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Characterization of key aroma compounds and relationship between aroma compounds and sensory attributes in different aroma types of Fu brick tea

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nerolidol (PubChem CID: 5284507)
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α-terpineol (PubChem CID: 17100)
acetophenone (PubChem CID: 7410)
(E,Z)-2,6-nonadienal (PubChem CID: 643731)
hexanal (PubChem CID: 6184)
dihydro-*β*-ionone (PubChem CID: 519382)
(E,E)-2,4-nonadienal (PubChem CID: 5283339).

ABSTRACT

Aroma is one of the most important sensory properties of tea. Floral-fungal aroma type, ripe-fungal aroma type and fresh-fungal aroma type were the main aroma types of Fu brick tea by QDA. A total of 112 volatile compounds were identified and quantified in tea samples by HS-SPME/GC–MS analysis. Ten voaltiles in floral-fungal aroma type, eleven voaltiles in ripe-fungal aroma type, and eighteen voaltiles in fresh-fungal aroma type were identified as key aroma compounds for the aroma characteristics formation in three aroma types of Fu brick tea. In addition, PLS analysis revealed that 3,4-dehydro-*β*-ionone, dihydro-*β*-ionone, (+)-carotol and linalool oxide *II* were the key contributors to the 'floral and fruity' attribute, *α*-terpineol contributed to 'woody' and 'stale' attributes, and thirteen aroma compounds related to 'green' attribute. Taken together, these findings will provide new insights into the formation mechanism of different aroma characteristics in Fu brick tea.

1. Introduction

Fu brick tea is a typical post-fermented dark tea, which is mainly produced in Hunan, Shaanxi and Zhejiang provinces of China. The manufacturing process of Fu brick tea includes steaming, piling, pressing, fermentation, and drying (Li et al., 2019; Ling et al., 2010; Xu, Mo, Yan, & Zhu, 2007). Microbial fermentation was considered as the key step in the process of Fu brick tea, which helped to form its unique 'fungal flower' aroma and mellow taste (Ling et al., 2010). In recent years, Fu brick tea has attracted global interest owing to its special flavor and health benefits, such as anti-obesity, anti-hyperlipidemia, anti-oxidant, anti-tumor, anti-microbial, and others (Fu et al., 2011; Li et al., 2013; Mo, Zhang, Li, & Zhu, 2008; Xu, Wang, Wen, Liu, Liu, & Li, 2011).

Tea aroma is one of the most important sensory properties reflecting the quality of tea (Zhu et al., 2018). 'Fungal flower' aroma is the basic aroma characteristics of Fu brick tea, which is formed by a variety of volatile compounds with different aroma attributes mixed together (Li et al., 2020). However, the aroma characteristics of Fu brick tea vary greatly with cultivation conditions, processing, regions and storage time. Previous study indicated that the 'fungal flower' aroma of aged Fu brick tea was dominated with 'stale' attribute and was enriched by methyl hexadecanoate, 1-octanol, methyl laurate, methyl tetradecanoate, 1-heptadecanol (Huang, Wang, Zeng, & Lai, 2011). Otherwise, the 'fungal flower' aroma of Fu brick tea processing in Hunan province was dominated with 'floral', 'woody', 'green' attributes and was enriched in methyl salicylate, acetophenone, cedrol, benzyl alcohol (Li et al., 2020). The 'fungal flower' aroma of Fu brick tea processing in

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Shaanxi province was dominated with 'roasted' attribute and enriched in heteroxic compounds, such as 2-pentylfuran and tetramethylpyrazine. The 'fungal flower' aroma of Fu brick tea producing from Zhejiang province was usually dominated by the 'green' attribute and was riched in low grade fatty aldehydes such as hexanal (Cao et al., 2018; Zhao, Xu, Wu, Jiang, & Zhu, 2017). The 'fungal flower' aroma of Fu brick tea processed from Sichuan province was dominated by 'floral and fruity' attribute and its main volatile components are methyl salicylate, geranylacetone and β-ionone (Nie et al., 2019). To sum up, it is reasonable to speculate that there are different 'fungal flower' aroma characteristics, which might result in different aroma types in Fu brick tea. But up to now, there was no research focus on the aroma characteristics in different aroma types of Fu brick tea.

Therefore, the aims of the present study were to (a) classify the aroma characteristics of Fu brick tea in different aroma types according to the sensory evaluation; (b) identify the key aroma compounds contributing to the formation of different aroma characteristics in Fu brick tea; (c) illuminate the relationship between the aroma compounds and sensory attributes in Fu brick tea. This study is of significant importance for providing information to enhance our understanding of the mechanisms on different aroma characteristics formation in Fu brick tea.

2. Method and material

2.1. Chemicals

The C_7-C_{40} *n*-alkanes and ethyl decanoate (99.99%) were obtained from Sigma-Aldrich (St. Louis, MO). Forty two authentic standards were purchased from J&K chemical Ltd. (Beijing, China). All the chromatographic solvents were of chromatography grade and all of the chemicals were of analytical reagent grade unless otherwise stated.

2.2. Samples and sensory analysis

Sixty Fu brick tea samples were collected from the tea market all over China. Sensory analysis was approved by Hunan Agricultural University Institutional Review Board Committee (#TSF-780-2020). Sensory evaluation was performed by eight well-trained panelists from the Tea Science Department in Hunan Agricultural University (four males and four females, aged from 25 to 55 years) according to the Chinese standards "Methodology of Sensory Evaluation of Tea" (GB/T 23776-2018). Before the experiment, each panelist had to complete a 60-hours of sensory evaluation training with different aroma types of Fu brick tea in 20 days. All participants received written information about the study, and they signed informed consent to participate. Sensory quantitative description analysis (QDA) was performed according previous study (Li, Luo, Wang, Fu, & Zeng, 2019). Briefly, 3.0 g tea sample was infused with 150 mL of boiled water for 7 min in a special tea cup. Each sample was coded with a three-digit number and randomly offered to panelists after brewing. The aroma descriptors were evaluated and discussed by panelist panel. The intensity of aroma attribute was scored by panelists using a scale from 0 to 10. Each score was expressed as mean value. Each sample was evaluated three times by each panelist in different days.

2.3. Qualitative and quantitative analysis of the volatiles in Fu brick tea by HS-SPME/GC–MS analysis

The volatile profile of Fu brick tea was extracted by headspace solid-phase micro-extraction (HS-SPME) using a 65 μm polydimethylsiloxane/divinylbenzene (PDMS/DVB) fiber (Supelco, Bellefonte, PA, USA) and was analysed using a GC system (Agilent 5977B, Agilent Technologies Inc., CA, USA) equipped with a mass spectrometer (Agilent 5977A, Agilent Technologies Inc., CA, USA), according to our previous study (Li et al., 2020). Ethyl decanoate (8.64 mg/L) was used as internal standard. An Agilent HP-5MS capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness) was employed for separating the volatile compounds, and helium (purity > 99.999 %) was used as carrier gas with a constant flow rate of 1 mL/min. The injector temperature was set at 250 °C with splitless model. The GC oven temperature was set at 50 °C for 2 min, then increased at a rate of 3 °C/min to 160 °C and held for 1 min; lastly, it increased to 280 °C at a rate of 15 °C/min and kept for 1 min. The mass spectrometer conditions were set as follows: ionization mode, EI; ion source temperature, 230 °C; quadrupole temperature, 150 °C; electron energy, 70 eV; full scan mode, mass scan range 35–400 atomic mass units (amu). Each sample was analyzed three times.

Volatile compounds were identified based on retention indices (RIs), authentic standards, and mass spectra matching in the standard NIST17 library. C_7-C_{40} *n*-alkane mixture was employed for determination of RIs. Each standard was mix with 'volatile-free' tea to obtain the standard curves. The 'volatile-free' tea was prepared by vacuum concentration to remove volatile compounds according to a previous study (Du, Wang, Li, Xiao, Li, & Xu, 2013). For the volatile compounds without available standards, the quantitation was carried out using the standard that had the same carbon atom or a similar functional structure (Li et al., 2020).

2.4. Odor activity values (OAVs) calculation

OAV was calculated by dividing the concentration of volatile compound with its odor threshold (OT) reported by the references. Volatile compounds with OAV > 1 were considered as an aroma-active compound, which played an important role for the aroma characteristics formation of tea samples (Mao, Lu, Li, Ye, Wei, & Tong, 2018).

2.5. Statistical analysis

The data were preprocessed by mean centering and scaling prior to analysis. Principal component analysis (PCA), hierarchical cluster analysis (HCA), orthogonal partial least squares discriminant analysis (OPLS-DA) and partial least squares analysis (PLS) were performed by SIMCA-P+ (Version 14.0, Umetrics, Umea, Sweden). Analysis of variance (ANOVA) was performed by SPSS (Version 22.0, IBM, Armonk, NY, USA). All data were presented as the mean value ± SD. Significant differences between groups were declared significant at $p < 0.05$.

