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Comparative study of the volatile fingerprints of roasted and unroasted oolong tea by sensory profiling and HS-SPME-GC-MS

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ABSTRACT

Roasting plays important roles in shaping the volatile profile of oolong tea. In this study, the sensory attributes and volatile compositions of 153 roasted or unroasted oolong tea samples, belonging to four typical types, namely, High Mountain oolong tea (HMT), Tieguanyin tea (TGYT), Dongding oolong tea (DDT) and Wuyi rock tea (WRT), were studied in detail. Based on the sensory evaluation by tea evaluation experts, their respective sensory profiles were established and compared. Unroasted teas had more pronounced fresh and green flavors, while roasted teas had higher scores in pungent and caramel flavors. In particular, WRT demonstrated a unique fragrance of floral fruity flavors. By using HS-SPME-GC-MS analysis, a total of 128 compounds were identified across all samples. Notably, it was found that roasting largely increased the variety of volatile compounds in oolong tea. Furthermore, the characteristic volatile compounds of each type of tea were identified by PLS-DA modeling. Linalool and geraniol were the characteristic volatiles of HMT. Four volatiles, including *(E)*-nerolidol, jasmin lactone, benzeneacetaldehyde, and 4-methyl benzaldehyde oxime were identified as the characteristic volatiles of TGYT. Seven volatiles, including N-ethyl pyrrole, 3-(hydroxy methyl) pyridine, 4-pyridylcarbinol, 1-methyl pyrrole-2-carboxaldehyde, 2-ethyl-3,5-dimethyl pyrazine, 4-amino-2,3-xylenol, and 4,6-dimethyl pyrimidine were the characteristic volatiles of DDT. For WRT, 2,2,6-trimethyl cyclohexan-1-one, hexanoic acid, benzaldehyde, benzyl alcohol, β-cyclocitral, *(E)*-β-ionone, α-ionone, and octanoic acid were the characteristic volatiles. These findings expand our knowledge of the volatile fingerprints of oolong tea.

1. Introduction

Oolong tea, one of the six traditional Chinese tea categories, is a partially fermented tea type. It has a unique fruity and floral flavor, falling between unfermented green tea and fully fermented black tea (Dou et al., 2007). Many varieties of oolong teas exist in China, such as Wuyi Rock tea, Tieguanyin tea, Minnan Shuixian tea, High Mountain tea, Dongding tea, Oriental Beauty tea, Phoenix Dancong tea, Phoenix Shuixian tea, etc. These diverse oolong teas demonstrate distinctively different flavor profiles. For example, Wuyi rock tea (WRT), a top-ranking oolong tea, is famous for its characteristic "rock flavor" derived from a relatively high fermentation degree and a roasting process (Ho et al., 2015). Tieguanyin tea (TGYT), a popular light fermented and unroasted oolong tea, imparts floral and creamy notes. High Mountain tea (HMT), which is grown in the mountains of central Taiwan at altitude of over 1000 m above sea level, has floral and creamy notes. It undergoes a wide range of fermentation degree from 8% to 85% without further roasting. Dongding tea (DDT), similar to WRT, undergoes a robust roasting process after fermentation. It has a creamy, sweet, toasted nutty flavor. The divergent flavor properties, including aroma and taste, are rooted from many factors, such as the genotype, growth conditions, geographical origins, processing methods, and etc. Particularly, due to the distinctive manufacturing procedures conducted by experienced tea makers, the aroma and taste of oolong tea have almost unlimited possibilities (Ng et al., 2018).

The aroma of oolong tea is highly related to its chemical composition. Abundant volatile compounds, such as alcohols, aldehydes, hydrocarbons, and ketones, are associated with the aroma of oolong tea. A large number of studies have well documented the volatile fingerprints of different oolong tea varieties, such as Wang et al. (2022) found that

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among the 65 aroma-active compounds identified in WRT, 2-ethyl-3, 5-dimethylpyrazine and 6-methyl-5-hepten-2-one contributed the roasted and caramel-like aromas for WRT. Xu et al. (2018) identified 59 volatile compounds in TGYT, of which *(E)*-nerolidol, indole, and benzeneacetaldehyde were the most abundant compounds. Wang et al. (2008) detected 76 aroma compounds in DDT, showing a more complex volatile profile than those in green and black teas. It is noticed that the roasting plays critical roles in shaping the volatile fingerprints of oolong tea, such as WRT (Liu et al., 2022) and Qingxin oolong tea (Lan et al., 2022).

