

DIFFERENTIATION OF BLACK TEAS BY VOLATILE PROFILE ANALYSIS USING HS-SPME/GC-MS

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ABSTRACT

A total of fourteen different commercial brands of black teas including Orthodox (OTD) and Crush Tear Curl (CTC) teas were collected at different markets with origin from 07 countries, and their volatile flavour compounds (VFC) were compared by analyses using headspace solid-phase micro-extraction coupled with gas chromatography - mass spectrometry (HS-SPME/GC-MS). Among the **60 volatiles** identified, terpenes (10.28 % - 59.88 %), ketones (1.85 % - 22.02 %) and esters (3.13 % - 418 %) represented the most abundant compounds. The results showed that CTC teas have a lower number of volatile components detected, lower contents of aldehydes and alcohols, and higher content of esters than OTD teas. Both Orthodox and CTC teas could be classified using principal component analysis (PCA) and cluster analysis of their volatile profiles. The data presented in this studied also suggest that Terpene Index might be used as an additional mean for the determination of the geographical origin of teas.

Keywords: black tea, volatile compounds, geographic origin.

1. INTRODUCTION

Normally, the quality of black tea is determined by human sensory evaluation based on “shape, colour, aroma and taste”. Among these characteristics, aroma is an essential criterion in the evaluation of sensory scores and the commercial description of black tea. Besides the conventional sensory evaluation of aroma quality, gas chromatography-olfactometry (GC-O) and aroma extract dilution analysis (AEDA) are also commonly applied to odour description and the determination of potent odorants in tea products [1, 2]. Unfortunately, these methods all rely upon highly trained personnel, and are likely affected by individual and subjective factors, such as age, emotion, and preference. Recently, several attempts have been made towards an objective discrimination and quality evaluation of black tea by GC-mass spectrometry (GC-MS) along with different techniques for the extraction of volatiles compounds, e.g. the simultaneous distillation-extraction (SDE) [3] and the dynamic headspace solid phase (DHS) techniques [4]. However, the experimental procedures presented in these extraction methods were time-consuming or involved complex samples pre-treatment. Headspace solid phase micro-extraction

(HS-SPME) has been proven to be a fast, simple, and convenient method for the analysis of volatile compounds in teas [5, 6]. This technique has also been successfully applied to the quality assessment and authentication of many fruits and other products, such as apple, strawberries, tomatoes, olive oils and green teas [5, 7]. Therefore, it would be interesting to investigate the feasibility of this method as a tool to authenticate and/or discriminate the black tea products. Since the aroma is one of the most typical features of the food, the characterization of aroma profile can represent a useful tool to evaluate the organoleptic quality and it could be used to guarantee its authenticity [8, 9]. Really, the aromatic profile represents a chemical “fingerprint” of the product, since the nature and the relative amount of the compounds present in the volatile fraction are distinctive features of the product.

On the basis of these remarks, this study was aimed at the characterization of the volatile fraction of black tea with the objective to differentiate this product in term of geographic origin. For this purpose, the HS-SMPE technique coupled to GC-MS was used. Secondly, the PCA was applied in order to detect the volatile compounds able to differentiate the fourteen different commercial brands of black tea from different regions investigated as well as kind of products (CTC and Orthodox black teas). This chemometric approach has been widely reported in literature to classification problems involving the authentication of food stuffs [10]. Then, the Terpene Index was calculated in order to differentiate the various geographic regions of black teas investigated.

2. MATERIALS AND METHODS

2.1. Materials

A total of fourteen different commercial brands of black teas were collected at different markets with origin from 7 Tea-producing countries and regions:

- Vietnam (VN) : 02 brands (Phuquy-Orthodox and Lipton-CTC);
- Russia (RUS): 02 brands (Russisch Westchiff-Orthodox and Russisch Teekanne-CTC);
- Sri-Lanka (SRI): 03 brands (English Breakfast-Orthodox; English Breakfast Teekanne-CTC and English Breakfast AHMAD-CTC);
- Indonesia (INDO): 02 brands (Heritage-Orthodox and SariWangi-Orthodox);
- Kenya (KEN): 02 brands (Nero Teekanne-CTC and Gold Teekanne-Orthodox);
- China (CHI): 01 brand (Puerh-Orthodox);
- India (IND): 02 brand (Darjeeling-Orthodox and Ceylon-CTC).

