling				C-DT-1		C-2025		Keemun
PN	KI[a]	compound	A-SDE	A-brewed	B-brewed	C-brewed	SDE	brewed	SDE	brewed	brewed
1	991	1-penten-3-one	0.12	b	b	b	0.24	b	0.58	b	b
2	1064	hexanal	1.63	0.05	0.10	0.05	2.42	0.08	4.88	0.07	0.37
5	1110	4-methyl-3-penten-2-one	0.06	b	b	b	0.10	b	0.17	b	b
5′	1114	(*Z*)-3-hexenal	b	b	b	b	0.11	b	0.79	b	b
6	1139	1-penten-3-ol	0.55	0.19	0.45	0.26	1.09	0.38	2.95	1.51	0.97
8	1160	heptanal	0.07	b	b	b	0.22	b	0.17	b	b
8′	1168	(*E*)-3-hexenal	0.07	b	b	b	0.21	b	0.39	b	b
9	1174	3-methylbutanol	0.17	b	b	b	0.16	0.54	b	b	b
11	1192	(*E*)-2-hexenal	1.11	0.13	0.64	0.57	4.56	0.35	10.59	0.69	1.00
12	1215	2-pentylfuran	0.06	b	b	b	0.35	b	0.21	b	b
14	1231	pentanol	0.22	b	b	b	0.71	0.16	0.53	b	0.56
16	1292	perillen	0.04	b	b	b	0.03	b	0.03	b	b
16′	1300	(*Z*)-3-hexenyl acetate	0.07	b	b	b	0.35	b	0.28	b	b
18	1302	(*E*)-2-pentenol	0.68	0.38	0.62	0.63	1.39	0.60	3.18	1.62	0.52
20	1320	6-methyl-5-hepten-2-one	0.15	b	b	b	0.15	b	0.10	b	b
21	1332	hexanol	0.80	0.42	0.48	0.62	0.69	0.82	0.61	0.47	0.12
22	1361	(*Z*)-3-hexanol	3.07	1.84	2.12	3.00	2.48	0.90	4.75	2.91	0.82
24	1368	nonanal	0.40	b	b	b	0.19	b	0.14	b	b
25	1380	(*E*)-3-hexenol	0.09	b	b	b	0.02	b	0.05	b	b
26	1386	(*E*)-2-hexenol	0.70	0.49	0.42	0.67	1.79	0.64	0.64	0.78	0.04
27	1415	linalool oxide I (*trans*, furanoid)	5.45	4.57	4.51	3.95	5.67	3.43	2.07	2.01	1.63
28	1418	1-octen-3-ol	0.34	b	b	b	0.21	b	0.37	b	b
28′	1420	furfural	b	b	b	b	b	b	b	0.63	0.25
28″	1428	(*Z*)-3-hexenyl butyrate	b	b	b	b	0.13	b	0.19	b	b
29′	1430	(*E*)-2,(*Z*)-4-heptadienal	0.24	b	b	b	0.39	b	0.45	b	b
30	1440	linalool oxide II (*cis*, furanoid)	18.35	16.52	16.66	14.47	24.70	20.01	7.87	8.95	3.81
33	1453	2-ethylhexan-1-ol	0.33	b	b	b	b	b	b	b	b
34	1456	(*E*)-2,(*E*)-4-heptadienal	b	0.03	0.05	0.09	0.41	0.10	0.80	0.27	0.12
36	1482	benzaldehyde	0.32	0.13	0.16	0.19	0.25	0.07	0.24	0.20	0.81
39	1522	linalool	20.35	7.24	6.94	5.78	24.46	7.43	21.24	9.56	0.97
40	1534	octanol	0.35	b	b	b	0.69	b	0.39	b	b
45	1570	2,6,6-trimethyl-2-hydroxycyclohexanone	0.11	b	b	b	0.15	b	0.23	b	b
46	1573	3,7-dimethyl-1,5,7-octatrien-3-ol	1.50	0.44	0.28	0.37	0.32	0.05	0.25	0.10	0.42
47	1573	4-butanolide	b	0.74	0.74	0.74	b	0.16	b	0.38	2.78
49	1593	β-cyclocitral	0.04	b	b	b	0.25	b	0.25	b	b
51	1596	safranal	0.15	b	b	b	0.24	b	0.95	b	b
53	1626	(*Z*)-3-hexenyl hexanoate	0.18	b	b	b	0.44	b	1.42	b	b
