

AROMA CHARACTERISTICS OF DARJEELING TEA

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Summary

Darjeeling Teas cultivated in Darjeeling valley tea estates in India were analyzed. Aroma concentrates were prepared by using the brewed extraction method, and were analyzed by GC/MS. The common components of Darjeeling tea aroma were linalool oxide I, II, III, and IV, linalool, geraniol, methyl salicylate, benzyl alcohol, 2-phenylethanol, dihydroactinidiolide, hexanoic acid, (*Z*)-3-hexenoic acid, (*E*)-2-hexenoic acid, *trans*-geranic acid, 3,7-dimethyl-1,5,7-octatrien-3-ol, and 2,6-dimethyl-3,7-octadiene-2,6-diol. The last 2 components were the most important aroma components of Darjeeling tea, and were produced by green flies (*E. flavescens*), which infest tea shoots. The results of the sensory test correlate with the results of Cluster and principle component analyses performed using the main eleven components.

Keywords

Black tea Aroma, Tea flavor, Brewed extraction method, PCA analysis, Cluster analysis

Introduction

In a previous paper¹, we reported on the aroma composition of Darjeeling black tea prepared by the brewed extraction method. In this paper, the aroma concentrations of 35 kinds of Darjeeling tea were prepared by the brewed extraction method and analyzed by GC and GC-MS.

To investigate each aroma profile in more detail, Cluster and Principle Component Analyses were performed using the main eleven aroma components.

Materials and Methods

Thirty-five kinds of Darjeeling tea plucked at the Tea Estates in Darjeeling, West Bengal, India were conducted on Sensory test and GC-MS analysis. One hundred grams of a powdered tea sample was brewed in 1000 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 centrifuged and dried over anhydrous sodium sulfate for 12 hours. After the sodium sulfate was filtrated out, the solvent was removed by using a Kuderna-Danish evaporator. Each aroma concentrate was then analyzed by GC and GC-MS. A Hitachi Model G-3000 GC equipped with an FID and a 60 m x 0.25 mm HP-wax capillary column was used. The peak area was integrated with a Hitachi D-2500 integrator. A Hewlett-Packard 5972 MS interfaced with a 5890 GC was used for identification. The GC and GC-MS conditions were detailed in the previous paper¹.

Results and Discussion

Samples, D17, D18, D11 and D36 had the best Darjeeling tea aroma according to the sensory test. Each aroma concentrate from Darjeeling tea strongly resembles that of tea infusion. Gas chromatograms of one aroma concentrate (D17) having a nice Darjeeling flavor are shown in Figure 1. One-hundred twelve compounds were identified in Darjeeling tea aroma extracts. The aroma profile of samples having the characteristic nice and off flavors are shown in Figure 2, and the main components and relative quantities of each component as calculated by the peak area are listed in Table 1.

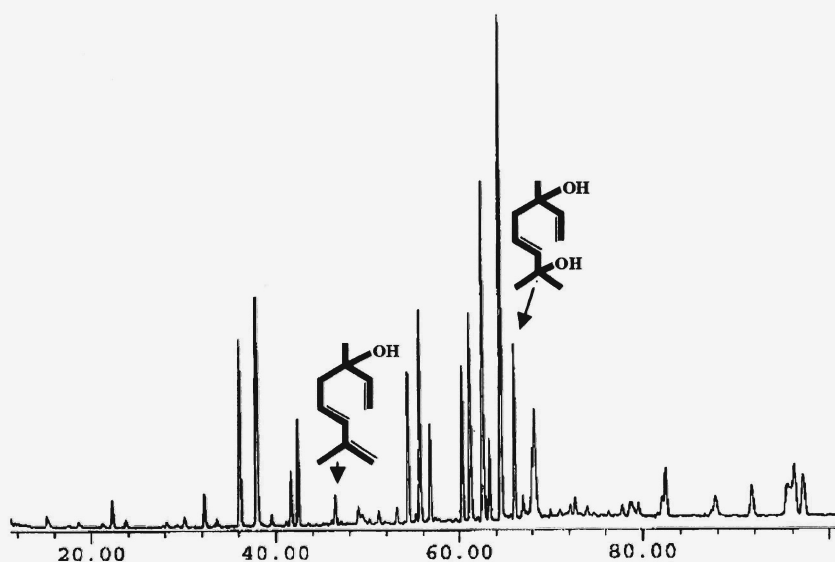


Figure 1 D17 Gas Chromatogram.