The black tea samples were collected in 2013 and stored at ambient temperature for further study.

2.2. Sample Preparations

One gram of black tea samples were infused with distilled water (5 ml) in a 10-ml glass septum vial by heating on a hot plate for 10 min. After the equilibration, commercially available SPME fibre (Supelco, Bellefonte PA, USA) coated with 65 μm polydimethylsiloxane/divinylbenzene (PDMS/DVB) was rapidly inserted into the headspace of the vial. The absorption step was kept at 90 °C for 30 min. The PDMS/DVB fibre was preconditioned for 5 min in the injection port of the GC at 220 °C before each analysis. Sample analyses are carried out in duplicates [5].

2.3. GC-MS analysis

A Thermo trace GC Ultra gas chromatograph coupled with the DSQ II mass spectrometer was used to perform the aroma analysis on HP-5 capillary column (30 m × 0.25 mm × 0.25 μm) with purified helium as the carrier gas at a constant flow rate of 1 ml min⁻¹. After extraction, the fibre was desorbed in the injector port of the GC at 220 °C for 5 min. The oven temperature was held at 50 °C for 3 min and then increased to 190 °C at a rate of 5 °C min⁻¹ and held at 190 °C for 1 min, and then increased to 240 °C at a rate 20 °C min⁻¹, held at this temp. for 3 min. Ion source temperature was at 200 °C and spectra was produced in the electron impact (EI) mode at 70 eV. The mass spectrometer was operated in the full scan, and the peak area was determined by Xcalibur software (Thermo Technologies) [11]. Volatile compounds were identified by retention time, electron impact mass spectrum and **similarity** match index. Then, the Terpene Index was calculated by using the ratio of the levels of linalool to the sum of linalool and geraniol.

2.4. Statistical analysis

Principal component analysis (PCA) and hierarchical cluster analysis were performed by SPAD 5.5 software (Programs developed by Optima Company, France).

3. RESULTS AND DISCUSSION

3.1. Volatile compounds

A total of 60 volatile substances were identified in fourteen samples of black tea, belonging to different chemical classes. A total ion current (TIC) chromatogram obtained for a Black tea samples can be seen in Fig. 1. Among the compounds identified terpenes (10.28 % - 59.88 %, ketones (1.85 % - 22.02 %) and esters (3.13 % - 418 %) represented the most abundant classes. In the last few years, many studies have been performed on characterization of the aromatic profile of black as well as green tea [12]. As known, the typical aroma of tea products is ascribed to a large number of volatiles, the nature and relative amount of which can be related to the precursor flavour composition (amino acid, fatty acid, carotenoids, etc...), the geographical area of cultivation, the breeds and the processing conditions, such as plucking, fermentation, withering and heating, thus determining a “fingerprint” of the product. Terpenoids form the most abundant class of aroma compounds detected in all samples (Table 1).