3. Results and discussion

3.1. Aroma characteristics of Fu brick tea

After sensory evaluation, 25 samples with typical flavor characteristics of Fu brick tea were selected from 60 samples and used for subsequent sensory QDA analysis. In total, 40 attributes that described the aroma characteristics were obtained (Fig. 1A). The flavor wheel was consist of 3 tiers, 1 first-tier descriptor, 10 s-tier descriptors and 40 third-tier descriptors. The third-tier descriptors were precise descriptions to Fu brick tea, such as cherry fragrance, orchid sweet, old house smell and mushroom scent. The second-tier descriptors were the summary of the third-tier descriptors, that is, the concrete aroma attributes of Fu brick tea, which were split into 10 categories, including fermented, fruity, floral, nutty, stale, woody, green, hay flavor, fishy smell and herbal. All of the second-tier descriptors have been often used to describe the characteristics of Fu brick tea in other reports (Li et al., 2019; Li et al., 2020; Lv, Wu, Li, Xu, Liu, & Meng, 2014; Nie et al., 2019). It is worth mentioning that the 'fungal flower' aroma attribute could be a complex aroma, which might be formed by a variety of volatiles with woody, floral and mint attributes mixed together (Li et al., 2020). So, this descriptor was not presented in the flavor wheel independently. After discussion, all panelists agreed to use the five attributes with higher odor intensity as useful terms for subsequent evaluation of the aroma characteristics of Fu brick tea, including 'floral and fruity', 'stale', 'woody', 'green' and 'herbal'.

Using the QDA method, 25 samples were clustered into three groups

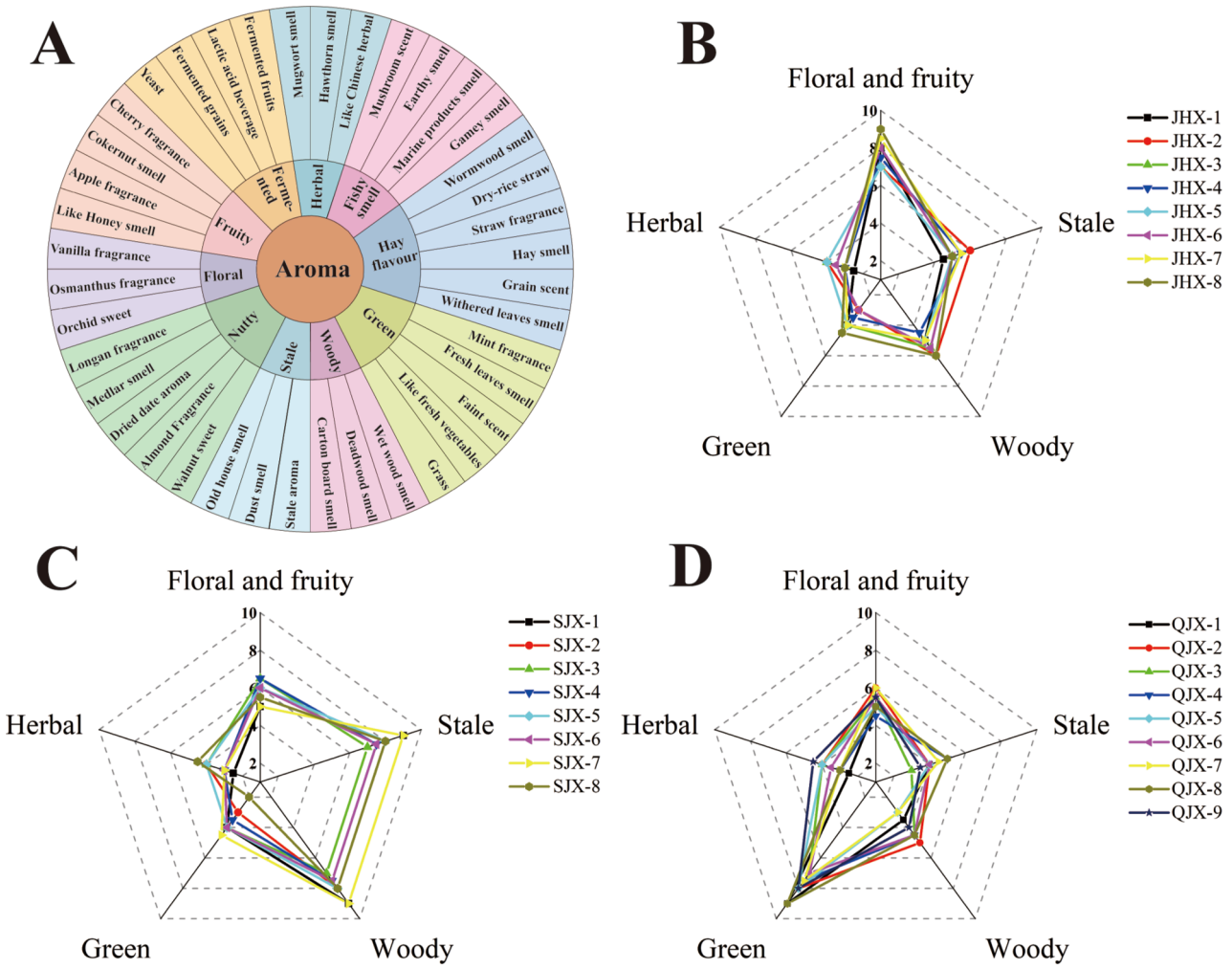

Fig. 1. Quantitative descriptive analysis of different aroma types of Fu brick tea. (A) Flavor wheel. (B) JHX: floral-fungal aroma type. (C) SJX: ripe-fungal aroma type. (D) QJX: fresh-fungal aroma type.

based on sensory intensity (Fig. 1B–D). The first group had strong 'floral and fruity' attribute, while weaked in the other attributes. The second group, with a typical 'stale' attribute, also was rich in 'woody' attribute. The third group had strong 'green' attribute, whereas the other attributes were moderate. These noticeable differences suggested that the samples had significantly different flavor and intensities. Generally, eight Fu brick tea samples with strong 'floral and fruity' attribute were classified as 'floral-fungal' aroma (JHX) type, eight samples with strong 'stale' and 'woody' attributes were classified as 'ripe-fungal' aroma (SJX) type, and nine samples with strong 'green' attribute were classified as 'fresh-fungal' aroma (QJX) type based on the sensory evaluation.

3.2. Comparison of volatile profiles in three aroma types of Fu brick tea

To illuminate the volatile profiles of three aroma types of Fu brick tea, a comprehensive analysis was performed by HS-SPME/GC–MS. A total of 112 volatile compounds were identified and quantified in Fu brick tea samples by absolute quantitative method (Table 1 and Table S1). For clarification, the identified compounds were further classified into 10 different sub-classes, including 26 hydrocarbons, 21 ketones, 20 alcohols, 20 aldehydes, 9 esters, 6 heteroxy compounds, 3 phenols, 2 lactones, 1 pyrrole and 4 others. Generally, the total content of volatile compounds in QJX type was much higher than that in JHX and SJX types of samples (Fig. 2A). Alcohols, ketones, aldehydes, hydrocarbons were the predominant volatile categories in Fu brick tea, which totally accounted for 85.47% in the identified volatile compounds (Fig. 2B). Previous studies also reported that alcohols, ketones, aldehydes, hydrocarbons might be the main contributing substance to the characteristic aroma formation of Fu brick tea, which was consistent with our present results (Li et al., 2019; Xu, Mo, Yan, & Zhu, 2007). Notably, alcohols were the most volatiles category in Fu brick teas (29.11%, 26.91%, and 24.32%), followed by ketones, aldehydes and hydrocarbons in the three aroma types of Fu brick tea, but it was not consistent with previous studies that reported ketones was the most abundant volatile category (Lv et al., 2014; Shi, Zhu, Zhang, Lin, & Lv, 2019). This difference might be due to the absolute quantitative method used in our present study, otherwise, the relative quantitative method, such as peak area normalization method used in previous studies, might result in the lower relative content of alcohol compounds (Lv et al., 2014; Shi et al., 2019). Furthermore, alcohols, ketones and hydrocarbons were more abundant in JHX type, whereas ketones and aldehydes were more abundant in SJX and QJX types of Fu brick tea.

To obtain a preliminary overview of similarities and differences among the different aroma types of Fu brick tea, unsupervised PCA were carried out based on the absolute content of identified volatile compounds, and most of the chemical phenotypes of the samples were well discriminated according to the aroma types of Fu brick tea. As shown in Fig. 3A (PC1, 26.4%; PC2, 13.1%; R2X = 0.852), there was a clear differentiation among the three aroma types of Fu brick tea samples, suggesting that the volatile profile of tea samples differed dramatically in their content levels. Furthermore, HCA analysis was also performed to distinguish the similarities and differences of volatile composition and

Table 1
Qualitative and quantitative results of volatile components among three aroma types of Fu brick tea.