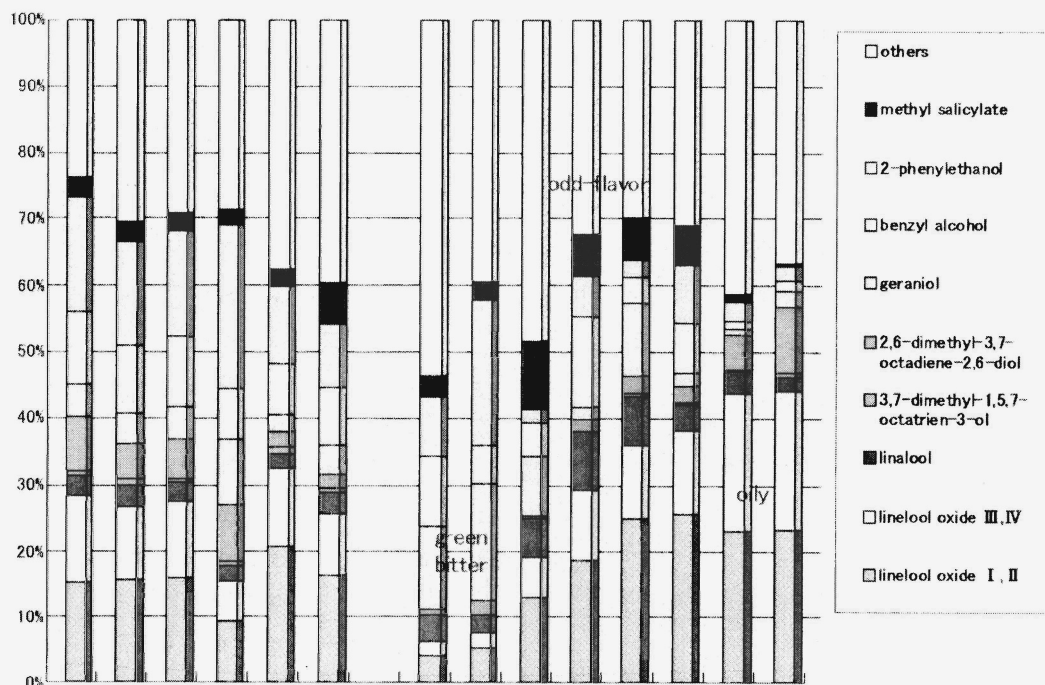


Figure 2 Aroma profiles of Darjeeling tea

Table 1. Composition of the aroma concentrates of Darjeeling Teas

K.I.	Compound	D11	D17	D18	D36	D29	D04	D10	D19	D20	D28	D33	D38
1007	3-methyl-2-butenal						0.01						
1024	methylbenzene	0.07		0.49	0.03	0.26	0.01		0.27	0.9	0.33	0.18	0.33
1089	hexanal	0.26	0.31	0.12	0.29	0.26	0.41	0.1	0.23	0.67	0.15	0.99	1.07
1141	(<i>E</i>)-2-pentenal						0.05						
1145	4-methyl-3-penten-2-one		0.05	0.01	0.03	0.03							0.08
1169	1-penten-3-ol	0.09	0.15	0.02	0.07	0.05	0.24	0.01	0.07	0.27	0.13	0.18	0.17
1205	limonene					0.05							
1219	1,8-cineole					0.03							
1224	3-methylbutanol		0.13										
1235	(<i>E</i>)-2-hexenal	0.51	0.78	0.42	0.5	0.55	0.4	0.28	0.56	1.27	0.63	0.85	0.86
1265	pentanol	0.07	0.2	0.01	0.12	0.13	0.2	0.01	0.05	0.32	0.04	0.36	0.28
1284	methylpyrazine				0.09		0.07		0.05				0.02
1310	3-hydroxy-2-butanone				0.03		0.01						
1325	1-hydroxy-2-propanone	0.02			0.03								
1335	(<i>E</i>)-2-pentanol		0.17	0.01	0.01	0.13	0.15			0.19	0.13	0.16	0.07
1340	2,5-dimethylpyrazine				0.17		0.27						
1345	2,6-dimethylpyrazine						0.04						
1347	2-heptanol							0.28					
1350	ethylpyrazine				0.04		0.08	0.03					
1362	2,3-dimethylpyrazine		0.03				0.03						