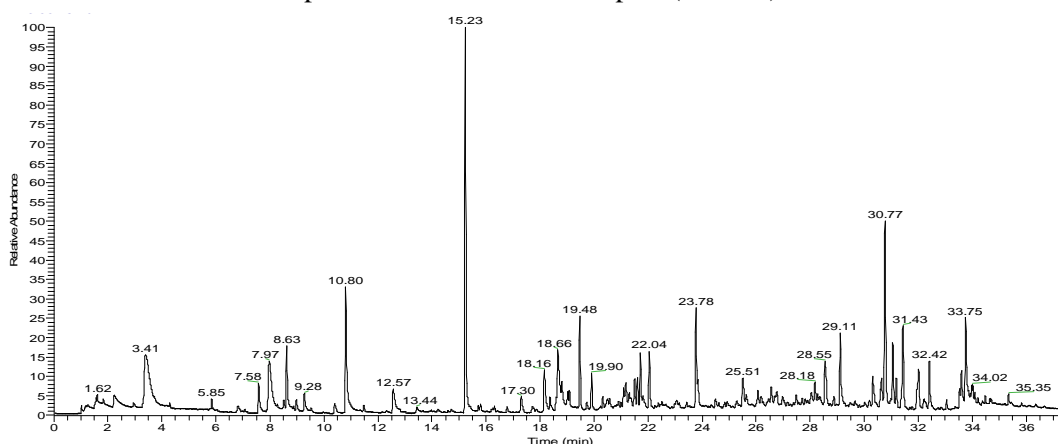


Figure 1. Typical total ion chromatogram of the volatile compounds in a Black tea sample.

Table 1. Volatile compounds detected in 14 Black tea samples by HS-SPME/GC-MS.

S.No	Compounds	Peak area ratio (%)													
		PQ-O	LT-C	PU-O	D-O	C-C	EW-O	ET-C	EH-C	RW-O	RT-C	N-C	G-O	H-O	S-O
		VN	VN	CHI	IND	IND	SRI	SRI	SRI	RUS	RUS	KEN	KEN	INDO	INDO
1	3-methyl-butanal	0.02	0.2	0.27	nd	nd	0.03	nd	0.34	0.04	0.1	0.07	0.28	0.09	0.07
2	pentanal	0.03	0.28	nd	0.04	nd	0.05	nd	0.48	0.05	0.14	0.09	0.36	0.12	0.09
3	2-methyl-2-pentenal	0.06	nd	nd	0.07	nd	0.08	nd	0.2	0.07	0.11	nd	0.36	0.16	0.08
4	hexanal	0.09	nd	0.25	0.29	nd	0.09	nd	nd	0.6	nd	nd	0.45	0.09	0.07
5	heptanal	0.08	nd	0.12	0.1	nd	0.1	nd	nd	0.12	nd	nd	nd	0.1	0.09
6	(<i>E</i>)-2-hexenal	1.8	0.62	nd	nd	nd	nd	nd	nd	nd	nd	0.76	0.21	nd	nd
7	<i>trans</i> -2-pentenal	nd	nd	nd	2.13	nd	nd	nd	nd	1.05	nd	nd	0.39	nd	nd
8	<i>n</i> -2-pentyl furan	nd	nd	0.49	nd	1	0.17	nd	nd	nd	0.59	nd	0.66	nd	nd
9	<i>cis</i> -3-hexenol	2.1	nd	0.12	0.4	nd	0.54	nd	nd	0.11	nd	nd	0.15	0.25	0.32
10	ocimene	nd	nd	nd	0.4	nd	nd	0.9	2.26	nd	nd	nd	nd	nd	nd
11	D-limonene	0.11	nd	0.21	2.65	2.57	0.11	10.5	11.9	0.81	1.94	nd	1.4	nd	nd
12	benzeneacetaldehyde	0.09	0.06	0.74	0.12	0.12	0.03	0.45	0.18	0.08	0.03	0.09	0.11	0.11	0.08
13	1-ethyl-2-formylpyrrole	nd	nd	8.14	1.84	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
14	<i>trans</i>-linalool oxide	0.19	nd	2.42	0.47	nd	0.03	nd	nd	nd	nd	nd	nd	0.18	0.08
15	furfuryl alcohol	0.56	nd	3.77	2.98	nd	0.08	nd	nd	nd	nd	nd	0.67	0.57	nd
16	β-linalool	12.9	1.1	6.35	16.3	0.43	0.55	1.66	2.55	1.12	1.07	0.16	12	1.2	0.54
17	<i>n</i> -nonaldehyde	nd	nd	1.55	0.6	nd	0.09	nd	nd	nd	nd	nd	nd	nd	nd
19	safranal	5.73	nd	1.68	nd	nd	1.27	nd	nd	1.61	nd	nd	0.9	1.32	1.29
20	decanal	0.6	0.55	0.95	0.65	nd	0.08	0.12	nd	0.22	nd	nd	0.2	0.15	0.1
21	β -cyclocitral	0.43	0.25	1.73	0.35	0.35	0.53	1.02	nd	0.23	0.26	nd	1.18	0.63	0.41