54	1631	isovaleric acid	b	0.28	0.35	0.37	b	0.35	b	0.67	0.77
54′	1632	nonanol	0.22	b	b	b	0.48	b	0.38	b	b
55	1636	(*E*)-2-hexenyl hexanoate	0.14	b	b	b	0.43	b	0.13	b	b
55′	1649	neral	0.14	b	b	b	0.05	b	0.04	b	b
57	1660	4-hexanolide	b	0.37	0.36	0.52	b	0.23	b	0.99	0.63
58	1662	α-terpineol	0.60	b	b	b	0.52	b	0.63	b	b
61	1689	valeric acid	b	0.22	0.20	0.21	b	0.16	b	0.39	0.19
62′	1693	geranial	0.10	b	b	b	0.02	b	b	b	b
63	1698	linalool oxide III (*trans*, pyranoid)	0.70	1.65	1.96	1.65	0.54	2.44	0.21	0.82	1.79
64	1700	2-hexen-4-olide	b	0.23	0.33	0.50	b	0.03	b	0.45	0.14
65	1727	methyl salicylate	6.65	2.93	3.10	3.16	6.80	2.40	14.43	7.72	0.39
67	1739	linalool oxide IV (*cis*, pyranoid)	3.30	7.26	6.70	6.46	4.47	13.58	2.35	5.38	3.46
68	1766	nerol	0.58	0.17	0.18	0.29	0.61	0.19	0.87	0.31	0.08
73	1807	hexanoic acid	0.33	5.38	4.89	6.26	0.41	2.64	1.18	10.68	9.12
74	1812	geraniol	23.15	9.75	9.46	9.34	1.59	0.58	1.84	0.64	3.10
76	1820	geranylacetone	0.26	b	b	b	b	b	b	b	b
77	1833	benzyl alcohol	1.87	5.39	5.65	5.35	1.32	14.31	0.32	2.47	14.79
78	1836	*N*-ethylsuccinimide	b	1.78	1.88	1.99	b	1.26	b	1.23	9.63
80	1863	2-phenylethanol	0.95	4.25	4.71	4.74	0.13	0.75	0.13	1.80	15.99
83	1889	β-ionone	0.22	b	b	b	b	b	0.65	b	b
84	1891	*cis*-jasmone	1.03	0.50	0.33	0.43	1.30	1.31	0.78	0.94	0.13
85	1907	2,6-dimethyl-3,7-octadiene-2,6-diol	0.11	2.96	3.15	3.29	0.06	1.08	0.14	1.04	1.82
85	1907	(*Z*)-3-hexenoic acid	b	3.29	3.43	4.55	b	2.80	b	9.40	2.03
87	1922	(*E*)-2-hexenoic acid	b	5.60	5.44	5.63	b	3.52	b	3.51	2.86
90	1954	5,6-epoxy-β-ionone	0.13	b	b	b	0.05	b	0.12	b	b
91	1956	phenol	b	b	b	0.54	b	b	b	b	0.69
93	1991	nerolidol	0.35	b	b	b	2.26	b	2.23	b	b
101	2087	bovolide	b	b	b	b	0.36	b	0.68	b	b
102	2096	nonanoic acid	b	0.45	0.47	0.41	b	0.70	b	0.39	0.60
104	2128	theaspirone	b	0.38	0.45	0.30	b	0.48	b	0.57	0.27
107	2157	jasmine lactone	b	0.45	0.45	0.74	b	2.39	b	3.35	0.69
108	2159	methylethylmaleimide	b	0.20	0.18	0.11	b	0.10	b	0.15	1.00
109	2163	decanoic acid	b	0.65	0.63	0.60	b	2.02	b	1.05	1.17
110	2251	dihydroactinidiolide	b	1.50	1.32	1.35	0.30	1.40	0.74	3.76	4.97
111	2260	methyl jasmonate	b	b	b	b	b	0.61	b	1.18	b
112	2294	*trans*-geranic acid	1.36	8.12	6.36	6.74	0.26	1.19	0.35	1.90	3.26
114	2365	indole	b	b	b	b	0.96	4.49	0.97	4.52	0.47
		total, %	99.71	97.16	96.15	96.92	98.49	96.73	96.95	95.31	95.23

