Variation of Major Volatile Constituents in Various Green Teas from Southeast Asia

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A total of 15 green tea samples were prepared from fresh tea leaves obtained from three different countries: two from Laos, seven from Myanmar, and six from Vietnam. The volatile aroma constituents of the 15 samples were analyzed by gas chromatography/mass spectroscopy. Eleven aroma constituents were chosen from over 100 chemicals found in the samples to compare differences among various teas. They were hexanal, 1-penten-3-ol, heptanal, 1-pentenal, (*Z*)-2-penten-1-ol, (*Z*)-3-penten-1-ol, linalool oxide (*trans*-furanoid), linalool oxide (*cis*-furanoid), linalool, linalyl propanoate, and geraniol. Generally, concentrations of linalool and hexanal seem to play an important role in the quality of green teas. Green teas from Laos and Myanmar contained heterocyclic compounds, such as pyridines and pyrazines, formed by high-temperature processing. The presence of these heterocyclic compounds suggested that the temperature used for tea processing plays an important role in the formation of aroma chemicals in green teas.

Keywords: *aroma chemicals, geraniol, green teas, hexanal, linalool*

INTRODUCTION

Tea is one of the most popular beverages in the world. It is a member of the camellia evergreen tree species and is related to *Camellia* and *Sasanqua*. The first records of drinking tea date back to 59 BC in China (*1*). Tea was imported into Japan around 815 AD, and green tea began to be produced around the same time. The British East India Company brought tea to India over 200 years ago, and black tea began to be produced there. The same tea leaf, *Camillia sinensis,* with different degrees of fermentation, resulted in different black teas.

Typical teas are green tea, oolong tea, and black tea. Among these three teas, black tea is the most widely consumed in the world. Green tea is popular in the Southeast Asian countries. The origin of green tea is the southwestern plateau region of China and golden delta area located partially in China, Vietnam, and Myanmar (Burma). Green teas differ depending on the type of processing—steamed and pan-fired. Generally, green tea is steamed in Japan and pan-fired in the other Southeast Asian countries. Green tea is light yellowish-green when it is brewed. High-quality green tea has a slight greenish flavor given by certain terpenes such as linalool. There have been many reports on the aroma constituents of green teas from China and Japan (*2*). However, even though countries in Southeast Asia, including Laos, Myanmar, and Vietnam, are major green tea producing countries, there are only a few reports on the aroma investigation of green teas from Southeast Asia.

In the present study, aroma constituents of various pan-fired green teas obtained from Southeast Asia were analyzed to investigate their characteristic flavors.

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EXPERIMENTAL PROCEDURES

Chemicals. Authentic compounds were purchased from Aldrich Chemical Co. (Milwaukee, WI); Tokyo Kasei Organic Chemicals (Tokyo, Japan); Wako Pure Chemical Industries, Ltd. (Osaka, Japan); and Fluka Chemical Co. (Ronkonkoma, NY) or obtained from TAKATA KORYO Co., Ltd. (Osaka, Japan) as a gift.

Pan-Fired Green Teas. A total of 15 pan-fired green teas were prepared from fresh tea leaves taken from tea plants in Laos, Myanmar, and Vietnam. New leaves picked from tea trees were fired/heated in a drum-shaped container. After the heated leaves were rolled by hand, they were dried in sunlight or in a dryer. Tea leaf processing in the above countries is a type of household industry that has been manufacturing teas using a consistent method for over 100 years. Therefore, their products are also very consistent. The sources of pan-fired green teas are shown in Table 1.

Sample Preparations. Processed tea leaves were homogenized using a blender for 1 min. The homogenized tea sample (50 g) was placed in a 3 L round-bottom flask with 1 L of preboiled distilled water. The steam distillates were extracted with 100 mL of dichloromethane, containing 100 ppm of ethyl decanoate as an internal standard (*3*), simultaneously for 60 min by using a modified Likens−Nickerson apparatus (*4*). The extracts were dried over anhydrous sodium sulfate for 12 h. After the sodium sulfate was filtered out, the solvent was removed using a Kuderna−Danish evaporative concentrator. The solvent was further reduced under a purified nitrogen stream until the extract volume was 400 μL. Samples prepared were analyzed by gas chromatography (GC) and gas chromatography/mass spectrometry (GC/MS).