In addition to the characterization of volatile fingerprints of individual product, the comparison of multiple products from different varieties or geographical origins was also widely used to assess the flavor quality and authenticity of food. The direct comparison of different oolong tea varieties with the same experimental setup and analytical approach facilitates the comprehensive understanding of the similarities and differences among them. Volatile fingerprint comparison among different oolong tea varieties have been reported in several studies. For example, by employing identical manufacturing procedures, Guo et al. (2021a,b) compared the volatile profiles of roasted oolong tea made from three cultivars, including Shuixian, Huangmeigui and Zimudan. A total of 27 distinguishing volatiles were reported in this study. In another study, the volatile fingerprints of 75 oolong tea samples, belonging to five varieties (Tieguanyin, Benshan, Maoxie, Huangjingui and Jinguanyin) and with diverse prices, were established (Lin et al., 2013). A collection of 18 volatiles were identified as the most differentiated features. However, the information of manufacturing process of these teas was not reported in detail. To our knowledge, these comparative studies did not specifically focus on the effect of roasting in shaping the volatile fingerprints of oolong tea. It is of great academic, industrial and public interests to explore the role of roasting from the approach of comparing multiple products.

Aiming to further our understanding of the role of roasting and better characterize the aroma of different oolong teas, we selected two representative roasted oolong teas (WRT & DDT), and two representative unroasted oolong teas (TGYT & HMT) for comparative study of their volatile fingerprints. In order to provide more representative data, 153 oolong tea samples, including 44 HMT, 30 TGYT, 27 DDT, and 52 WRT, from multiple geographic locations and with different fermentation degrees were collected for analysis. Firstly, the sensory attributes of all oolong tea samples were evaluated by tea evaluation experts. Subsequently, head space – solid – phase microextraction – gas chromatography mass spectrometry (HS-SPME-GC-MS) was used to qualitatively and semi-quantitatively analyze the volatile compounds of the four types of oolong tea. Lastly, multivariate statistical analyses, including principal component analysis (PCA) and partial least squares - discriminant analysis (PLS-DA), were employed to identify their characteristic volatiles.

2. Materials and methods

2.1. Chemicals

Standards of linalool, geraniol, 2-ethyl-3,5-dimethylpyrazine, benzyl alcohol and benzaldehyde were purchased from Macklin Biochem (Shanghai, China); β-cyclocitral, benzeneacetaldehyde and jasmin lactone were purchased from Shanghai Yuanye Co., Ltd. (Shanghai, China); α-ionone, and *(E)*-β-ionone were purchased from CATO (Eugene, USA); and *(E)*-nerolidol was purchased from Bide Pharmatech Co., Ltd. (Shanghai, China). Internal standard of 2-octanol was purchased from Dr. Ehrenstorfer (Augsburg, GER). N-alkanes mixed standard (C_7–C_{40}) was purchased from O2SI smart solutions (Charleston, USA). Analytically pure sodium chloride (NaCl) was purchased from Sinopharm Chemical Reagent (Shanghai, CHN).

2.2. Tea samples

A total of 153 authentic oolong tea samples of four typical types, including 44 HMT, 30 TGYT, 27 DDT, and 52 WRT were collected. The details of these samples and collection locations were listed in Supplementary Material Table S1.

2.3. Sensory evaluation

The sensory attributes of all samples were evaluated by a sensory panel comprising of five tea evaluation experts who have been officially qualified as senior tea evaluators. The sensory evaluation process, including sensory description and scoring, was performed according to the Tea Sensory Evaluation National Standard of China (GB/T 23776-2018). In specific, 5 g of oolong tea samples were accurately weighted into a 110 mL tea bowl. Subsequently, 100 mL of boiling demineralized water was poured into the tea bowl and brewed for 2 min. The tea infusion was then transferred to an inspection bowl. The residue tea leaves were brewed for two more times with the same procedure, and the three infusions were combined for sensory evaluation by the sensory panel. Results of sensory evaluation in terms of aroma attribute descriptors and scorings were recorded.

2.4. Volatile compounds extraction

The volatile compounds of all oolong tea samples were extracted with a previously reported HS-SPME method (Liu et al., 2022). Briefly, 0.1 g powdered oolong tea sample, together with 1.5 g NaCl and 20 μL 2-octanol (10 mg/L, as internal standard) were placed into an 18 mL headspace vial. Then, 5 mL boiling demineralized water was added into the vial to brew the tea infusion for 5 min. The vial was sealed with a silicone septum. Subsequently, the vial was kept in a 50 °C water bath for 10 min. Then, SPME fibers were inserted into the headspace of the vial for the extraction of volatile compounds for 50 min in the water bath. Next, the fibers were inserted into the gas chromatograph injector and thermally desorbed at 250 °C for 5 min.

2.5. GC–MS analysis

The volatile compounds were analyzed by using an Agilent 7890A gas chromatograph (Agilent, Palo Alto, CA, USA) equipped with HP-INNOWAX column (30.0 m × 0.25 mm × 0.25 μm, Agilent, USA), and coupled to an Agilent 5975C mass spectrometer (Agilent, Palo Alto, CA, USA). The temperature of the injection port was set to 250 °C; the injection mode was selected as splitless injection. The carrier gas was high purity He (purity >99.999%), and the flow rate was set to 1 mL/min. The temperature program used was as follows: initial column temperature 40 °C for 5 min; temperature ramp at 3 °C/min to 120 °C; further temperature ramp at 6 °C/min to 240 °C and held at 240 °C for 5 min, the post-run temperature was 240 °C, and the post-run time was 5 min. The temperatures of the ion source and quadrupole were 230 °C and 150 °C, respectively. The ion energy for electron impact was 70 eV, and the mass spectra were obtained in full scan mode from 35 to 450 amu.