1362	4-hydroxy-4-methyl-2-pentanone						0.03						
1365	hexanol	0.14	0.3	0.03	0.3	0.26	0.07	0.14	0.09	0.16	0.23		0.01
1397	(<i>Z</i>)-3-hexenol	0.85	0.99	0.38	1.53	0.79	0.27	0.71	0.79	0.98	0.97	0.22	0.17
1406	2-ethyl-5-methylpyrazine						0.15						
1417	trimethylpyrazine						0.04						
1420	(<i>E</i>)-2-hexenol	0.07	0.25	0.09	0.22	0.47	0.11	0.07	0.07	0.1	0.35		0.01
1426	maleic anhydride						0.01						
1454	linalool oxide I	5.98	5.67	4.56	6.78	2.89	3.99	0.67	1.13	2.62	2.23	8.23	6.57
1479	(<i>E</i>)-2,(<i>Z</i>)-4-heptadienal												0.38
1485	linalool oxide II	10.1	10.1	10.8	14.1	6.45	11.2	3.47	4.23	16.2	10.9	15.2	16.6
1490	acetic acid												0.1
1507	(<i>E</i>)-2,(<i>E</i>)-4-heptadienal	0.14	0.33	0.24	0.32	0.5	0.67	0.18	0.27	0.63	0.25	0.5	1.45
1524	2-acetylfuran				0.03		0.04						
1534	(<i>E</i>)-3,(<i>Z</i>)-5-octadienone	0.05	0.15		0.1	0.11	0.36		0.11	0.21	0.12	0.23	0.28
1537	benzaldehyde	1.37	1.66	0.8	0.5	1.45	0.51	0.57	0.5	0.51	0.33	0.29	0.68
1561	linalool	2.9	3.18	3.07	2.16	2.16	3.68	3.99	2.7	8.56	5.66	2.08	3.3
1568	propionic acid						0.07	0.01					
1586	(<i>E</i>)-3,(<i>E</i>)-5-octadienone						0.13	0.03	0.07		0.01		
1586	5-methylfurfural				0.04								
1593	2,2-dimethylpropionic acid						0.04					0.08	