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22	cis-geraniol	3.61	0.32	0.2	19.7	0.49	0.68	1.9	2.98	0.89	0.9	nd	0.47	0.14	0.07
23	keto-isophorone	0.23	0.24	nd	nd	0.08	0.15	0.43	nd	0.14	nd	nd	0.42	nd	0.09
24	α -ionol	0.43	0.31	6.28	6.68	0.21	0.63	0.52	0.31	0.2	0.38	nd	0.58	nd	0.15
25	copaene	1.12	0.77	nd	0.71	0.1	0.26	nd	nd	0.21	nd	nd	nd	nd	nd
26	β -damascenone	0.36	0.62	2.62	9.6	0.27	0.73	nd	0.46	nd	0.62	nd	0.95	0.31	0.15
27	ethyl caprylate	nd	nd	nd	4.62	nd	nd	13.7	nd	nd	nd	12.9	nd	nd	nd
28	tetradecane	nd	nd	0.66	nd	0.19	0.27	nd	3.97	0.19	0.82	nd	nd	1.97	1.84
29	T-neoclovene	nd	1.12	nd	nd	0.29	0.72	nd	nd	0.53	0.62	nd	nd	nd	0.25
30	α -Ionone	2.1	0.83	3.27	0.68	0.62	3.89	2.33	3.94	0.98	1.16	0.43	0.73	1.38	1.06
31	geranylacetone	0.76	nd	1.98	1.59	nd	6.44	nd	nd	nd	nd	nd	nd	1.74	nd
32	β -Ionone	6.6	7.36	8.33	3.06	3.24	4.87	21.5	9.06	3.82	5.75	44.4	10.2	7.97	4.49
33	unknown	0.22	2.77	0.66	nd	0.97	nd	8.2	11.5	1.27	12.22	11.6	2.18	1.78	11.3
34	α -farnesene	0.93	nd	0.21	1.41	nd	0.45	nd	nd	nd	nd	nd	nd	nd	nd
35	β -muurolene	0.76	nd	0.22	nd	nd	0.19	1.71	nd	nd	0.75	nd	9.67	nd	nd
36	β -guaiene	2.06	1.37	1.11	1.17	0.94	0.48	nd	nd	0.85	1.82	nd	0.88	0.4	1.37
37	dihydroactinidiolide	1.77	nd	1.33	nd	nd	1.36	nd	nd	nd	nd	nd	nd	nd	nd
38	nerolidol	1.89	nd	0.37	0.79	nd	2.02	nd	nd	nd	nd	nd	nd	nd	nd
39	3-hexen-1-ol benzoate	18.4	nd	nd	0.45	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.49
40	propanoic acid, 2-methyl	0.32	2.36	nd	4.05	18.2	nd	0.39	nd	21.76	7.45	nd	nd	nd	nd
41	hexadecane	nd	1.23	2.44	nd	nd	9.12	nd	6.68	nd	0.84	0.13	nd	5.52	7.23
42	unknown	0.16	1.82	nd	nd	nd	nd	nd	4.89	nd	7.52	nd	nd	nd	2
43	α -cadiol	0.34	nd	0.21	nd	nd	0.34	nd	nd	nd	nd	nd	nd	nd	nd
44	γ -ionol acetate	0.2	4.07	0.68	0.47	1.07	1.37	7.34	7.12	1.95	13.33	11.4	1.16	4.71	10.2
45	heptadecane	nd	5.83	nd	nd	0.57	2.24	nd	4.34	0.95	0.65	nd	nd	6.97	10