Analysis of the Volatile Constituents of Tea Samples. The GC Kovats retention index *I* (*5*) and the MS fragmentation pattern of each component were compared to those of the authentic compound to identify the volatiles in the samples. A Hewlett-Packard (HP) MS ChemStation Data system was also used to confirm MS identification of the GC components. A HP 5890A Series II GC equipped with a 30 m × 0.25 mm i.d. (Df = 0.1 μm) DB-WAX bonded phase, fused-silica capillary column (J & W Scientific, Folsom, CA), and a flame ionization detector (FID) was used. The linear velocity of helium carrier gas was 30 cm/s. The injector and the detector temperatures

10.1021/jf001321x CCC: $20.00
Published on Web 02/21/2001

Table 1. Sources of Pan-Fired Green Teas

no.	country	location	source	type	year obtained[a]
1.	Laos	South Lao Pakse	tea plant	high quality	1995
2.	Laos	South Lao Pakse	tea plant	low quality	1995
3.	Myanmar	Namhsan	tea plant		1994
4.	Myanmar	Tha Moe Ngay	tea plant	100	1998
5.	Myanmar	Tha Moe Ngay	tea plant	200	1998
6.	Myanmar	Tha Moe Ngay	tea plant	300	1998
7.	Myanmar	Tha Moe Ngay	tea plant	400	1998
8.	Myanmar	Naung Lang	tea plant		1998
9.	Myanmar	Kyaukme	tea retail store		1998
10.	Vietnam	Tuyen Quang	tea retail store		1996
11.	Vietnam	Phong Tho	city market		1996
12.	Vietnam	Lang Son	city market		1996
13.	Vietnam	Sapa	manufacturer		1997
14.	Vietnam	Yen Bai	city market		1997
15.	Vietnam	Hoa Bin	tea plant		1998

[a] Tea leaves were picked and processed within the year shown.

Table 2. Major Volatile Chemicals Identified and Their Relative Amounts in Green Teas from Laos

compound	*I*[a]	high quality	low quality
hexanal	1076	0.470[b]	0.601
1-penten-3-ol	1327	trace	0.150
heptanal	1167	0.204	0.282
1-pentenal	1124	0.216	0.250
(*Z*)-2-penten-1-ol	1327	0.730	0.350
(*Z*)-3-hexen-1-ol	1385	0.201	0.235
linalool oxide (*trans*-furanoid)	1439	0.246	0.695
linalool oxide (*cis*-furanoid)	1466	0.271	1.436
linalool	1554	4.065	3.461
linalyl propanoate	1690	0.906	0.554
geraniol	1847	0.845	0.372

[a] Kovats Index on DB-WAX. [b] Values are GC peak amount of compound/GC peak amount of internal standard.

were 250 °C at the split ratio of 1/20. The oven temperature was held at 40 °C for 5 min and then programmed to 180 °C at 2 °C/min and held for 20 min.

A HP 5890A Series II GC interfaced to a HP 5972 mass-selective detector was used for mass spectral identification of the GC components at an MS ionization voltage of 70 eV. Column and oven conditions were as stated above.

RESULTS AND DISCUSSION

Table 2 shows the major volatile chemicals identified in green teas from Laos. High-quality tea generally contained more volatile chemicals—in particular terpenes—in the present study. For example, the relative level of linalool is higher in high-quality tea (4.065) than in low-quality tea (3.461). Relative levels of linalyl propanoate and geraniol are also higher in high-quality tea than in low-quality tea. Some difference between high- and low-quality teas was observed. For example, an appreciable amount of (*E*)-2-hexenal (0.474) was found in low-quality tea but not in high-quality tea. (*E*)-2-Hexenal is known to possess a strong green note (*6*). The presence of pyridines and pyrazines—which are known to possess considerably low odor threshold and a toasted flavor (*7*)—was recognized in all green teas from Laos by GC/MS. They were 2-methylpyridine, 2,6-

Table 3. Major Volatile Chemicals Identified and Their Relative Amounts in Green Teas from Myanmar

compound	Namhsan	Tha Moe Ngay 100	Tha Moe Ngay 200	Tha Moe Ngay 300	Tha Moe Ngay 400	Naung Lang	Kyaukme
hexanal	0.718[a]	1.025	0.158	0.391	0.601	0.263	0.417
1-penten-3-ol	0.265	0.196	0.096	0.167	0.236	0.378	0.260
heptanal	0.306	trace	trace	trace	trace	trace	0.182
1-pentenal	0.165	trace	trace	trace	trace	trace	trace
(*Z*)-2-penten-1-ol	0.221	0.156	trace	0.112	trace	trace	0.171
(*Z*)-3-hexen-1-ol	0.222	0.115	0.158	0.373	0.451	trace	0.267
linalool oxide (*trans*-furanoid)	0.138	0.231	0.153	0.159	trace	0.199	0.278
linalool oxide (*cis*-furanoid)	0.162	0.420	0.337	0.363	trace	trace	0.122
linalool	9.299	6.565	2.634	5.046	0.415	4.627	8.559
linalyl propanoate	2.506	trace	0.514	0.887	1.016	0.983	trace
geraniol	2.000	0.860	0.506	0.912	1.450	1.004	1.781

[a] Values are GC peak amount of compound/GC peak amount of internal standard.