continued

K.I.	Compound	D11	D17	D18	D36	D29	D04	D10	D19	D20	D28	D33	D38
1601	6-methyl-(<i>E</i>)-3,5-heptadien-2-one						0.05	0.31	0.02				
1619	sulfinylbis methane	0.09	0.18	0.19	0.22	0.18	0.4	0.32	0.23	0.48	0.29	0.3	0.15
1623	2,6,6-trimethyl-2-hydroxycyclohexanone			0.07			0.31		0.45		0.21		0.18
1628	1-ethyl-2-formylpyrrole	0.07	0.15				0.33				0.08		
1633	3,7-dimethyl-1,5,7-octatrien-3-ol	0.56	0.86	0.65	1.24	0.79	0.33			0.16		0.62	0.26
1657	4-butanolide	0.53	0.51	0.26	0.89	1.05	0.14	0.52	0.68	0.6	0.54	0.15	0.07
1657	1-ethyl-2-acetylpyrrole						0.13						
1665	2-phenylacetaldehyde	0.17	0.5	0.7	0.13	0.53	0.23	0.03			0.01		0.83
1679	<i>N</i> -ethylacetamide						0.04						
1684	dihydro-4,4-dimethyl-2(3 <i>H</i>)-furanone	0.07	0.17	0.03	0.12		0.17	0.04	0.09		0.01	0.21	0.26
1690	neral							0.06					
1697	4-methyl-5-hexen-4-olide			0.03			0.16				0.01		0.02
1700	isovaleric acid	0.34	0.38	0.23	0.22	0.18	0.27	0.57	0.38	0.29	0.3		0.03
1717	α -terpineol		0.08	0.07		0.03	0.27	0.35			0.11		0.12
1733	4-hexanolide	0.48	0.5	0.28	0.53	0.34	0.13	0.42	0.5	0.63	0.45		0.07
1736	geranial							0.14					
1738	2-methyl-6,7-dihydro-5 <i>H</i> -cyclopentapyrazine						0.01						
1748	2-isopropylpyrrolidine						0.04						0.02
1754	naphthalene						0.43						
1763	linalool oxide III	4.91	4.48	4.42	3.92	2.89	3.56	0.24	0.68	1.43	0.75	8.69	6.61
1772	valeric acid	0.29	0.26	0.09	0.22		0.16	0.28	0.36	0.24	0.27		0.21
1779	1-propyl-2-formylpyrrole						0.08						
1789	linalool oxide IV	6.49	6.44	8.5	7.5	3.42	8.42	1.88	1.62	9.07	5.28	12	13.9
1796	methyl salicylate	2.9	2.86	3.2	2.63	2.37	3.09	2.97	2.7	6.18	10.4	0.62	1.11
1813	M82,43,111+4,5,5-trimethyl-2(5 <i>H</i>)-furanone	0.07	0.13	0.12	0.1		0.23					0.29	0.17
1824	nerol	0.07	0.1				0.15	0.21	0.16				
1830	(<i>E</i>)-2,(<i>Z</i>)-4-decadienal						0.06						
1830	5-hexanolide						0.03		0.16				
1830	1-phenylethanol							0.17					
1851	3-methyl-2,5-furandione							0.21					
1870	geraniol	4.78	4.65	4.77	2.41	9.86	3.46	12.6	17.5	1.74	9.05	2.46	1.02
1876	hexanoic acid	6.66	6.28	2.99	6.59	5.52	5.23	7.79	9.23	7.22	9.66	5.24	5.53
1890	3,3-dimethyl-2,7-octadione	0.05	0.15	0.05			0.09	0.17	0.23		0.13		0.07
1904	benzyl alcohol	10.8	10.4	10.9	7.72	7.37	1.73	10.5	5.74	13.7	5.01	1.46	1.22
1915	<i>N</i> -ethylsuccimide	3.07	2.4	1.55	2.25	2.5	12.9	3.26	2.93	2.22	4.84	6.47	10.2
1935	2-phenylethanol	15.7	15.5	17.1	11.7	24.7	7.46	9.06	22	6.18	1.93	1.99	2.94
1946	sulfonylbis methane				0.1		0.16			0.1	0.08	0.49	0.79
1948	benzylcyanide	0.03	0.1	0.1			0.21		0.05		0.02		
1948	b-ionone		0.05	0.02			0.09		0.05		0.03		0.01
1957	<i>cis</i> -jasnone				0.09		0.6	0.24	0.23	0.24	0.19	0.31	
1969	2-ethylhexanoic acid												