The mass spectrum corresponding to each chromatographic peak was compared with the NIST11.L standard spectral library, and the chromatographic peak was qualitatively analyzed by comparing with the NIST11.L standard spectral library and combining the retention index (RI, determined by n-alkanes C_7–C_{40}) method as defined by Vandendool and Kratz (1963). Volatile compounds were semi-quantitated by using the internal standard of 2-octanol. Additionally, the compounds that were considered as characteristic volatiles (see below) were further quantified based on the respective external calibration curves with authentic standards (supplementary materials Table S3).

2.6. Statistical analysis

The comparison of volatile compounds among different tea types and the identification of the representative volatile compounds of each tea type were conducted by using PCA and PLS-DA with R software (version 3.6.1), respectively. All data were expressed as mean ± SD of three replications. Graphing was performed by using GraphPad Prism software (Version 8.0.2).

3. Results and discussion

3.1. Sensory evaluation

As the first step, sensory evaluation, including the sensory description and sensory scoring, of the 153 oolong tea samples were conducted by the five tea evaluation experts. Results of the sensory evaluation were listed in Table S2. A total of 15 descriptors were used for the characterization of the tea aroma profiles. These descriptors and the scores were summarized in a radar chart as shown in Fig. 1. For HMT, the characteristic aromatic attributes were determined as floral, pure, tender, green, grassy and fruity. For TGYT, the commonly used descriptors were floral, pure, tender, green, thick, gloomy and grassy. For DDT, caramel and cloudy were considered as the dominant features. And for WRT, the characteristic aromatic attributes were floral, aging, orange peel, fruity and pungent. Notably, analogous odor profiles of TGYT and HMT were perceived, with comparable scores in fresh flavors, including floral, pure, tender, green and grassy. Among them, the fruity aroma of HMT was more obvious, while the green, thick and cloudy taste of TGYT was more prominent. In addition, small variations were shown within TGYT samples and also HMT samples, as reported by the panelists. With regard to WRT and DDT, generally higher sensory scores of floral, fruity, and roast flavors were perceived. The flavor variations of these samples were higher than those of TGYT and HMT samples, even for those from the same cultivars and geographic locations.

3.2. Analysis of aroma composition of oolong tea

Through GC-MS analysis, a total of 128 volatile compounds were detected in the 153 oolong teas. These volatile compounds consisted of 18 alcohols, 14 aldehydes, 5 acids, 7 hydrocarbons, 18 ketones, 41 heterocyclic compounds and 25 esters. Their RI, odor description and average contents in each type of tea were shown in Table 1. The odor descriptions were derived from Flavor and Extracts Manufacturers Association (FEMA) database or published literatures. The comparisons of the number and total content of each category of volatiles across all samples were demonstrated in Fig. 2. It was shown that alcohols, aldehydes, ketones, and esters were the dominant volatile categories of TGYT and HMT. For DDT and WRT, in addition to alcohols, aldehydes, ketones and ester, heterocyclic compounds were also abundantly found. It can be seen that the numbers and contents of heterocyclic compounds in roasted oolong tea were significantly higher than those in unroasted ones.

Based on the semi-quantitative information of each volatile, PCA was utilized for further comparison (Fig. 3). The variances of the first two principal components were 45.72% and 18.79%, respectively, accounting for 64.51% of the total variability of volatile components, which was sufficient to explain the information of volatile components in each tea sample. In general, the four types of tea can be well distinguished in PCA analysis. Among them, the difference between TGYT and HMT was relatively small, as the scatter plots from these two teas gathered more closely and distanced away from those from DDT and WRT ($P < 0.05$). Therefore, it is obvious that the roasting procedure significantly alter the volatile profiles of oolong tea. When comparing the volatile profiles of DDT and WRT, distinctive differences were shown. Moreover, large variations within each roasted teas were observed as well, which was in line with the results of sensory test. The reason for the large variations of the roasted oolong tea may partially due to the diverse roasting conditions performed. Indeed, Yang et al. (2021) also found that the aroma characteristics of WRT with medium and heavy roasting were similar, but significantly different from those with light roasting. The results from PCA scatter plot highlighted the significance of the roasting in shaping the volatile profiles of tea. Aiming to further elucidate the effect of roasting, more details regarding the differences of volatile categories between the roasted and the unroasted oolong teas were compared and discussed.