46	trimethyltetradecane	nd	1.35	nd	nd	nd	nd	nd	0.81	nd	nd	nd	nd	nd	8.14
47	unknown	nd	nd	nd	nd	2.54	1.76	nd	nd	2.72	2.54	nd	nd	nd	nd
48	unknown	nd	nd	nd	nd	1.49	1.68	nd	nd	1.31	1.09	nd	nd	nd	1.83
49	unknown	nd	nd	nd	nd	0.76	1.39	nd	nd	1.02	0.96	nd	nd	nd	nd
50	ricinoleic acid	nd	nd	nd	nd	0.76	1.49	nd	nd	1.96	nd	nd	nd	nd	nd
51	naphthalene	nd	4.86	nd	nd	1.1	nd	nd	1.09	1.76	nd	nd	nd	5.35	8.7
52	Z-9-hexadecen-1-ol	nd	nd	nd	nd	2.47	2.61	nd	nd	4.15	nd	nd	nd	nd	nd
53	hexahydrofarnesylacetone	0.5	11.6	3.54	0.7	12.8	10.33	7.21	2.37	18.58	6.39	1.04	9.24	2.24	2.97
54	farnesyl acetone	nd	5.64	nd	nd	2.04	4.37	nd	nd	3.03	2.34	nd	2.42	3.47	3.68
55	methyl palmitate	2.74	5.8	10.5	1.63	4.36	4.05	0.68	1.73	5.62	nd	nd	6.55	0.89	nd
56	dibutyl phthalate	nd	nd	nd	nd	nd	nd	nd	nd	nd	4	nd	nd	36.2	7.37
57	unknown	nd	nd	nd	nd	0.7	nd	0.68	1.41	1.06	3.15	2.81	nd	0.89	nd
58	methyl oleate	0.08	nd	4.36	nd	3.53	1.5	nd	nd	0.75	3.12	nd	nd	nd	nd
59	methyl linoleate	0.11	nd	7.15	nd	nd	1.59	nd	1.74	0.64	nd	nd	nd	nd	nd
60	phytol	16.7	25.0	0.5	0.4	23.4	15.34	3.95	4.8	6.43	2.94	1.18	22.8	1.08	1.94
terpenoid		44.43	38.43	35.18	54.77	32.64	32.45	45.99	37.83	14.95	17.59	46.19	59.88	12.98	10.28
aldehydes		8.50	1.71	5.56	4.00	0.12	1.82	0.57	1.20	3.84	0.38	1.01	3.26	2.03	1.87
alcohols		2.66	0.00	3.89	3.38	2.47	3.23	0.00	0.00	4.26	0.00	0.00	0.82	0.82	0.32
esters		3.13	9.87	22.7	6.72	8.96	8.51	21.72	10.6	8.96	16.45	24.3	7.71	41.8	17.6
ketones		1.85	18.1	8.14	11.9	15.19	22.02	7.64	2.37	21.75	9.35	1.04	3.79	7.76	6.89
others		26.57	20.22	9.95	6.34	28.28	18.12	9.27	35.12	35.12	41.83	14.55	12.08	22.59	51.53
Total % composition		87.1	88.3	85.4	87.1	87.66	86.15	85.19	87.1	88.88	85.6	87.1	87.5	87.98	88.5
Terpene Index¹ (TI)		0.77	0.77	0.97	0.45	0.47	0.44	0.47	0.46	0.56	0.55	1.00	0.96	0.79	0.78

nd: not detected; PQ-O: Phuquy-Orthodox; LT-C: Lipton-CTC; PU-O: Puerh-Orthodox; D-O: Darjeeling-Orthodox; C-C: Ceylon-CTC; EW-O: English Breakfast-

Orthodox; ET-C: English Breakfast Teekanne-CTC; EH-C: English Breakfast AHMAD-CTC; RW-O: Russisch Westchiff-Orthodox; RT-C: Russisch Teekanne-CTC; N-C: Nero Teekanne-CTC; G-O: Gold Teekanne-Orthodox; H-O: Heritage-Orthodox; S-O: SariWangi-Orthodox.