Compounds	CAS	Quantitative ions (*m/z*)[a]	RI[b]/RI[c]	Identification[d]	Concentration (μg/L)			Proportion (%)		
					JHX	SJX	QJX	JHX%	SJX%	QJX%
Esters										
Sebacic acid, di(2-hexyl) ester	1000355–65-8	185,98,143,166	1851/-	MS	1.49 ± 0.93	1.96 ± 1.33	1.46 ± 0.62	0.03 ± 0.01	0.04 ± 0.03	0.02 ± 0.01
Octyl 4-methoxycinnamate	5466–77-3	161,178,133,207	2329/-	MS	9.58 ± 0.16	9.71 ± 0.44	9.69 ± 0.37	0.17 ± 0.03	0.18 ± 0.05	0.11 ± 0.02
Diethyl phthalate	84–66-2	149,177,105,121	1592/1602	MS RI	13.17 ± 4.57	10.81 ± 1.18	29.42 ± 47.91	0.23 ± 0.1	0.21 ± 0.07	0.35 ± 0.52
Diisobutyl phthalate	84–69-5	149,57,223,104	1870/1869	MS RI	26.67 ± 7.3	18.37 ± 7.02	20.76 ± 7.48	0.46 ± 0.16	0.35 ± 0.06	0.24 ± 0.06
Methyl hexadecanoate	112–39-0	74,143,87,227	1923/1926	MS RI	2.81 ± 0.23	2.6 ± 0.17	2.87 ± 0.26	0.05 ± 0.01	0.05 ± 0.01	0.03 ± 0
Ethyl palmitate	628–97-7	88,101,157,241	1990/1992	MS RI	0.66 ± 0.49	0.42 ± 0.07	0.54 ± 0.13	0.01 ± 0.01	0.01 ± 0	0.01 ± 0
Dibutyl phthalate	84–74-2	149,150,223,205	1965/1965	MS RI	14.09 ± 0.54	11.51 ± 0.59	12.5 ± 1.08	0.24 ± 0.05	0.22 ± 0.07	0.15 ± 0.03
Methyl salicylate	119–36-8	120,152,92,121	1191/1191	MS RI STD	294.09 ± 169.47	225.83 ± 219.33	167.88 ± 68.53	5.11 ± 3.06	4.28 ± 2.73	1.97 ± 0.91
2,2,4-trimethyl-1,3-pentanediol diisobutyrate	6846–50-0	71,111,43,83	1595/-	MS	31.62 ± 18.18	12.12 ± 4.83	26.03 ± 13.98	0.55 ± 0.51	0.23 ± 0.13	0.31 ± 0.14
Heteroxy compounds										
1,2-dimethoxybenzene	91–16-7	138,123,95,77	1144/1148	MS RI STD	37.29 ± 13.06	58.66 ± 32.57	107.37 ± 51.96	0.65 ± 0.28	1.11 ± 0.46	1.26 ± 0.67
1,3-dimethoxybenzene	151–10-0	138,107,122,95	1164/-	MS	26.7 ± 6.28	29.62 ± 23.61	30.47 ± 14.86	0.46 ± 0.28	0.56 ± 0.25	0.36 ± 0.17
5-methoxy-6,7-dimethylbenzofuran	35355–35-2	161,131,145,176	1465/1468	MS RI	24.85 ± 5.68	62 ± 16.59	46.55 ± 14.11	0.43 ± 0.22	1.18 ± 0.43	0.55 ± 0.17
4-methylanisole	104–93-8	122,77,91,107	929/-	MS STD	6.77 ± 1.87	5.65 ± 1.53	22.61 ± 9.98	0.12 ± 0.02	0.11 ± 0.05	0.27 ± 0.13
2-pentylfuran	3777–69-3	82,53,81,138	987/988	MS RI STD	8.73 ± 2.53	4.41 ± 2.87	20.86 ± 4.11	0.15 ± 0.03	0.08 ± 0.03	0.25 ± 0.05
cis-anethol	104–46-1	148,133,121,77	1281/1283	MS RI	5.32 ± 2.03	5.16 ± 1.84	8.28 ± 6.86	0.09 ± 0.03	0.1 ± 0.03	0.1 ± 0.12
Ketones										
Geranylacetone	3796–70-1	43,69,151,107	1451/1449	MS RI STD	186.72 ± 67.15	145.01 ± 60.04	241.66 ± 54.02	3.24 ± 0.88	2.75 ± 1.74	2.84 ± 0.8
Dihydro-*β*-ionone	17283–81-7	126,161,176,121	1436/1438	MS RI	35.05 ± 0.44	35.8 ± 1.19	35.47 ± 0.56	0.61 ± 0.12	0.68 ± 0.22	0.42 ± 0.07
α-ionone	127–41-3	121,93,136,192	1425/1425	MS RI STD	96.18 ± 22.32	78.04 ± 25.69	120.34 ± 30.4	1.67 ± 0.27	1.48 ± 0.71	1.41 ± 0.39
β-ionone	79–77-6	177,123,135,159	1484/1483	MS RI STD	37.6 ± 12.36	15.59 ± 6.08	45.33 ± 12.72	0.65 ± 0.18	0.3 ± 0.1	0.53 ± 0.14
3,4-dehydro-*β*-ionone	1203–08-3	175,91,147,190	1481/1482	MS RI	26.29 ± 7.1	13.89 ± 2.27	26.13 ± 7.49	0.46 ± 0.07	0.26 ± 0.06	0.31 ± 0.1
Benzophenone	119–61-9	105,182,77,51	1624/1623	MS RI	6.99 ± 0.94	7.07 ± 1.35	8.2 ± 4.35	0.12 ± 0.03	0.13 ± 0.05	0.1 ± 0.05
(E)-3-nonen-2-one	18402–83-0	125,55,97,140	1137/1134	MS RI	17.49 ± 5.45	14.67 ± 3.21	26.89 ± 4.53	0.3 ± 0.09	0.28 ± 0.12	0.32 ± 0.1
4-methyleneisophorone	20548–00-9	150,135,107,91	1215/-	MS	13.74 ± 0.96	15.37 ± 1.97	16.17 ± 1.17	0.24 ± 0.04	0.29 ± 0.09	0.19 ± 0.03
2,5-pyrrolidinedione,1-ethyl-	2314–78-5	127,56,84,112	1132/1134	MS RI	14.06 ± 3.43	20.98 ± 17.04	24.37 ± 12.93	0.24 ± 0.04	0.4 ± 0.42	0.29 ± 0.14
Dihydro-*α*-ionone	31499–72-6	121,136,95,176	1413/-	MS	79.8 ± 23.15	56.71 ± 33.74	121.26 ± 30.26	1.39 ± 0.37	1.08 ± 0.88	1.43 ± 0.38
Hexahydrofarnesylacetone	502–69-2	58,109,194,137	1845/1846	MS RI	62.23 ± 4.09	56.38 ± 2.32	64.92 ± 3.13	1.08 ± 0.16	1.07 ± 0.36	0.76 ± 0.16
3,5-dihydroxyacetophenone	51863–60-6	152,137,109,81	1152/-	MS	464.33 ± 146.01	133.35 ± 64.38	393.76 ± 131.95	8.07 ± 1.73	2.53 ± 0.74	4.63 ± 1.82
3-formyl-5,5-dimethyl-2-cycolohexen-1-one	56621–35-3	96,152,109,68	1140/-	MS	11.25 ± 4.58	7.9 ± 4.5	11.6 ± 2.73	0.2 ± 0.07	0.15 ± 0.14	0.14 ± 0.05
Ionone	8013–90-9	136,192,177,121	1274/-	MS	24.78 ± 2.24	21.03 ± 1.82	26.75 ± 2.72	0.43 ± 0.1	0.4 ± 0.14	0.31 ± 0.07
4-*t*-butylpropiophone	81561–77-5	147,175,190,105	1379/-	MS	12.79 ± 2.06	10.73 ± 0.54	12.8 ± 1.81	0.22 ± 0.05	0.2 ± 0.06	0.15 ± 0.02
Pulegone	89–82-7	110,152,81,95	1244/1237	MS RI STD	81.33 ± 35.64	126.8 ± 40.55	149.09 ± 45.32	1.41 ± 0.61	2.41 ± 1.29	1.75 ± 0.72
2,2,6-trimethyl-cyclohexanone	2408–37-9	82,69,140,56	1030/1030	MS RI	27.27 ± 6.13	7.47 ± 4.21	25.48 ± 7.67	0.47 ± 0.12	0.14 ± 0.05	0.3 ± 0.06
(*E,E*)-3,5-octadien-2-one	30086–02-3	95,81,109,43	1068/1074	MS RI	44.83 ± 19.41	65.01 ± 35.37	201.01 ± 67.52	0.78 ± 0.33	1.23 ± 0.71	2.36 ± 0.73
Methylheptenone	110–93-0	108,93,69,83	982/981	MS RI STD	24.5 ± 7.36	30.11 ± 10.62	51.9 ± 10.24	0.43 ± 0.13	0.57 ± 0.32	0.61 ± 0.21

(*continued on next page*)

Table 1 (*continued*)