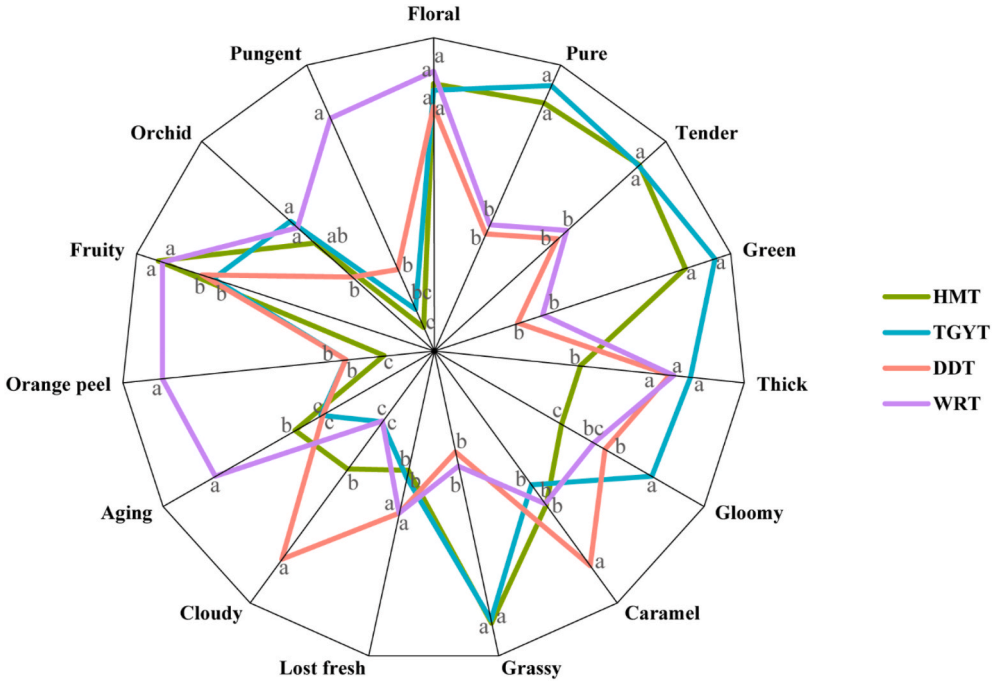

Fig. 1. Sensory description and scoring radar map of four types of oolong tea. Different letters of a–c indicate significantly different sensory scorings ($P < 0.05$).

Table 1
Concentration of volatiles in tea leaves of typical oolong tea.