¹ (Linalool+ *trans*-linalool oxides)/(linalool+oxides+geraniol).

The peak numbers refer to the order of their appearance in the chromatogram of Fig. 1.

Phytol, an acyclic diterpene alcohol, has the highest amount (0.4 % - 25 %), followed by hexahydrofarnesylacetone (0.5 % - 18.6 %), farnesyl acetone (2.04 % - 5.6 %), β -linalool (0.43 % - 16.31 %) and β -ionone (3.06 % - 44.4 %), α -ionone (0.4 % - 3.9 %). Among non-terpenoids, ester compounds like methyl palmitate (0.7 % - 10.5 %), methyl linoleate (0.11 % - 7.15 %) were present in a relatively high amount. Other compounds like aldehydes (hexanal, pentanal, phenyl acetaldehyde, etc...), alcohols (3-hexen-1-ol, furfuryl alcohol, etc...) were detected in somewhat lower amounts. The HS-SPME/GC-MS method was shown to be fully suitable for the analysis of volatile compounds in black tea due to its selectivity and sensitivity [13].

It has been reported that the fermentation rate of tea shoots crushed by the CTC roller was faster than that of tea shoots crushed by the orthodox roller. Furthermore, the non- or light withered leaves showed stronger fermentation ability than did the heavily-withered leaves after rolling [14]. Therefore, it was considered that the low total contents of aldehydes and alcohols in the volatile profile of CTC tea in comparison with OTD tea in comparison between the samples in the same region, as showed in Table 1, might be a result of the higher fermentation rate of light withered leaves, while the contents of esters in the CTC teas is higher than those of OTD teas may be due to a decrease in the contents of alcohols. On the other hand, the high number of detected compounds in orthodox tea i.e. in comparison between two Vietnam samples, 39 detected compounds for orthodox and 27 detected compounds for CTC, might be related to the lower fermentation rate of heavily-withered leaves. The difference in the volatile profile between Orthodox and CTC black tea may affect the aroma characteristics of both teas.

3.2. Statistical analysis

The common and large peaks allowed faster compound identification, and were more stable, with better repeatability of quantification. For these reasons, some volatile compounds that appeared in low or trace quantities were excluded in statistical analysis.

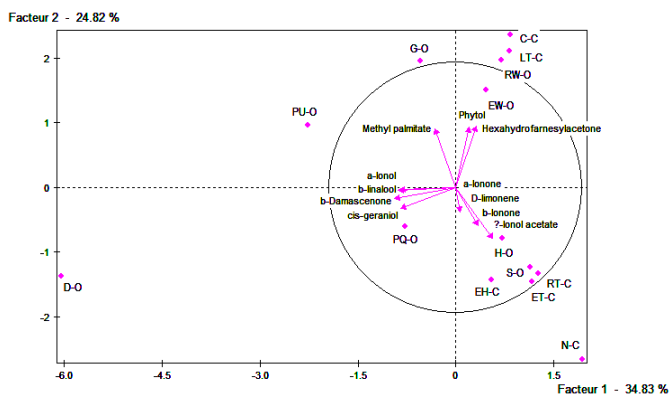


Figure 2. PCA score plots of samples based on the relative content of 11 volatile compounds.