Compounds	CAS	Quantitative ions (*m/z*)[a]	RI[b]/RI[c]	Identification[d]	Concentration (μg/L)			Proportion (%)		
					JHX	SJX	QJX	JHX%	SJX%	QJX%
Acetophenone	98–86-2	105,120,77,51	1061/1061	MS RI STD	37.65 ± 6.71	83.13 ± 79.03	58.11 ± 10.66	0.65 ± 0.16	1.58 ± 2.44	0.68 ± 0.11
3-octen-2-one	1669–44-9	55,111,126,97	1035/1040	MS RI STD	0 ± 0	0 ± 0	3.02 ± 2.09	0 ± 0	0 ± 0	0.04 ± 0.03
Hydrocarbons										
Dehydro-*ar*-ionene	30364–38-6	157,172,142,109	1350/1349	MS RI	77.44 ± 15.89	32.02 ± 13.1	59.79 ± 19.76	1.35 ± 0.26	0.61 ± 0.17	0.7 ± 0.2
α-ionene	475–03-6	159,174,160,131	1353/1352	MS RI	39.59 ± 7.15	21.85 ± 9.08	33.77 ± 9.44	0.69 ± 0.13	0.41 ± 0.06	0.4 ± 0.11
1,2,3-trimethoxybenzene	634–36-6	168,153,177,105	1311/1316	MS RI STD	36.37 ± 10.22	136.91 ± 90.13	187.02 ± 68.07	0.63 ± 0.27	2.6 ± 1.6	2.2 ± 0.75
1-methylnaphthalene	90–12-0	142,166,141,115	1304/1306	MS RI STD	3.28 ± 0.95	1.06 ± 0.98	5.71 ± 2.01	0.06 ± 0.02	0.02 ± 0.02	0.07 ± 0.03
Naphthalene	91–20-3	128,64,77,102	1178/1178	MS RI STD	4.68 ± 2.8	1.67 ± 1.99	7.72 ± 2.52	0.08 ± 0.07	0.03 ± 0.03	0.09 ± 0.03
Phenylethylene	100–42-5	104,78,103,58	883/888	MS RI	29.96 ± 9.59	32.67 ± 11.56	50.62 ± 22.12	0.52 ± 0.2	0.62 ± 0.28	0.6 ± 0.18
1-ethylcyclohexene	1453–24-3	67,81,95,110	1008/-	MS	62.25 ± 37.55	123.2 ± 57.79	319.75 ± 128.28	1.08 ± 0.59	2.34 ± 1.44	3.76 ± 1.16
2,6,10,14-tetramethylpentadecane	1921–70-6	71,57,113,85	1703/1703	MS RI	8.28 ± 2.6	4.6 ± 1	6.97 ± 2.94	0.14 ± 0.08	0.09 ± 0.04	0.08 ± 0.03
2,2′,5,5′-tetramethylbiphenyl	3075–84-1	195,210,165,180	1680/1675	MS RI	5.95 ± 2.18	4.56 ± 0.56	6.04 ± 1.44	0.1 ± 0.05	0.09 ± 0.03	0.07 ± 0.03
α-cedrene	469–61-4	119,161,204,93	1409/1409	MS RI STD	139.09 ± 128.7	244.08 ± 231.94	125.48 ± 55.02	2.42 ± 2.15	4.63 ± 4.44	1.48 ± 0.52
Calamenene	483–77-2	159,204,137,123	1521/1522	MS RI	33.15 ± 14.82	31.97 ± 18.2	33.85 ± 8.1	0.58 ± 0.26	0.61 ± 0.16	0.4 ± 0.07
3,4-dimethoxytoluene	494–99-5	152,137,167,109	1236/1241	MS RI STD	11.9 ± 2.51	10.15 ± 8.01	20.11 ± 14.7	0.21 ± 0.21	0.19 ± 0.1	0.24 ± 0.17
3,5-dihydroxyamylbenzene	500–66-3	124,180,137,81	1523/1528	MS RI	111.34 ± 30.54	89.39 ± 36.43	162.6 ± 37.48	1.93 ± 0.44	1.7 ± 0.67	1.91 ± 0.42
(+)-*β*-cedrene	546–28-1	161,204,120,93	1418/1420	MS RI	25.43 ± 13.95	37.82 ± 26.96	25.57 ± 6.71	0.44 ± 0.26	0.72 ± 0.51	0.3 ± 0.06
1,6-dimethylnaphthalene	575–43-9	156,141,177,121	1412/1410	MS RI	23.73 ± 4.56	17.24 ± 4.2	29.21 ± 4.88	0.41 ± 0.09	0.33 ± 0.11	0.34 ± 0.07
3,4-diethylbiphenyl	61141–66-0	195,165,210,180	1708/-	MS	5.13 ± 1.25	4.11 ± 0.35	5.11 ± 1.04	0.09 ± 0.03	0.08 ± 0.03	0.06 ± 0.02
Hexadecane	638–36-8	71,57,43,85	1806/1809	MS RI	5.12 ± 1.43	3.27 ± 0.39	4.29 ± 1.14	0.09 ± 0.04	0.06 ± 0.02	0.05 ± 0.01
2,6-di-*tert*-butyl-p-benzoquinone	719–22-2	135,220,177,121	1464/-	MS	30.61 ± 12.98	24.49 ± 7.52	34.02 ± 19.35	0.53 ± 0.2	0.46 ± 0.18	0.4 ± 0.25
2-vinylnaphthalene	827–54-3	154,153,128,76	1374/1381	MS RI	28.88 ± 7.07	20.29 ± 5.3	34.13 ± 7.65	0.5 ± 0.09	0.38 ± 0.08	0.4 ± 0.1
Fluorene	86–73-7	166,165,83,109	1573/1572	MS RI	71.62 ± 40.97	35.29 ± 37.67	94.06 ± 64.19	1.24 ± 0.67	0.67 ± 0.44	1.11 ± 1.09
Heptadecane	629–78-7	57,71,85,43	1697/1700	MS RI	12.4 ± 0.36	11.8 ± 0.09	12.06 ± 0.19	0.22 ± 0.05	0.22 ± 0.07	0.14 ± 0.02
m-xylene	108–38-3	91,105,77,56	860/864	MS RI	150.04 ± 41.27	116.47 ± 68.89	267.14 ± 106.14	2.61 ± 0.63	2.21 ± 0.97	3.14 ± 0.91
Phenanthrene	85–01-8	178,176,111,151	1775/1778	MS RI	4.96 ± 0.47	4.64 ± 0.7	5.53 ± 0.76	0.09 ± 0.01	0.09 ± 0.02	0.07 ± 0.01
Acenaphthene	83–32-9	153,77,154,123	1476/-	MS	26.92 ± 14.2	13.16 ± 3.39	14.97 ± 2.27	0.47 ± 0.22	0.25 ± 0.05	0.18 ± 0.04
Octadecane	593–45-3	57,71,85,99	1796/1800	MS RI	11.9 ± 0.14	11.63 ± 0.04	11.72 ± 0.06	0.21 ± 0.04	0.22 ± 0.07	0.14 ± 0.02
2-methylnaphthalene	91–57-6	142,141,115,139	1286/1285	MS RI STD	4.35 ± 1.45	1.22 ± 1.36	8.81 ± 3.37	0.08 ± 0.03	0.02 ± 0.02	0.1 ± 0.05
Aldehydes										
trans-2-hexenal	6728–26-3	69,83,98,55	843/846	MS RI STD	321.75 ± 87.14	308.27 ± 86.85	565.18 ± 237.91	5.59 ± 1.2	5.85 ± 2.11	6.64 ± 2.07
Hexanal	66–25-1	56,82,95,100	788/800	MS RI STD	129.62 ± 96.94	233.01 ± 88.35	572.45 ± 256.08	2.25 ± 1.8	4.42 ± 2.38	6.73 ± 2.35
Benzaldehyde	100–52-7	106,51,78,106	954/957	MS RI STD	62.64 ± 8.63	57.39 ± 17.62	123.65 ± 31.86	1.09 ± 0.11	1.09 ± 0.22	1.45 ± 0.3
Citral	5392–40-5	69,84,97,137	1266/1268	MS RI	8.74 ± 1.83	19.73 ± 11.09	17.72 ± 6.94	0.15 ± 0.02	0.37 ± 0.14	0.21 ± 0.09
(E)-2-octenal	2548–87-0	70,55,41,83	1054/1056	MS RI	31.09 ± 9.04	39.92 ± 10.16	77.26 ± 28.24	0.54 ± 0.18	0.76 ± 0.36	0.91 ± 0.28
Octanal	124–13-0	43,84,100,110	1000/1001	MS RI	20.24 ± 14.75	17.4 ± 17.68	12.83 ± 18.77	0.35 ± 0.28	0.33 ± 0.42	0.15 ± 0.24
Nonanal	124–19-6	57,98,82,114	1101/1101	MS RI	56.71 ± 36.96	39.64 ± 24.15	177.34 ± 85.64	0.99 ± 0.67	0.75 ± 0.57	2.08 ± 0.78

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Table 1 (*continued*)