continued

K.I.	Compound	D11	D17	D18	D36	D29	D04	D10	D19	D20	D28	D33	D38
1971	2,6-dimethyl-3,7-octadiene-2,6-diol	5.81	5.28	8.21	2.26	8.42	1.6	0.96	2.25	1.82	0.49	9.99	5.15
1979	heptanoic acid	0.77	0.66	0.16	0.29			1.84	2.48	0.56	0.95		0.5
1983	methylmaltol						1.04						1.24
1983	maltol	0.68		0.52	2.16	0.39	0.77	2.62	0.34		0.38	1.69	0.5
1983	2-acetylpyrrole				1.64	0.11	3.2		0.18			0.92	0.96
1988	(Z)-3-hexenoic acid	1.88	1.99	1.08	1.88	1.58	0.42	3.83	2.03	3.33	3.68		0.38
1990	(E)-2-hexenoic acid	3.24	3.23	3.88	3.14	5	1.23	8.64	3.83	2.17	7.49	0.81	0.81
2001	5,6-epoxy-b-ionone	0.17				0.05	0.27		0.38				0.02
2033	3,7-dimethyl-1-octen-3,7-diol	0.07		0.21			0.2						0.12
2042	2-formylpyrrole	0.12	0.17	0.12	0.53	0.05	0.03	0.35			0.13	0.38	
2063	octanoic acid	0.51	0.51	0.23	0.43	0.32	0.8	1.16	1.24	0.63	0.89	0.51	0.5
2068	4-nonanolide?		0.31	0.02	0.07		0.2				0.07		
2108	5-methyl-2-formylpyrrole			0.24	0.22		0.27						
2123	3,7-dimethyl-1,7-octadiene-3,6-diol	0.05	0.17	0.19	0.25		0.23			0.48	0.14	0.49	0.18
2136	2-phenoxyethanol						0.48						
2152	nonanoic acid	0.31	0.36	0.19	0.09		0.31	1.42	1.58	0.63	0.87	0.46	0.21
2159	theaspirone				0.03		0.2			0.08			0.17
2167	dihydrobovolide						0.09		0.23		0.11		
2171	(E)-2-octenoic acid						0.13						
2177	5-decanolide	0.43			0.17					0.16		0.31	
2177	3-methoxy-4-acetyl-2(5H)-furanone						0.91						
2185	1,3-dimethyl-2,4,5-trioxoimidalidine		0.05	0.09			0.27				0.16		0.31
2187	1,3,5-trimethyl-1,3,5-triazine- 2,4,6(1H,3H,5H)-trione				0.17		0.2	0.71			0.3		0.94
2241	jasmine lactone			0.1	0.22		0.17	0.5		0.24	0.38	1.72	0.18
2251	3-ethyl-4-methyl-1H-pyrrole-2,5-dione	0.26	0.18	0.09	0.2		0.17	0.71	0.29	0.16	0.27	0.46	0.45
2301	2,6-dimethyl-2,7-octadiene-1,6-diol	0.77	0.93	0.56	0.58		1.2	1.27	0.34	0.63	0.38	0.69	1.57
2312	methyl jasmonate			0.16									
2327	trans-geranic acid	0.85	0.94	2.2	1.3	1.84	3.46	9.03	6.41	1.98	8.48	2	1.29
2336	dihydroactinidiolide	1.28	0.4	0.73	0.14	1.32	2	1.98	1.13	1.43	2.12	1.54	1.57
Total		97.9	96.7	96.5	91.7	97.4	94.4	97.8	99.9	98.4	99.4	92.8	95.3

The aroma concentrates from Darjeeling tea primarily consist of high levels of linalool oxides I, II, III, IV, linalool, geraniol, benzyl alcohol, 2-phenylethanol, methyl salicylate, hexanoic acid, (Z)-3-hexenoic acid, (E)-2-hexenoic acid, *t*-geranic acid, dihydroactinidiolide, *N*-ethylsuccinimide, 2,6-dimethyl-3,7-octadiene-2,6-diol, and 3,7-dimethyl-1,5,7-octatrien-3-ol. The last two components have apple-Muscat grape like flavor and fresh greenish aroma, and contribute to Darjeeling Muscat grape flavor. 3,7-Dimethyl-1-octen-3,7-diol, 3,7-dimethyl-1,7-octadiene-3,6-diol and 2,6-dimethyl-2,7-octadiene-1,6-diol were newly identified in this study. A series of these terpene-diol compounds shown in Figure 3 are characteristic in Darjeeling tea. Samples such as D17, D18, D36, and D11 having a nice Darjeeling flavor contained a high amount of these terpene-diol compounds. Taiwanese

red-oolong teas (Chan Pin Oolong and Pom Fon Oolong) also contains some of them^{1), 2)}. These compounds are probably produced from tea leaves infested with green flies (*E. flavescense*). The terpene-diol compounds are formed as an anti-insect by abnormal biosynthesis pathway, and these are easily dehydrated to mono-terpene alcohol by heating such as a firing process^{3, 4}. Terpene-diol compounds are also components of Muscat grapes, and they have been reported as the thermal degradation in grape juices⁵.