NO.	Compounds	Odor description[a]	RI	Concentration (μg/g dry weight of tea leaves) [b]			
				HMT	TGYT	DDT	WRT
Alcohols							
1	4-Pyridylcarbinol	Hazelnut, coffee, nutty	1242	n.d.	n.d.	0.312 ± 0.389	n.d.
2	1-Pentanol	Pungent, solvent-like	1251	0.027 ± 0.027	0.005 ± 0.014	0.011 ± 0.057	0.008 ± 0.032
3	4-Pyridinemethanol	Hazelnut, coffee, nutty	1278	n.d.	n.d.	0.263 ± 0.461	n.d.
4	1-Hexanol	Pungent, sweet, green	1346	0.001 ± 0.005	n.d.	n.d.	0.048 ± 0.069
5	3-Hexen-1-ol	Green, leafy	1373	0.025 ± 0.049	n.d.	n.d.	0.005 ± 0.028
6	*(E)*-Linalool oxide	–	1425	0.278 ± 0.122	0.213 ± 0.106	0.887 ± 0.713	1.933 ± 0.823
7	1-Octen-3-ol	Earthy, green	1442	0.030 ± 0.038	0.083 ± 0.031	0.023 ± 0.058	0.188 ± 0.166
8	1-Decanol	Fatty, waxy	1444	0.002 ± 0.010	0.015 ± 0.032	0.001 ± 0.007	0.026 ± 0.094
9	*(Z)*-Linalool oxide	–	1454	0.221 ± 0.123	0.114 ± 0.073	0.512 ± 0.228	0.793 ± 0.337
10	Linalool	Citrus, floral	1539	1.154 ± 0.625	0.764 ± 0.255	0.486 ± 0.175	0.777 ± 0.543
11	1-Octanol	Waxy, green, orange	1543	0.016 ± 0.038	0.012 ± 0.038	n.d.	0.153 ± 0.121
12	Hotrienol	Fresh, floral, woody	1597	0.981 ± 0.599	0.666 ± 0.267	2.692 ± 1.442	2.153 ± 2.709
13	α-Terpineol	Pine, terpenic, lilac	1674	0.000 ± 0.001	n.d.	n.d.	0.026 ± 0.065
14	Nerol	Sweet, natural, neroli	1780	n.d.	n.d.	n.d.	0.005 ± 0.020
15	Geraniol	Sweet, floral, fruity	1830	0.645 ± 0.416	0.071 ± 0.121	0.235 ± 0.157	0.423 ± 0.456
16	Benzyl alcohol	Floral, rose, phenolic	1850	0.076 ± 0.049	0.070 ± 0.050	0.100 ± 0.049	0.319 ± 0.143
17	Phenylethyl alcohol	Floral, rose	1883	0.371 ± 0.395	0.585 ± 0.432	0.152 ± 0.119	0.728 ± 0.453
18	*(E)*-Nerolidol	Floral, green, citrus	2020	0.792 ± 0.624	3.471 ± 1.031	0.208 ± 0.201	0.785 ± 0.579
	Total alcohols			4.618 ± 1.616	6.068 ± 1.458	5.882 ± 2.049	8.371 ± 4.531
	Proportion			46.7% ± 14.1%	34.6% ± 4.1%	25.3% ± 10.0%	28.6% ± 11.8%
Aldehydes							
19	Hexanal	Fresh, green, fatty	–	0.076 ± 0.135	0.342 ± 0.169	0.027 ± 0.063	0.331 ± 0.314
20	2-Hexenal	Sweet, almond, bitter	1201	n.d.	0.000 ± 0.001	n.d.	0.035 ± 0.059
21	1-Methyl pyrrole-2-carboxaldehyde	–	1276	n.d.	n.d.	0.349 ± 0.425	n.d.
22	*(E,E)*-2,4-Heptadienal	Fatty, green, oily	1447	0.101 ± 0.113	0.529 ± 0.213	0.642 ± 0.787	0.537 ± 0.730
23	*(Z,E)*-2,4-Heptadienal	Fatty, green, oily	1474	0.119 ± 0.106	0.543 ± 0.235	0.454 ± 0.489	1.092 ± 0.684
24	Decanal	Sweet, aldehydic, waxy	1479	0.016 ± 0.044	0.023 ± 0.051	0.003 ± 0.011	0.013 ± 0.045
25	Benzaldehyde	Sharp, sweet, bitter	1496	0.155 ± 0.137	0.404 ± 0.202	0.399 ± 0.184	1.408 ± 0.556
26	*(E,Z)*-2,6-Nonadienal	Green, fatty, dry	1562	n.d.	n.d.	n.d.	0.011 ± 0.025
27	β-Cyclocitral	Tropical, saffron, herbal	1584	0.057 ± 0.058	0.146 ± 0.098	0.072 ± 0.085	0.347 ± 0.130
28	Benzeneacetaldehyde	Green, sweet, floral	1612	0.535 ± 0.673	1.881 ± 0.929	n.d.	0.102 ± 0.355
29	2-Hydroxy-5-methyl benzaldehyde	–	1753	n.d.	n.d.	n.d.	0.002 ± 0.012