Compounds	CAS	Quantitative ions (*m/z*)[a]	RI[b]/RI[c]	Identification[d]	Concentration (μg/L)			Proportion (%)		
					JHX	SJX	QJX	JHX%	SJX%	QJX%
trans-2-decenal	3913–81-3	70,83,121,55	1257/1258	MS RI	15.23 ± 0.19	16.74 ± 2.9	16.83 ± 0.95	0.26 ± 0.05	0.32 ± 0.09	0.2 ± 0.04
(*E,E*)-2,4-heptadienal	4313–03-5	81,67,79,110	993/997	MS RI STD	23.04 ± 17.14	37.17 ± 22.92	89.97 ± 62.12	0.4 ± 0.25	0.71 ± 0.56	1.06 ± 0.65
(*E,E*)-2,4-decadienal	25152–84-5	81,95,152,67	1288/-	MS	2.95 ± 0.04	2.97 ± 0.08	3.37 ± 0.34	0.05 ± 0.01	0.06 ± 0.02	0.04 ± 0.01
(*E,Z*)-2,6-nonadienal	557–48-2	41,70,94,69	1150/1155	MS RI	66.22 ± 3.83	64.37 ± 5.24	86.92 ± 14.74	1.15 ± 0.21	1.22 ± 0.31	1.02 ± 0.07
(*E,E*)-2,4-nonadienal	5910–87-2	81,138,95,67	1210/1213	MS RI STD	8.92 ± 10.36	26.47 ± 22.54	123.15 ± 68.78	0.15 ± 0.17	0.5 ± 0.59	1.45 ± 0.79
Decanal	112–31-2	57,82,95,112	1203/1200	MS RI STD	1.46 ± 2.42	0 ± 0	11.12 ± 9.05	0.03 ± 0.04	0 ± 0	0.13 ± 0.09
Heptaldehyde	111–71-7	70,81,55,96	896/899	MS RI STD	22.93 ± 8.56	21.24 ± 6.64	59.13 ± 22.31	0.4 ± 0.14	0.4 ± 0.13	0.7 ± 0.2
β-cyclocitral	432–25-7	152,137,123,109	1217/1218	MS RI STD	29.31 ± 4.65	21.27 ± 6.36	40.08 ± 8.4	0.51 ± 0.07	0.4 ± 0.11	0.47 ± 0.1
2,3-dihydro-2,2,6-trimethylbenzalhyde	116–26-7	107,121,150,91	1196/1196	MS RI	19.78 ± 3.66	10.16 ± 1.26	18.56 ± 2.74	0.34 ± 0.05	0.19 ± 0.05	0.22 ± 0.05
2-undecenal	2463–77-6	70,83,97,121	1361/1362	MS RI	15.12 ± 0.08	15.16 ± 0.11	15.54 ± 0.22	0.26 ± 0.05	0.29 ± 0.09	0.18 ± 0.03
1-cyclohexene-1-acetaldehyde	472–67-2	104,107,151,123	1253/1254	MS RI	7.04 ± 0.32	7.18 ± 0.92	9.71 ± 3.05	0.12 ± 0.02	0.14 ± 0.05	0.11 ± 0.05
trans-2-heptenal	18829–55-5	83,55,83,70	950/956	MS RI	5.47 ± 2.81	11.51 ± 5.54	25.73 ± 10.37	0.1 ± 0.05	0.22 ± 0.17	0.3 ± 0.11
Benzylcarboxaldehyde	122–78-1	91,120,92,65	1039/1040	MS RI STD	9.87 ± 18.83	3.34 ± 5.96	20.5 ± 10.23	0.17 ± 0.26	0.06 ± 0.08	0.24 ± 0.12
Phenols										
4-amino-3-methylphenol	2835–99-6	123,122,106,94	1045/-	MS	164.56 ± 50.11	66.69 ± 14.1	132.95 ± 47.7	2.86 ± 0.93	1.26 ± 0.42	1.56 ± 0.71
4-(2-butyl)phenol	99–71-8	121,150,93,109	1279/-	MS	38.22 ± 2.53	37 ± 0.95	42.69 ± 3.75	0.66 ± 0.13	0.7 ± 0.22	0.5 ± 0.13
2,4-di-*tert*-butylphenol	96–76-4	191,57,162,206	1509/1514	MS RI STD	0.19 ± 0.44	0.11 ± 0.28	0.26 ± 0.63	0 ± 0.01	0 ± 0.01	0 ± 0.01
Alcohols										
α-terpineol	98–55-5	121,136,93,81	1188/1188	MS RI STD	68.35 ± 19.5	254.28 ± 276.39	48.79 ± 60.26	1.19 ± 0.34	4.82 ± 3.59	0.57 ± 0.81
Geraniol	106–24-1	69,123,93,111	1251/1251	MS RI STD	40.92 ± 30.75	128.08 ± 251.68	39.99 ± 14.37	0.71 ± 0.43	2.43 ± 3.28	0.47 ± 0.13
1-octen-3-ol	3391–86-4	57,72,85,99	974/974	MS RI STD	27.41 ± 4.44	23.56 ± 3.97	44.71 ± 5.07	0.48 ± 0.05	0.45 ± 0.15	0.53 ± 0.11
Cedrol	77–53-2	95,150,135,119	1598/1598	MS RI STD	64.44 ± 55.53	106.33 ± 113.46	84.06 ± 43.35	1.12 ± 1.07	2.02 ± 2.27	0.99 ± 0.5
Linalool	78–70-6	93,121,71,136	1098/1098	MS RI STD	140.67 ± 74.52	109.3 ± 134.63	97.9 ± 55.73	2.44 ± 1.46	2.07 ± 1.89	1.15 ± 0.67
Phenethyl alcohol	60–12-8	91,122,92,65	1109/1108	MS RI STD	232.35 ± 66.59	230.19 ± 156.58	446.62 ± 208.42	4.04 ± 1.03	4.37 ± 1.97	5.25 ± 2.72
cis-1-p-menthanol	3901–95-9	71,98,123,141	1128/-	MS	5.04 ± 1.85	4.09 ± 0.86	7.26 ± 2.76	0.09 ± 0.05	0.08 ± 0.04	0.09 ± 0.05
(-)-*α*-cadinol	481–34-5	95,161,105,204	1656/1652	MS RI	0.73 ± 1.03	0.16 ± 0.26	0.54 ± 0.79	0.01 ± 0.02	0 ± 0	0.01 ± 0.01
(R,R)-(+)-hydrobenzoin	52340–78-0	107,108,79,77	1020/-	MS	20.57 ± 1.57	19.39 ± 6.11	18.4 ± 2.3	0.36 ± 0.06	0.37 ± 0.16	0.22 ± 0.04
2,6-dimethylcyclohexanol	5337–72-4	71,95,110,128	1105/-	MS	57.69 ± 13.78	38 ± 11.06	77.47 ± 12.79	1 ± 0.18	0.72 ± 0.31	0.91 ± 0.16
Ledol	577–27-5	58,107,182,95	1402/-	MS	12.39 ± 0.57	11.97 ± 0.27	12.79 ± 0.46	0.22 ± 0.04	0.23 ± 0.07	0.15 ± 0.03
t-cadinol	5937–11-1	161,95,204,121	1642/1647	MS RI	11.37 ± 1.9	9.95 ± 0.54	10.3 ± 0.39	0.2 ± 0.06	0.19 ± 0.05	0.12 ± 0.02
Linalool oxide (*trans*-pyranoid)	39028–58-5	68,94,151,59	1166/-	MS	62.29 ± 35.63	50.02 ± 22.94	47.01 ± 18.87	1.08 ± 0.45	0.95 ± 0.21	0.55 ± 0.16
Terpinen-4-ol	562–74-3	71,93,111,154	1174/1179	MS RI STD	17.16 ± 4.83	9.47 ± 3.13	10.7 ± 4.72	0.3 ± 0.08	0.18 ± 0.04	0.13 ± 0.06
2-ethylhexanol	104–76-7	57,98,112,83	1025/1026	MS RI	184.45 ± 148.43	196.17 ± 131.88	456.38 ± 387.04	3.2 ± 2.85	3.72 ± 3.23	5.37 ± 4.08
Benzyl alcohol	100–51-6	108,79,91,107	1029/1031	MS RI STD	522.72 ± 307.55	215.1 ± 180.57	517.89 ± 219.95	9.08 ± 4.09	4.08 ± 2.81	6.09 ± 2.14
Linalool oxide *I*	5989–33-3	59,94,111,68	1070/1068	MS RI	49.73 ± 12.63	46.54 ± 17.17	37.56 ± 7.71	0.86 ± 0.2	0.88 ± 0.09	0.44 ± 0.13
Linalool oxide *II*	34995–77-2	59,111,137,94	1086/1085	MS RI	79.4 ± 35.27	67.39 ± 41.48	55.33 ± 15.11	1.38 ± 0.58	1.28 ± 0.41	0.65 ± 0.17
Nerolidol	7212–44-4	69,81,93,107	1561/1569	MS RI STD	72.32 ± 98.35	7.9 ± 15.08	28.79 ± 21.15	1.26 ± 1.39	0.15 ± 0.32	0.34 ± 0.25

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Table 1 (*continued*)

Compounds	CAS	Quantitative ions (*m/z*)[a]	RI[b]/RI[c]	Identification[d]	Concentration (μg/L)			Proportion (%)		
					JHX	SJX	QJX	JHX%	SJX%	QJX%
Heptanol	111–70-6	70,56,69,98	964/969	MS RI STD	20.01 ± 2.36	21.3 ± 2.03	27.08 ± 4.43	0.35 ± 0.05	0.4 ± 0.13	0.32 ± 0.11
Pyrrole										
Tetramethylpyrazine	1124–11-4	136,54,42,95	1083/-	MS STD	31.66 ± 2.51	131.83 ± 166.84	186.31 ± 148.7	0.55 ± 0.08	2.5 ± 2.06	2.19 ± 1.34
Lactones										
Dihydroactinolide	17092–92-1	111,137,180,109	1525/1526	MS RI STD	81.5 ± 82.12	7.03 ± 13.12	128.46 ± 56.33	1.42 ± 1.08	0.13 ± 0.17	1.51 ± 0.59
γ-nonanolactone	104–61-0	85,119,162,91	1359/1360	MS RI	90.57 ± 43.11	60.18 ± 25.26	111.24 ± 64.52	1.57 ± 0.57	1.14 ± 0.79	1.31 ± 1.17
Others										
Caffeine	58–08-2	194,109,85,71	1846/1846	MS RI	1.12 ± 0.41	1.16 ± 0.47	1.38 ± 0.31	0.02 ± 0	0.02 ± 0.01	0.02 ± 0
Oleic acid	112–80-1	57,69,95,71	2172/2140	MS RI	11.54 ± 0	11.53 ± 0	11.54 ± 0	0.2 ± 0.04	0.22 ± 0.07	0.14 ± 0.02
Palmitic acid	57–10-3	73,129,57,83	1958/1963	MS RI	6.98 ± 2.34	5.33 ± 1.68	7.57 ± 2.03	0.12 ± 0.02	0.1 ± 0.03	0.09 ± 0.02
(+)-carotol	465–28-1	161,119,71,85	1768/-	MS	9.32 ± 0.22	9.46 ± 0.41	9.45 ± 0.23	0.16 ± 0.04	0.18 ± 0.05	0.11 ± 0.02

[a] Ions monitored for quantitation. The underlined ions were the quantified ones while the others were the identified ones.
[b] Retention index of compounds on HP-5MS.
[c] Retention index of compounds in reference.
[d] "MS" mass spetrum comparison using NIST17 library. "RI" retention index in agreement with literature value. "STD" confirmed by authenic standards.