Table 4. Major Volatile Chemicals Identified and Their Relative Amounts in Green Teas from Vietnam

compound	Tuyen Quang	Phong Tho	Lang Son	Sapa	Yen Bai	Hoa Bin
hexanal	1.411[a]	1.743	2.055	0.365	0.256	0.087
1-penten-3-ol	0.207	0.311	0.391	0.167	0.193	0.157
heptanal	0.722	1.019	0.982	0.187	0.375	0.085
1-pentenal	0.365	0.345	0.339	0.184	0.209	0.184
(*Z*)-2-penten-1-ol	0.717	1.106	1.098	0.198	0.481	0.091
(*Z*)-3-hexen-1-ol	0.214	trace	0.359	trace	0.125	trace
linalool oxide (*trans*-furanoid)	trace	trace	0.148	0.108	trace	trace
linalool oxide (*cis*-furanoid)	0.217	trace	0.513	0.148	0.239	trace
linalool	5.930	2.384	4.561	3.055	2.594	2.180
linalyl propanoate	1.112	0.512	0.864	0.666	0.480	0.419
geraniol	1.124	0.260	0.386	0.617	0.459	0.410

[a] Values are GC peak amount of compound/GC peak amount of internal standard.

dimethylpyridine, 3-methylpyridine, 4-methylpyridine, methylpyrazine, 2,3-dimethylpyrazine, 2,5-dimethylpyrazine, trimethylpyrazine, and tetramethylpyrazine. Because the teas from Laos were treated by high temperature during a pan-firing process, pyridines and pyrazines were formed (*8*). Heterocyclic compounds including pyridines and pyrazines have been reported in many browning model systems consisting of a sugar and an amino acid (*9*).

Table 3 shows the major volatile chemicals identified in green teas from different areas in Myanmar. Among Tha Moe Ngay teas, the relative amount of linalool was much higher in high-quality tea (Tha Moe Ngay 100, 6.565) than in low-quality tea (Tha Moe Ngay 400, 0.415). On the other hand, low-quality teas contained relatively high levels of geraniol. Tea from Kyaulme contained the highest level of geraniol. Hexanal content was highest in Tha Moe Ngay 100, which is classified as a high-quality tea. It is difficult to rationalize clearly the relationship between quality and aroma constituents of tea. Generally, hexanal and linalool give a pleasant greenish note to green teas. Because Tha Moe Ngay 100 and Tha Moe Ngay 400 were prepared from tea trees grown in the same area, quality differences may be due to different picking months in 1998. A black tea from Myanmar (Burma) also contained high levels of linalool and linalool oxide, but the hexanal content in the same tea was somewhat low (*10*). Only one heterocyclic compound (2-methylpyridine) was detected in the green teas from Myanmar.

Table 4 shows the major volatile chemicals identified in green teas from different areas in Vietnam. Teas from Tuyen Quang and Lang Son are well-known as high-quality teas. As was the case with teas from Laos and Myanmar, high-quality teas from Vietnam contained relatively high levels of hexanal and linalool. Vietnamese green tea processed by pan-firing was also previously reported to contain a high level of linalool (*11*). Tuyen Quang tea contained the highest level of geraniol among the Vietnamese green teas examined. High levels of geraniol were also found in a Chinese green tea “Hwang Shan Mao Fong” (*12*, *13*).

As mentioned above, tea is one of the most popular beverages in the world. In particular, green teas are widely consumed in Southeast Asia. There are different varieties of green teas, but there have been only a few articles reporting aroma differences among them. Aromas of green teas vary significantly among different sources. Several aroma chemicals were shown to contribute to the quality of green tea in the present study. Generally, concentrations of linalool and hexanal seem to play an important role in the quality of green teas.

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Received for review November 7, 2000. Revised manuscript received January 22, 2001. Accepted January 24, 2001.

JF001321X