30	2,4-Dimethylbenzaldehyde	Naphthyl, cherry, almond	1779	0.002 ± 0.005	n.d.	n.d.	0.001 ± 0.004
31	α-Ethylidene-benzeneacetaldehyde	–	1896	n.d.	0.001 ± 0.004	0.002 ± 0.009	0.023 ± 0.029
32	1H-Pyrrole-2-carboxaldehyde	Musty, beefy, coffee	1989	n.d.	n.d.	n.d.	0.040 ± 0.049
	Total Aldehydes			1.061 ± 0.934	3.870 ± 1.555	1.948 ± 1.171	3.942 ± 1.691
	Proportion			8.8% ± 5.2%	21.6% ± 5.0%	8.4% ± 5.1%	13.3% ± 4.0%
Acids							
33	Hexanoic acid	Sour, fatty, sweaty, cheesy	1588	n.d.	0.018 ± 0.068	0.018 ± 0.041	0.881 ± 0.497
34	Heptanoic acid	Sour, fatty, sweaty, cheesy	1929	n.d.	n.d.	n.d.	0.095 ± 0.083
35	*(E)*-3-Hexenoic acid	Fruity, honey, acidic	1935	n.d.	n.d.	n.d.	0.033 ± 0.087
36	2-Hexenoic acid	–	1944	n.d.	n.d.	n.d.	0.035 ± 0.090
37	Octanoic acid	Fatty, waxy, rancid	2034	n.d.	n.d.	n.d.	0.074 ± 0.060
	Total Acids			n.d.	0.018 ± 0.068	0.018 ± 0.041	1.117 ± 0.707
	Proportion			n.d.	0.1% ± 0.3%	0.1% ± 0.2%	3.8% ± 2.1%
Hydrocarbons							
38	p-Xylene	–	1120	n.d.	n.d.	0.266 ± 0.248	0.479 ± 0.632
39	Limonene	Citrus, orange, fresh, sweet	1158	0.001 ± 0.007	n.d.	n.d.	0.034 ± 0.098
40	1-Methyl cycloheptene	–	1183	n.d.	n.d.	0.002 ± 0.009	0.002 ± 0.011
41	*(E)*-β-Ocimene	Sweet, herbal	1231	0.028 ± 0.050	n.d.	0.015 ± 0.046	n.d.
42	Vinylcyclohexane	–	1449	0.012 ± 0.035	n.d.	n.d.	n.d.
43	Azulene	–	1698	n.d.	n.d.	0.008 ± 0.041	n.d.
44	α-Farnesene	Floral, woody	1732	0.301 ± 0.485	0.601 ± 0.461	0.050 ± 0.176	0.023 ± 0.065
	Total hydrocarbons			0.342 ± 0.527	0.601 ± 0.461	0.341 ± 0.255	0.537 ± 0.637
	Proportion			2.3% ± 2.9%	3.4% ± 2.7%	1.4% ± 1.2%	1.7% ± 1.4%
Ketones							
45	2-Octanone	Earthy, weedy, natural	1269	0.000 ± 0.002	n.d.	n.d.	0.045 ± 0.049
46	2,2,6-Trimethyl cyclohexan-1-one	Pungent, thujonic, labdanum	1286	0.006 ± 0.015	0.001 ± 0.005	0.003 ± 0.008	0.181 ± 0.096
47	2,3-Octanedione	Dill, asparagus, cilantro	1309	0.005 ± 0.018	0.003 ± 0.017	n.d.	n.d.
48	2-Methyl-3-octanone	–	1318	0.007 ± 0.023	n.d.	n.d.	n.d.
49	6-Methyl-5-hepten-2-one	Citrus, green, musty	1326	0.158 ± 0.135	0.516 ± 0.513	0.094 ± 0.184	0.749 ± 0.702
50	3-Octen-2-one	Earthy, spicy, herbal	1387	0.002 ± 0.011	n.d.	n.d.	0.007 ± 0.024
51	3,4-Dihydroxyacetophenone	–	1486	n.d.	n.d.	n.d.	0.078 ± 0.150
52	3,5-Octadien-2-one	Fruity, fatty, mushroom	1498	0.079 ± 0.067	0.225 ± 0.113	0.289 ± 0.283	0.513 ± 0.364
53	*(E,E)*-3,5-Octadien-2-one	Fruity, fatty, mushroom	1547	0.037 ± 0.158	0.017 ± 0.016	0.040 ± 0.040	0.160 ± 0.099
54	3,5,5-Trimethyl-2-cyclopenten-1-one	–	1571	0.003 ± 0.015	n.d.	0.002 ± 0.012	n.d.
55	Acetophenone	Sweet, pungent, hawthorn	1620	0.003 ± 0.013	0.000 ± 0.002	0.094 ± 0.090	0.220 ± 0.201
56	2,6,6-Trimethyl-2-cyclohexene-1,4-dione	Musty, woody, sweet	1662	n.d.	n.d.	n.d.	0.005 ± 0.023
57	1-Phenyl-1-propanone	Hawthorn, lilac, floral	1690	n.d.	n.d.	0.010 ± 0.026	0.184 ± 0.204
58	α-Ionone	Sweet, woody, floral	1818	0.004 ± 0.008	0.023 ± 0.023	0.012 ± 0.012	0.064 ± 0.036