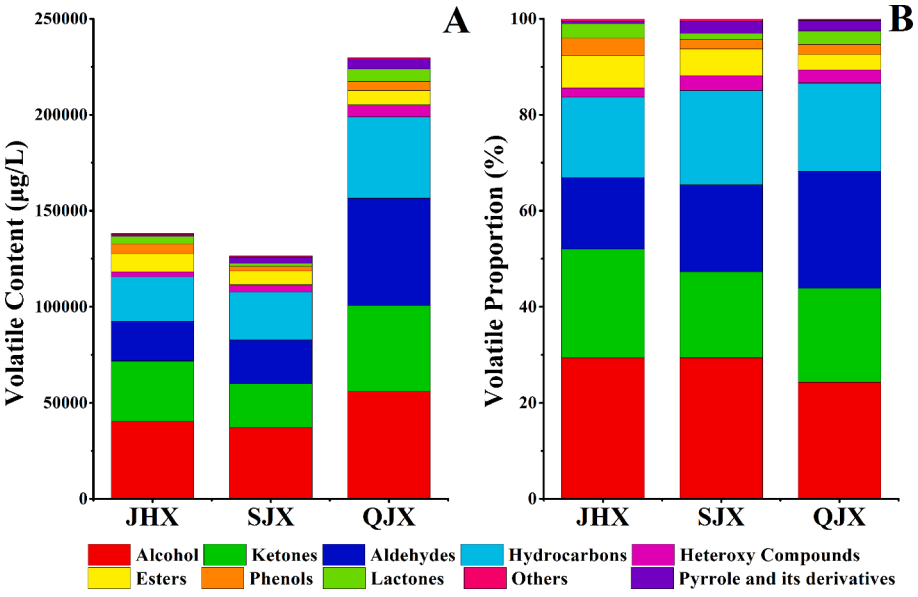

Fig. 2. Differences of the volatiles content (A) and proportion (B) in three aroma types of Fu brick tea. JHX: floral-fungal aroma type. SJX: ripe-fungal aroma type. QJX: fresh-fungal aroma type.

content in different tea samples, which was consistent with the PCA results (Fig. 3B). Overall, Fu brick teas could be divided into three groups based on their aroma types: JHX type (JHX-1 ~ JHX-8), SJX type (SJX-1 ~ SJX-8) and QJX type (QJX-1 ~ QJX-9). However, the absolute content of volatile compounds could not reflect the contribution of each compounds for the aroma characteristics formation in tea samples due to the absolute content of volatiles in QJX type samples was much higher than that in the other two types. Therefore, the percentage content of each volatile was used to explore the different volatile components in three types of Fu brick tea. Based on these results, three OPLS-DA models were established to investigate the differential volatiles contributing to distinguish different aroma types, including OPLS-DA model I (JHX type and SJX type), OPLS-DA models II (JHX type and QJX type) and OPLS-DA models III (SJX type and QJX type), respectively (Fig. S1). Two conditions need to be fulfilled for identifying the discriminatory volatiles in three aroma types Fu brick tea: the values of predictive component variable importance in the projection (VIP) ≥ 1 and p-value ≤ 0.05. Based on these criteria, 75 volatiles were identified as discriminatory volatile compounds (VIP > 1.0, $p < 0.05$) among the three groups. Obviously, the level of these discriminatory volatiles showed a dramatically different among the three aroma types of Fu brick tea (Fig. S2). In total, there were 29 discriminatory volatile compounds displayed the highest amount in JHX type samples, while 27 and 19 discriminatory volatile compounds had the highest content in SJX type and QJX type samples, respectively. Moreover, the higher level of discriminatory ketone and alcohol compounds in JHX type of Fu brick tea, including β-ionone, 3,5-dihydroxyacetophenone, ionone, benzyl alcohol, linalool, nerolidol, linalool oxide (*trans*-pyranoid), might cause

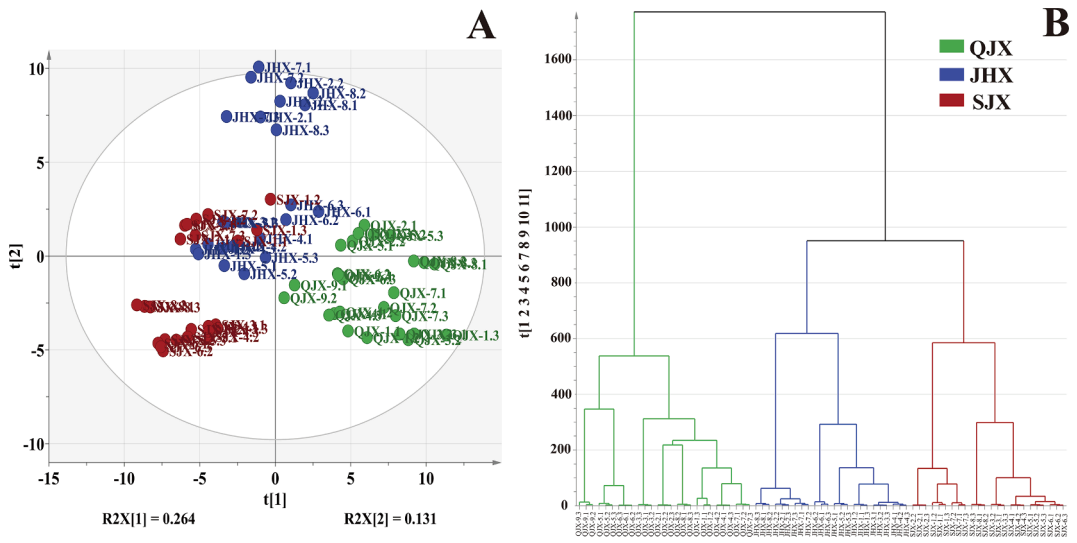

Fig. 3. PCA (A) and HCA (B) analysis based on the identified volatile compounds in different aroma types of Fu brick tea. JHX: floral-fungal aroma type. SJX: ripe-fungal aroma type. QJX: fresh-fungal aroma type.