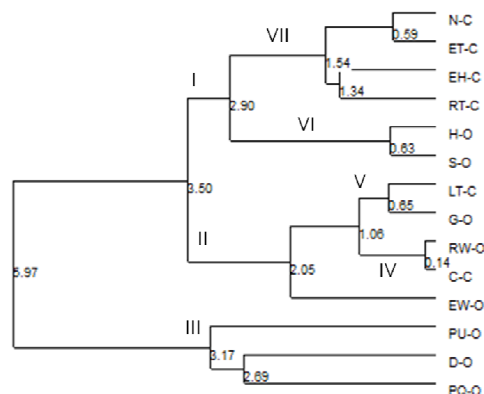


Figure 3. Clustering of 14 samples based on the relative content of 11 volatile compounds.

In order to have a better visualization of the samples structure and variety similarity, hierarchical cluster analysis was performed based on the relative content of 11 volatile compounds which have the peak area ratio higher than 1 % (D-limonene, β -linalool, cis-

geraniol, α -ionol, β -damascenone, α -ionone, β -ionone, γ -ionol acetate, hexahydrofarnesylacetate, methyl palmitate and phytol). Three clusters could be generated at a distance of higher than 3.17 but lower than 3.50 in the dendrogram, while cluster VII and VI could be assigned to types of CTC and Orthodox teas, respectively (Fig. 3). Cluster IV and I, however, could not distinguish CTC from OTD teas, indicating that Lipton-CTC and Ceylon-Orthodox teas have more similar aromatic profiles with Gold Teekanne-Orthodox and Russisch Westchiff-Orthodox, respectively. The PCA score plots of black tea samples obtained when performing principal component analysis (PCA) using the complete data set, jointly accumulating for 59.65 % of the total variance (Fig. 2). A clear separation according to geographic regions is not achieved when producing region/geographic region was considered except Heritage-Orthodox and Sariwangi-Orthodox samples, indicating that PCA may not be effective for discrimination purpose when the samples are collected from different type of black teas (i.e Orthodox and CTC teas)

3.3. Terpene Index

The content of both linalools and geraniol have been shown to vary widely within and between different tea cultivars when they are subjected to different manufacturing and agronomic treatments, and when cultivars are grown in different geographical regions. However, every variety/clone of teas has a specific terpene index, which varies only with plucking method. This index has been shown to be a reliable statistic tool for the differentiation of tea cultivars [15]. A terpene index was calculated for all tea samples using the ratio of the levels of linalools (linalool and *trans*-linalool oxides) to the sum of linalools and geraniols (Table 1). Significant difference was noted between teas manufactured in differing countries. However, products from India and Sri Lanka showed no difference.

Normally, methods of production cause changes in chemical composition of teas. Comparing the effects of CTC and OTD technique, it was noted that generally the OTD technique produce black teas with higher number of volatile compounds than CTC technique [16]. However, TI is specific for the cultivar. It does not depend on processing methods. According to previous studies, however, use of TI as a chemotaxonomic criterion has some problems. Firstly, it was demonstrated that plucking standards affect TI. Secondly, TI can only have some values between 0 and 1; but there are many cultivars. This implies that some clones have same TI and therefore other additional parameters must be use to discriminate/authenticate tea products.

4. CONCLUSION

Among the 60 volatiles identified, terpenes (10.28 % - 59.88 %), ketones (1.85 % - 22.02 %) and esters (3.13 % - 418 %) represented the most abundant compounds. The results showed that CTC teas have a lower number of volatile components detected, lower contents of aldehydes and alcohols, and higher content of esters than OTD teas. PCA is an unsupervised statistical method, allowing to describe the behaviour of the data without the constraint of initial assumptions on samples. Analyzing the whole set of data, eleven principal components were needed to explain about 59 % of the total variance. Therefore, PCA did not reveal useful in the discrimination of the samples investigated from different regions, since clusters of them were not clearly detected. Combining PCA with Cluster analysis could **classify** types of teas, i.e. OTD

and CTC. In addition, the Terpene Index is also applied as additional information for differentiating geographic region of Black teas.

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