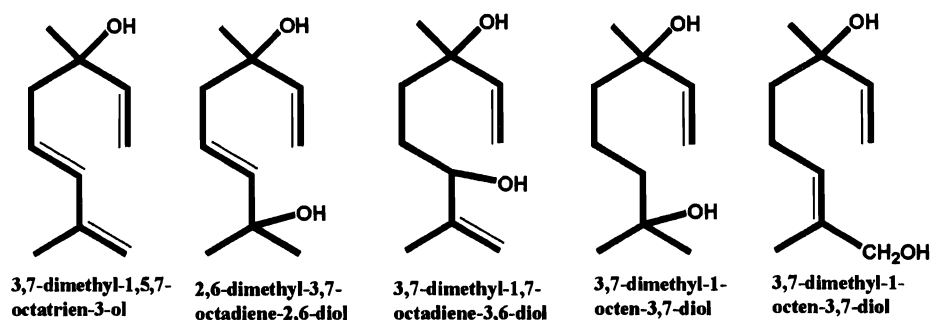


Figure 3 Characteristic components in Darjeeling tea aroma.

The results of Cluster analysis and PCA analysis using 11 main aroma components were shown in Figure 4 and Figure 5. The cluster table was divided two clusters, and both clusters contain high quality Darjeeling samples.

As the result of PCA analysis, compounds with a similar structure such as 2,6-dimethyl-3,7-octadiene-2,6-diol and 3,7-dimethyl-1,5,7-octatrien-3-ol have close vectors. Linalool oxide I +II and linalool oxide III+IV also have close vector. Alcohol, such as linalool, linalool oxides, (Z)-3-hexenol, geraniol, benzyl alcohol, 2-phenylethanol and methyl salicylate, were known to exist as glycosides in fresh tea leaves^{6, 7, 8}. Each compound has its own vector. The samples in a circle on the PCA Table had a nice Darjeeling flavor according to the sensory test. The PCA analysis result agree with the sensory test result. The results show that the importance for Darjeeling tea aroma is the balance among the main components, especially 2,6-dimethyl-3,7-octadiene-2,6-diol and 3,7-dimethyl-1,5,7-octatrien-3-ol.

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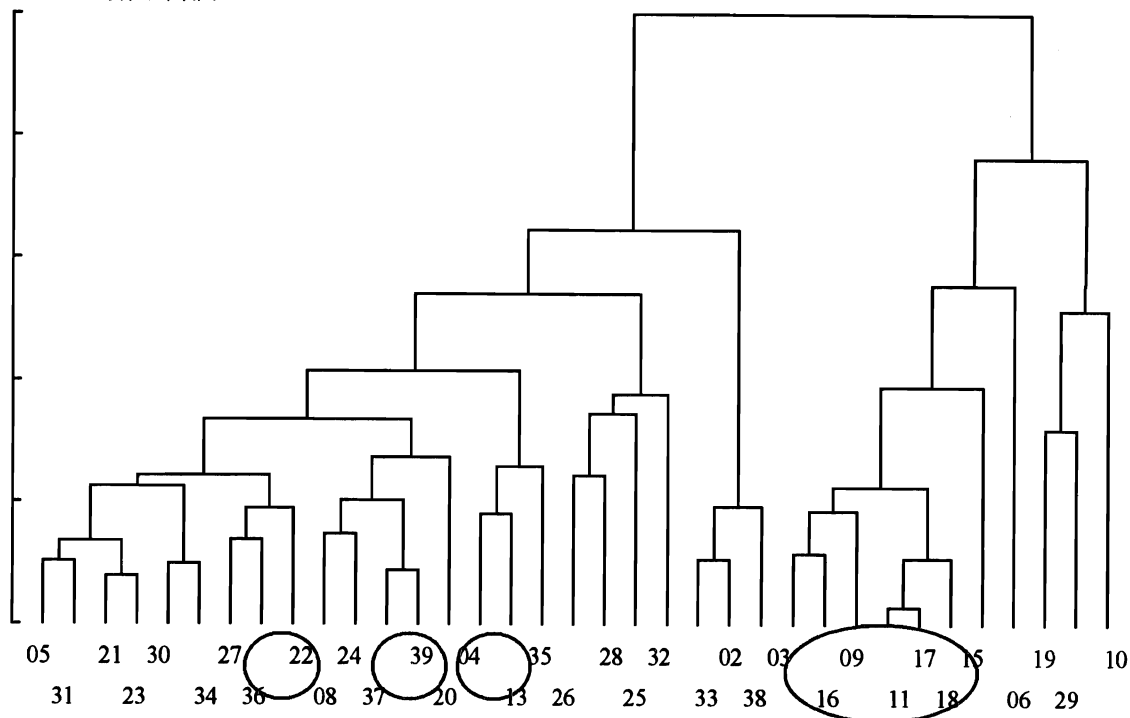