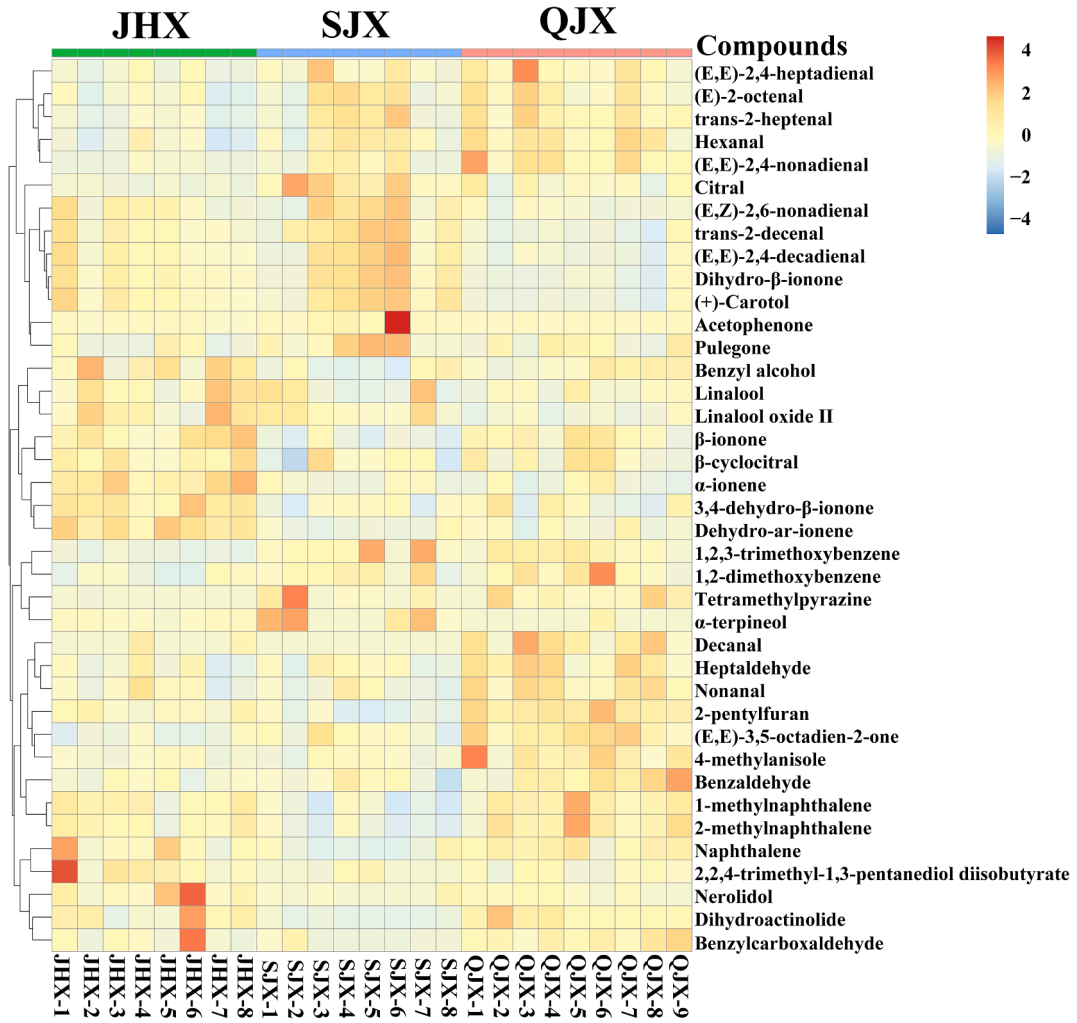

Fig. 4. Heatmap analysis for the discriminatory aroma compounds among the three aroma types of Fu brick tea. JHX: floral-fungal aroma type. SJX: ripe-fungal aroma type. QJX: fresh-fungal aroma type.

the aroma characteristics of JHX type of Fu brick tea differentiate from that in the other two aroma types (Huang et al., 2011; Lv et al., 2014; Shi et al., 2019). Also, the higher level of discriminatory ketone and aldehyde compounds in SJX type of Fu brick tea, such as acetophenone, dihydro-β-ionone, pulegone, (*E*,*Z*)-2,6-nonadienal, might contributed to the difference formation in the aroma characteristics of SJX type of Fu brick tea, as well as the higher level of discriminatory aldehyde compounds in QJX type of Fu brick tea, including hexanal, (*E*,*E*)-2,4-heptadienal, benzaldehyde, nonanal, (*E*,*E*)-2,4-nonadienal (Li et al., 2019; Li et al., 2020; Nie et al., 2019; Shi et al., 2019; Xu et al., 2007).

3.3. Differential aroma compounds in three aroma types of Fu brick tea

It is well known that the contribution of a volatile compound to the overall aroma of tea was not only depended on the concentration but also relied on its odor threshold. In our study, the odor descriptions and OAV values of volatile compounds were presented in Table S2, based on the reported references. Generally, there were 54 volatiles with OAV > 1 and 17 volatiles with OAV < 1. It has shown that the OAV value was directly proportional to the contribution degree of aroma (Liu, Zhou, & Xu, 2008). In this study, there are 28 aroma compounds with OAV > 10 were common to the three aroma types of Fu brick tea, including eight aroma compounds with floral attribute, nine with green attribute, four with woody attribute, two with fatty attribute, one with pungent attribute, two with stale attribute, one with earthy attribute and one with fruity attribute. All these aroma compounds provided important contribution to the basic aroma characteristics formation of Fu brick tea. Xu et al. (2007) reported that the compounds with floral and stale attributes in combination with some components of the raw material, which were together contributed to the 'fungal flower' aroma characteristics formation in Fu brick tea. However, some volatile compounds lacking OAV values might also contribute to the aroma characteristics formation of Fu brick tea. Fox example, (+)-β-cedrene and α-cedrene were found to be important compounds contributing to the woody flavour in Pu-erh tea (Xu et al., 2016; Lv et al., 2012). *t*-Cadinol was considered to have an important contribution to the formation of on the chestnut-like aroma in green tea (Wang, Hua, Jiang, Yang, Wang, & Yuan, 2020).

Combined with OPLS-DA results, 39 aroma compounds were considered as the discriminatory aroma compounds (VIP > 1, $p < 0.05$, OAV > 1) among the three aroma types of Fu brick tea (Fig. 4). For JHX type of Fu brick tea, there are 10 aroma compounds considered as discriminatory aroma compounds, including benzyl alcohol, linalool, linalool oxide *II* dehydro-*ar*-ionene, nerolidol, α-ionene, β-ionone, 2,2,4-trimethyl-1,3-pentanediol diisobutyrate, β-cyclocitral, 3,4-dehydro-β-ionone. The above discriminatory aroma compounds were presented at the highest level in the JHX type of Fu brick tea when compared with the other two aroma types. The aroma characteristics of JHX type of Fu brick tea was dominated by floral and fruity attributes, mixing with stale and woody attributes to form the floral-fungal aroma characteristics. Benzyl alcohol has the highest content in all volatile components of Fu brick tea, accounting for 6.4%. Also, previous studies reported that benzyl alcohol provided an important contribution to the floral attribute in Fu brick tea (Li et al., 2020; Wang et al., 2019). Linalool and linalool oxide II possessing typical floral attribute, could enhance the intense of floral attribute and decreased the intense of roasted attribute in instant white tea (Demyttenaere & Willemen, 1998). In addition, those two compounds were also identified as the important contributors to the 'fungal flower' aroma formation of Fu brick tea (Xu et al., 2007). Nerolidol is a sesquiterpene present in many teas, which was one of the most important factors to form the floral and fruity aroma in oolong tea and was also identified as the key contributor to the roses and apples aroma of black tea (Xu et al., 2016; Zhu et al., 2015). β-Ionone with floral attribute was derived from carotenoids, which played an important role in the aroma characteristic formation of many teas, such as Fu brick tea, due to its extremely low odor threshold (Li et al., 2020). Furthermore, β-cyclocitral, impart fruity attribute, which could be produced by the degradation of β-carotene, was also considered as one of the important compounds to form the 'fungal flower' aroma of Fu brick tea (Wang, Ma, Shi, Zhu, Lin, & Lv, 2020). Dehydro-*ar*-ionene presented with woody attribute, which was generated by the hydrolysis of aroma glycosides and contributed to the aroma characteristics formation in oolong tea (Guo, Ho, Wan, Zhu, Liu, & Wen, 2021).

For the SJX type of Fu brick tea, 11 aroma compounds presented the highest level compared with the other two aroma types were identified as discriminatory aroma compounds, including tetramethylpyrazine, α-terpineol, 1,2,3-trimethoxybenzene, pulegone, acetophenone, (*E*,*Z*)-2,6-nonadienal, dihydro-β-ionone, citral, *trans*-2-decenal, (+)-carotol, (*E*,*E*)-2,4-decadienal. Those compounds impart woody, stale and fatty attributes were found to dominate in SJX type of Fu brick tea, combining with those of which contributed to roasted, floral, fruity, green and mint scents to form the ripe-fungal aroma characteristics. α-Terpineol with woody note has been considered as an important aroma compound for the smoky or stale aroma characteristics in Pu-erh tea, which was hydrolyzed from glycoside precursors by microbial enzyme during the post-fermentation process (Lv, Zhong, Lin, Wang, Tan, & Guo, 2012). Dehydro-β-ionone with woody note was widely presence in tea, as the fermented product of β-ionone by fungal enzyme. It was also reported to be the key aroma compound for the 'aging fragrance' characteristics formation in the aging-storage Qingzhuan tea (Zhang et al., 2021). 1,2,3-trimethoxybenzene is well known as a typical stale compound that was considered to be the most important contributor to the stale characteristics of Pu-erh tea (Lv et al., 2014). It was transformed by fungal methylate action from gallic acid and the methylation of phenolic hydroxyl groups by *Aspergillus niger* during the pile-fermentation process (Lv et al., 2014). (*E*,*E*)-2,4-decadienal with fatty note was also considered as an important aroma compound in Chinese dark tea (Cao et al., 2018). Tetramethylpyrazine is a pyrazine compound with roasted attribute, which contributed to the characteristic aging fragrance formation of Chinese liquor and aged vinegar (Zhang, Si, Du, Li, Zhou, & Ye, 2020; Zhu, Qiu, & Li, 2016). Furthermore, acetophenone was mainly contributed to a floral note, which was identified as the basic aroma compounds of Fu brick tea (Li et al., 2020; Shi et al., 2019). Citral is a monoterpene aldehyde with fruity note that could be converted from carotenoids during the post-fermentation of tea, and it was considered to be a major constituent of essential oils in many lemon-scented aromatic plants (Dudai, Weinstein, Krup, Rabinski, & Ofir, 2005). (*E*,*Z*)-2,6-nonadienal was the key odorant in Chinese black teas and Darjeeling black tea, which could enhance green aroma in tea infusion (Chen et al., 2019; Greger & Schieberle, 2007). Pulegone was identified as the main aroma compound of peppermint that was usually used as herbal teas, spices and essential oil, due to its minty note and high relative odor intensity (Diaz Maroto, Castillo, Castro Vazquez, Gonzalez Vinas, & Perez Coello, 2007).