(*continued on next page*)

Table 1 (*continued*)

NO.	Compounds	Odor description[a]	RI	Concentration (μg/g dry weight of tea leaves) [b]			
				HMT	TGYT	DDT	WRT
59	*(E)*-β-Ionone	Dry, powdery, floral	1904	0.038 ± 0.048	0.095 ± 0.151	0.063 ± 0.096	0.389 ± 0.170
60	*(Z)*-Jasmone	Woody, herbal, floral	1908	0.167 ± 0.136	0.302 ± 0.139	0.164 ± 0.138	0.033 ± 0.101
61	4-(2,2,6-Trimethyl-7-oxabicyclo hept-1-yl)-3-buten-2-one	Fruity, sweet, berry	1959	0.006 ± 0.014	0.032 ± 0.033	0.002 ± 0.006	0.061 ± 0.046
62	5-Methyl-2-(1-methylethyl)-2-cyclohexen-1-one	–	2137	0.001 ± 0.004	0.002 ± 0.011	n.d.	0.004 ± 0.016
	Total ketones			0.514 ± 0.287	1.217 ± 0.533	0.771 ± 0.459	2.692 ± 1.340
	Proportion			5.1% ± 2.7%	7.0% ± 2.7%	3.4% ± 2.2%	9.1% ± 3.1%
Heterocyclic compounds							
63	N-Ethyl pyrrole		1169	0.018 ± 0.070	n.d.	2.485 ± 1.228	0.547 ± 0.668
64	3-(Hydroxymethyl) pyridine	Bitter, green	1245	n.d.	n.d.	0.371 ± 0.381	0.002 ± 0.017
65	4,6-Dimethyl pyrimidine	–	1302	n.d.	n.d.	0.101 ± 0.089	0.033 ± 0.081
66	2,5-Dimethyl pyrazine	Cocoa, roasted, nutty	1302	n.d.	n.d.	0.046 ± 0.097	n.d.
67	2,6-Dimethyl pyrazine	Burnt, almond, roasted	1304	0.002 ± 0.011	n.d.	0.117 ± 0.131	0.114 ± 0.141
68	2-Ethyl pyrazine	Peanut, butter, musty	1315	n.d.	n.d.	0.058 ± 0.079	0.057 ± 0.098
69	4-Methyl-1,3-benzenediamine	–	1365	n.d.	n.d.	0.006 ± 0.019	n.d.
70	2-Ethyl-3-methyl pyrazine	Nutty, peanut, musty	1366	n.d.	n.d.	0.045 ± 0.051	0.014 ± 0.057
71	2-Ethyl-5-methyl pyrazine	Coffee, beany, nutty	1371	0.005 ± 0.024	n.d.	0.288 ± 0.130	0.249 ± 0.175
72	4,5-Dihydro-5,5-dimethyl-4-isopropylidene-1H pyrazole	–	1375	n.d.	n.d.	n.d.	0.091 ± 0.174
73	2,6-Dimethyl-3-pyridinamine	–	1384	n.d.	n.d.	0.041 ± 0.056	0.062 ± 0.104
74	Furfural	Sweet, woody, almond	1448	n.d.	n.d.	0.277 ± 0.335	0.771 ± 0.917
75	3-Ethyl-2,5-dimethyl pyrazine	Potato, cocoa, roasted, nutty	1428	n.d.	n.d.	0.078 ± 0.278	n.d.
76	2,5-Diethyl pyrazine	Nutty, hazelnut	1437	n.d.	n.d.	0.019 ± 0.032	0.000 ± 0.002
77	2-Ethyl-3,5-dimethyl pyrazine	Burnt, almond, caramel, roasted	1440	n.d.	n.d.	0.056 ± 0.087	n.d.
78	3,5-Diethyl-2-methyl pyrazine	–	1473	0.002 ± 0.011	n.d.	0.077 ± 0.127	n.d.
79	2-Acetylfuran	Sweet, balsamic, almond	1484	0.003 ± 0.015	n.d.	0.083 ± 0.051	0.105 ± 0.085
80	2,5-Dimethyl-3-(2-methyl propyl) pyrazine	–	1503	n.d.	n.d.	0.002 ± 0.009	0.007 ± 0.018
81	5-Methyl furfural	Spicy, caramellic, maple	1552	0.004 ± 0.029	n.d.	0.298 ± 0.144	0.562 ± 0.451
82	Benzonitrile	–	1574	n.d.	n.d.	0.013 ± 0.047	n.d.
83	1-Ethyl-1H-pyrrole-2-carboxaldehyde	Burnt, roasted, smoky	1581	0.102 ± 0.198	0.015 ± 0.029	5.096 ± 2.153	2.887 ± 1.601
84	1-Methyl-1H-pyrrole-2-carboxaldehyde	–	1591	n.d.	n.d.	0.005 ± 0.018	0.042 ± 0.057
85	2-Methyl-4-methoxyaniline	–	1613	0.025 ± 0.110	n.d.	1.930 ± 0.913	1.147 ± 0.616
86	Phthalan	–	1623	0.019 ± 0.050	0.110 ± 0.154	n.d.	0.029 ± 0.207
87	1,2-Dihydro-1,5,8-trimethyl naphthalene	–	1707	n.d.	n.d.	0.045 ± 0.019	0.082 ± 0.084
88	Tetrahydro-2,2,6-trimethyl-6-vinyl-2H pyran-3-ol	Floral, honey	1716	0.072 ± 0.104	0.096 ± 0.099	0.006 ± 0.022	0.152 ± 0.208
89	4-Amino-2,3-xylenol	–	1721	0.010 ± 0.069	n.d.	0.657 ± 0.713	0.207 ± 0.351
90	6-Ethenyl tetrahydro-2,2,6-trimethyl-2H pyran-3-ol	Floral, honey	1747	0.006 ± 0.023	0.000 ± 0.003	n.d.	0.035 ± 0.062
91	2-Methyl-benzenemethanamine	–	1789	0.002 ± 0.009	n.d.	0.009 ± 0.027	0.019 ± 0.043
92	2-Cyclopropyl-benzenamine	–	1795	n.d.	n.d.	0.001 ± 0.006	0.033 ± 0.081