Figure 4. Cluster Analysis of Darjeeling Teas

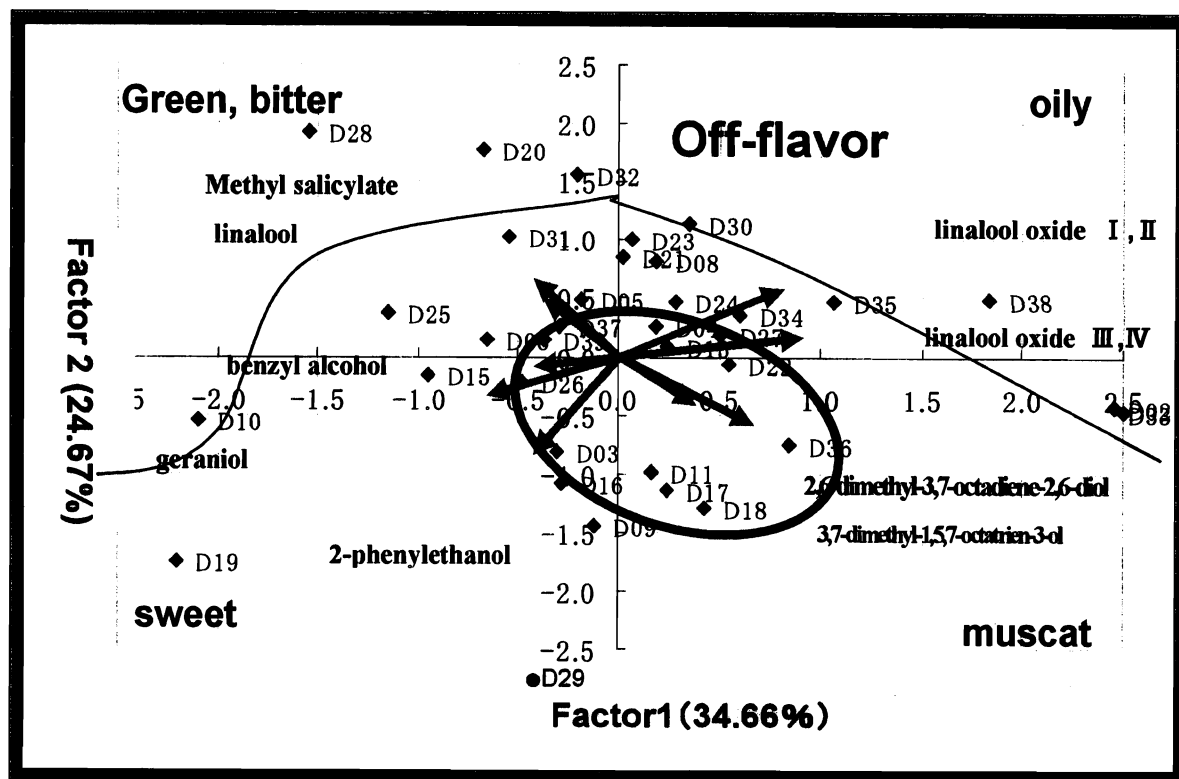


Figure 5. PCA analysis of Darjeeling tea.