For the QJX type of Fu brick tea, there were 18 compounds considered as discriminatory aroma compounds, including hexanal, (*E*,*E*)-3,5-octadien-2-one, nonanal, dihydroactinolide, benzaldehyde, (*E*,*E*)-2,4-nonadienal, 1,2-dimethoxybenzene, (*E*,*E*)-2,4-heptadienal, (*E*)-2-octenal, heptaldehyde, (*E*)-2-heptenal, 4-methylanisole, 2-pentylfuran, benzylcarboxaldehyde, decanal, 2-methylnaphthalene, naphthalene, 1-methylnaphthalene. The highest content of these 18 compounds were found in the fresh-fungal aroma type of Fu brick tea compared with the other types. The aroma characteristics of QJX type of Fu brick tea was mainly formed by green attribute, combining with fruity, stale and pungent notes to form the fresh-fungal aroma characteristics. (*E*,*E*)-3,5-Octadien-2-one, (*E*,*E*)-2,4-heptadienal, (*E*,*E*)-2,4-nonadienal, benzylcarboxaldehyde and decanal exhibit a freshly cut grass smell and were all identified as the key aroma compounds to form green attribute of the post-fermentation dark tea in previous studies, such as Fu brick tea and Pu-erh tea (Cao et al., 2018; Li et al., 2020; Lv et al., 2012; Shi et al., 2019). Hexanal, (*E*)-2-octenal, 2-pentylfuran and benzylcarboxaldehyde also with typical green attribute was often reported to contribute to form

the characteristic green aroma of non-fermentation or full-fermentation tea, such as green tea and black tea (Kang et al., 2019; Wang et al., 2020; Zhu et al., 2018). Benzaldehyde is an important odor compound with fruity note, which often present in the chestnut-like aroma of green tea (Zhu et al., 2018). Naphthalene, 1-methylnaphthalene and 2-methylnaphthalene contributed with pungent and herbal-like notes, which could be degraded by microorganisms (Tanguler, Selli, Sen, Cabaroglu, & Erten, 2017). Naphthalene with pungent note, converted from long-chain hydrocarbons, was identified as the key odorant in yellow tea and Wuyi rock tea (Liu et al., 2021; Shi et al., 2021). 1-Methylnaphthalene also possessing pungent note was reported to contribute to the aroma of Liupao tea, which was usually used as a potential indicator to evaluate the degree of fermentation (Ma et al., 2020). 1,2-Dimethoxybenzene mainly contributed to a stale note, which was regard as important compound in the formation of the special flavor in Pu-erh tea (Lv et al., 2012).

3.4. Relationship between aroma compounds and sensory attributes in three aroma types of Fu brick tea

To explore the relationships among the aroma compounds (OAV > 1), sensory attributes and tea samples, PLS analysis was performed. Two latent variables were included in the PLS model, which represents 82.8% of X-matrix variance, explained 79.6% of Y-matrix variance. As shown in Fig. 5, samples appeared to be divided into three groups according to the different aroma types. Among them, floral-fungal aroma type of samples were mainly distributed at the positive value of PC1 and PC2, ripe-fungal aroma type of samples were mainly located at the positive values of PC1 and negative values of PC2, fresh-fungal aroma type of samples were mainly distributed at the negative value of PC1 and PC2. The first PC was defined by the aroma descriptors showing 'green' attribute on the negative dimension and 'herbal', 'floral and fruity', 'stale', 'woody' attributes on the positive dimension.

The 'green' attribute was positively correlated to 13 compounds including decanal (M45), nonanal (M16), heptaldehyde (M4), (*E,E*)-2,4-nonadienal (M23), benzylcarboxaldehyde (M44), benzaldehyde (M5), naphthalene (M20), 1-methylnaphthalene (M30), 2-methylnaphthalene (M46), (*E,E*)-3,5-octadien-2-one (M51), 2-pentylfuran (M43), 4-methylanisole (M53) and dihydroactinolide (M40). Previous study has shown that most of aldehydes were generally associated with 'green' attributes (Xu et al., 2007). The 'floral and fruity' attribute was significantly positive related to 3,4-dehydro-*β*-ionone (M38), dihydro-*β*-ionone (M36), (+)-carotol (M42) and linalool oxide *II* (M14). It is well known that ketone compounds are very important volatile compounds in various teas, providing a special floral attribute, and appear to be the unique volatile compounds in Fu brick tea (Lv et al., 2014). The 'woody' and 'stale' attributes were only significantly related to *α*-terpineol (M21). *α*-Terpineol is a tertiary monoterpenoid alcohol, which could be produced by many microorganisms and was also considered to be an important component of the basic aroma of dark tea (Nie et al., 2019; Sales, Felipe, & Bicas, 2020). In addition, it should be noted that 'herbal' attribute is located in the inner ellipse, which indicated that this attribute was not well described by the PLS model, perhaps due to the masking interactions with other attributes.

4. Conclusion

In summary, Fu brick tea could be divided into three aroma types by QDA analysis according to the differences of aroma characteristics, including floral-fungal aroma type, ripe-fungal aroma type and fresh-fungal aroma type. Among them, floral-fungal aroma type of Fu brick tea had strong 'floral and fruity' attribute, ripe-fungal aroma type with strong 'stale' and 'woody' aroma attributes, and green-fungal aroma type with strong significant 'green' attribute. A total of 112 volatile compounds were identified and quantified in Fu brick tea samples by HS-SPME/GC–MS analysis. According to OAV analysis, 54 voaltile compounds were identified as aroma compounds. Combined with OPLS-DA analysis, ten aroma compounds dominated with floral and fruity attributes in JHX type samples, eleven aroma compounds dominated with woody, stale and fatty attributes in SJX type samples, and eighteen aroma compounds dominated with green attribute in QJX type samples were identified as key aroma compounds that were responsible for the different aroma characteristics formation in the three aroma types of Fu brick tea. Furthermore, 3,4-dehydro-*β*-ionone, dihydro-*β*-ionone,

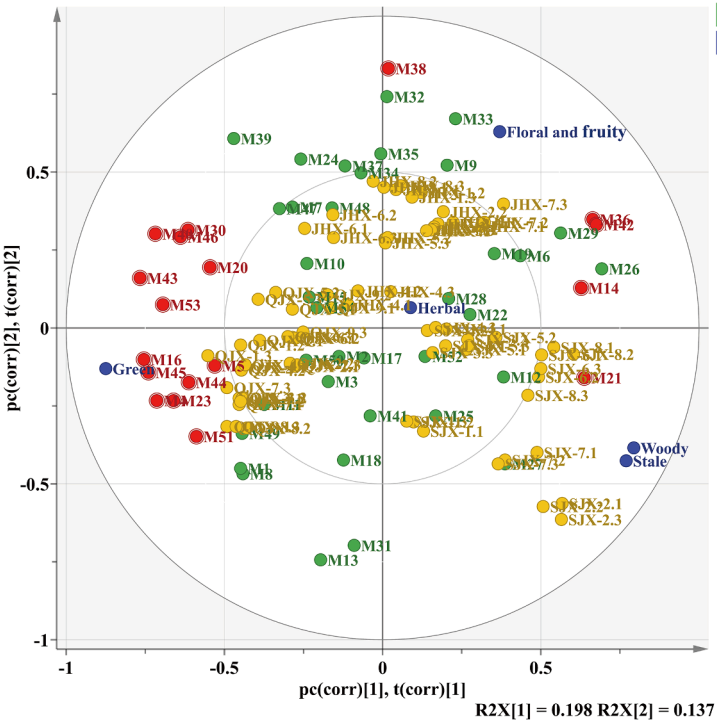

Fig. 5. PLS plots for the tea samples, the sensory attributes and aroma compounds (OAV > 1). M38: 3,4-dehydro-*β*-ionone. M36: dihydro-*β*-ionone. M42: (+)-carotal. M14: linalool oxide. M21: *α*-terpineol. M43: 2-pentylfuran. M45: decanal. M16: nonanal. M4: heptaldehyde. M40: dihydroactinolide. M53: 4-methylanisole. M23: (*E,E*)-2,4-nonadienal. M44: benzylcarboxaldehyde. M51: (*E,E*)-3,5-octadien-2-one. M46: 2-methylnaphthalene. M30: 1-methylnaphthalene. M20: naphthalene. M5: benzaldehyde.

(+)-carotol and linalool oxide *II* were identified as key contributors to 'floral and fruity' attribute, 'α-terpineol was contributed to 'woody' and 'stale' attributes, and thirteen aroma compounds that mainly composed of aldehydes were related to 'green' attribute by PLS analysis. This study provided the useful information for understanding the formation mechanisms of different aroma characteristic in Fu brick tea. Further research can focus on whether the volatile compounds with low OAVs have contribution to the aroma characteristics formation and the interaction of these aroma compounds in the different aroma types of Fu brick tea.

CRediT authorship contribution statement

Zheng Xuexue: Investigation, Visualization, Software, Writing – original draft. **Hong Xin:** Investigation, Visualization, Software, Writing – original draft. **Jin Youlan:** Investigation, Methodology, Software, Validation, Resources. **Wang Chao:** Validation, Writing – review & editing. **Liu Zhonghua:** Resources, Writing – review & editing. **Huang Jianan:** Conceptualization, Investigation, Writing – review & editing, Project administration, Funding acquisition. **Li Qin:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2022.100248.

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