[a] Kovats index on Carbowax 20M. [b] Not detected.

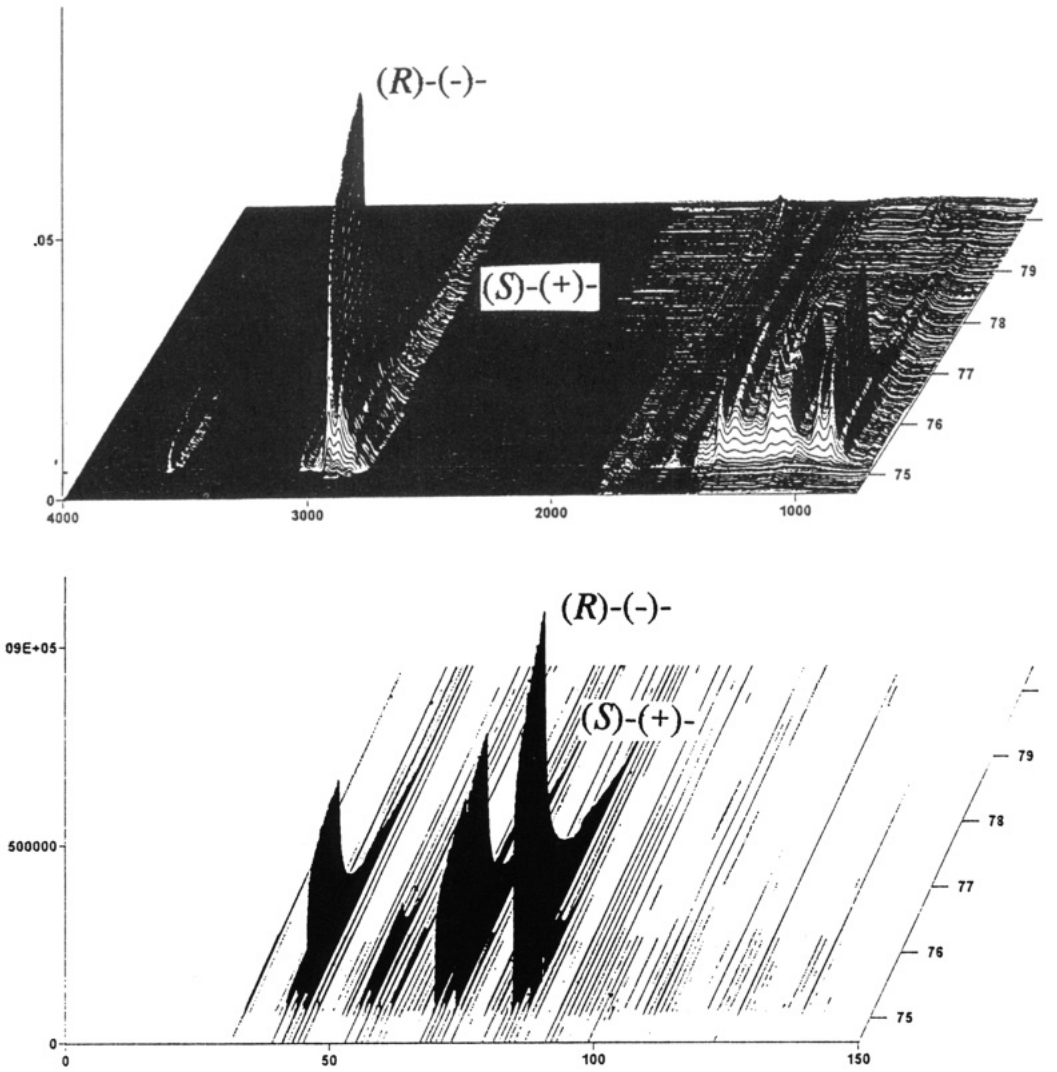

Figure 4. 3D IR and MS spectra of 2,6-dimethyl-3,7-octadiene-2,6-diol.

Table 3. *R* and *S* Ratio of Linalool, 3,7-Dimethyl-1,5,7-octatrien-3-ol, and 2,6-Dimethyl-3,7-octadiene-2,6-diol in Chan Pin Oolong and Darjeeling Teas

	Chan Pin oolong *R:S*	Darjeeling *R:S*
linalool	27.8:72.2	52.4:47.6
3,7-dimethyl-1,5,7-octatrien-3-ol	63.0:37.0	0:100
2,6-dimethyl-3,7-octadiene-2,6-diol	91.8:8.2	7.4:92.6

Table 3. In black tea flavor, only (*S*)-(+)-2,6-dimethyl-3,7-octadiene-2,6-diol (Etoh et al., 1980) and (*S*)-(+)-3,7-dimethyl-1,5,7-octatrien-3-ol (Nakatani et al., 1969) have been previously isolated, and both compounds were estimated to be produced from (S)-(+)-linalool (Etoh et al., 1980). Further work showed that 3,7-dimethyl-1,5,7-octatrien-3-ol has been produced *in vitro* from 2,6-dimethyl-3,7-octadiene-2,6-diol by dehydration at 120–130 °C (Hara, and Hotta, 1987). Chan Pin oolong tea mainly contains (*R*)-(−)-2,6-dimethyl-3,7-octadiene-2,6-diol and (*R*)-(−)-3,7-dimethyl-1,5,7-octatrien-3-ol, regardless of the *R*/*S* ratio of linalool, as shown in Figure 4. The result of Chan Pin oolong tea analysis supports the same dehydration pathway, from 2,6-dimethyl-3,7-octadiene-2,6-diol to 3,7-dimethyl-1,5,7-octatrien-3-ol. But the relation between 2,6-dimethyl-3,7-octadiene-2,6-diol and linalool is not clear, because (*R*)- and (*S*)-linalool are present in black and oolong teas (Wang et al., 1994).

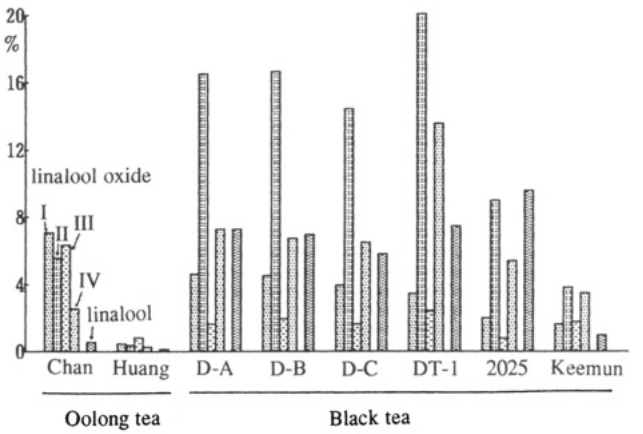

Figure 5. Comparison of the composition of linalool oxides and linalool between oolong tea and black tea: (Chan) Chan Pin oolong; (Huang) Huang Chin Kuei; (D-A) Darjeeling A; (D-B) Darjeeling B; (D-C) Darjeeling C; (DT-1) clone DT-1; (2025) clone 2025; Keemun.

The main components of the brewed Sri Lanka clone DT-1 extract were linalool oxide II, benzyl alcohol, linalool oxide IV, linalool, and indole. The aroma of Sri Lanka clone 2025 mainly consists of hexanoic acid, linalool, (*Z*)-3-hexenoic acid, linalool oxide II, methyl salicylate, and linalool oxide IV. These aroma patterns are also different from those of the SDE extracts, especially in the amount of terpene alcohols. The aroma

of Chinese Keemun contains large amounts of 2-phenylethanol, benzyl alcohol, *N*-ethylsaccinimide, hexanoic acid, and dihydroactinidiolide. This aroma pattern is different from that of the extracts, prepared by using a rotary evaporation apparatus (Aisaka et al., 1978).

Figure 5 shows the ratio of linalool and linalool oxides I, II, III, and IV. It is worth noting that oolong tea contains larger amounts of the *trans* form of linalool oxides such as I and III than of the *cis* form, while black tea contains larger amounts of the *cis* form such as II and IV than of the *trans* form. Terpene alcohols are produced mostly during the SDE process, and the quantity of linalool in tea aroma is not found to be as large as linalool oxides. Some new aroma components of tea were identified through this study by the use of the brewed extraction method. Using this method, further investigations on the various types of tea are now in progress.

ACKNOWLEDGMENT

We thank Dr. T. Yamanishi for valuable discussion, Dr. M. Iwamoto, Hasegawa Perfumery Co., Ltd., for supplying standard (*R*)-(−)- and (*S*)-(+)-2,6-dimethyl-3,7-octadiene-2,6-diol samples, and Dr. M. Hara, Mituinourin Co., Ltd., for supplying Chan Pin oolong.

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Received for review May 11, 1994. Accepted October 24, 1994.[⊗]

JF940237C

[⊗] Abstract published in *Advance ACS Abstracts,* December 1, 1994.