93	1-Furfuryl pyrrole	Plastic, green, waxy	1801	0.002 ± 0.011	n.d.	0.207 ± 0.086	0.165 ± 0.099
94	Benzyl nitrile	–	1894	0.631 ± 0.663	0.498 ± 0.273	0.667 ± 0.349	2.206 ± 1.320
95	2-Acetyl pyrrole	Musty, nut, skin, cherry	1940	0.006 ± 0.025	n.d.	0.133 ± 0.049	0.181 ± 0.075
96	4-(2,6,6-Trimethylcyclohexa-1,3-dienyl) but-3-en-2-one	–	1970	n.d.	n.d.	n.d.	0.010 ± 0.020
97	Phenol	Phenolic, plastic, rubbery	1978	n.d.	n.d.	0.001 ± 0.005	0.016 ± 0.026
98	3-Methyl phenol	–	2052	n.d.	n.d.	n.d.	0.005 ± 0.014
99	4-Methyl benzaldehyde oxime	–	2262	0.016 ± 0.046	0.048 ± 0.045	n.d.	n.d.
100	3,5-bis (1,1-Dimethylethyl) phenol	–	2283	0.001 ± 0.004	0.004 ± 0.008	n.d.	0.007 ± 0.012
101	Eugenol	Sweet, spicy, clove, woody	2309	0.015 ± 0.022	0.012 ± 0.014	0.004 ± 0.010	0.014 ± 0.030
102	2,3-Dihydro benzofuran	–	2352	n.d.	n.d.	0.027 ± 0.017	0.016 ± 0.019
103	Indole	Animal, floral, naphthyl, fecal	2396	2.679 ± 1.922	4.132 ± 1.260	0.892 ± 0.545	0.543 ± 0.386
	Total heterocyclic compounds			3.620 ± 2.598	4.916 ± 1.480	14.142 ± 5.551	10.411 ± 4.177
	Proportion			30.6% ± 10.5%	27.7% ± 4.4%	55.4% ± 11.7%	35.1% ± 9.5%
Esters							
104	*(Z)*-3-Hexenoic acid methyl ester	Fruity	1244	0.001 ± 0.004	n.d.	n.d.	0.141 ± 0.283
105	2-Methyl-propanoic acid hexyl ester	Fruity, green, apple	1394	0.002 ± 0.008	n.d.	n.d.	0.018 ± 0.044
106	Formic acid heptyl ester	Green, waxy, floral	1445	0.007 ± 0.025	0.008 ± 0.025	n.d.	0.005 ± 0.025
107	*(Z)*-3-Hexenyl-α-methyl butyrate	–	1457	0.001 ± 0.004	0.006 ± 0.013	0.003 ± 0.014	0.003 ± 0.016
108	2-Furanmethanol acetate	Sweet, fruity, banana	1522	0.000 ± 0.001	n.d.	0.007 ± 0.012	0.020 ± 0.027
109	2-Furoatemethyl	Fruity, mushroom, fungal	1559	n.d.	n.d.	0.072 ± 0.071	0.179 ± 0.208
110	Hexanoic acid hexyl ester	Herbal, fresh, green	1588	0.012 ± 0.027	0.001 ± 0.004	n.d.	0.076 ± 0.114
111	Caproic acid hexenyl ester	Fruity, apple, pear	1640	0.094 ± 0.072	0.066 ± 0.063	0.087 ± 0.065	0.186 ± 0.137
112	*(E)*-Hexanoic acid-2-hexenyl ester	Green, natural, cognac	1650	0.002 ± 0.010	n.d.	0.002 ± 0.011	0.034 ± 0.065
113	Methyl 5-hydroxynicotinate	–	1657	n.d.	n.d.	0.101 ± 0.149	0.091 ± 0.140
114	Methyl 2-oxo-1,2-dihydro-3-pyridinecarboxylate	–	1659	0.004 ± 0.015	n.d.	0.059 ± 0.117	0.027 ± 0.057
115	γ-Hexalactone	Herbal, coconut, sweet	1669	0.009 ± 0.014	0.002 ± 0.009	0.022 ± 0.023	0.007 ± 0.019
116	Acetic acid phenylmethyl ester	Sweet, floral, fruity	1703	0.000 ± 0.001	n.d.	0.012 ± 0.031	0.099 ± 0.131
117	Methyl phenylacetate	Sweet, floral, honey	1736	n.d.	n.d.	0.084 ± 0.172	0.217 ± 0.185
118	Methyl salicylate	Wintergreen, minty	1745	0.197 ± 0.147	0.204 ± 0.055	0.852 ± 0.516	0.998 ± 0.671
119	Formic acid 2-phenylethyl ester	Rose, green, hyacinth	1760	n.d.	n.d.	n.d.	0.005 ± 0.021

(*continued on next page*)

Table 1 (*continued*)

NO.	Compounds	Odor description[a]	RI	Concentration (μg/g dry weight of tea leaves) [b]			
				HMT	TGYT	DDT	WRT
120	Methyl 3-amino-4-hydroxybenzoate	–	1784	n.d.	n.d.	0.006 ± 0.012	0.003 ± 0.018
121	Acetic acid 2-phenylethyl ester	Floral, rose, sweet	1787	n.d.	0.001 ± 0.007	0.004 ± 0.017	0.093 ± 0.084
122	Phenethyl butyrate	Musty, sweet, floral	1933	0.023 ± 0.042	0.026 ± 0.038	0.009 ± 0.022	0.020 ± 0.067
123	2-Phenylethyl 2-methylbutyrate	Floral, green, sweet	1941	0.005 ± 0.019	0.032 ± 0.066	n.d.	0.003 ± 0.021
124	Benzoic acid 2-phenylethyl ester	Rose, balsamic, honey, floral	2081	0.188 ± 0.249	0.303 ± 0.184	0.011 ± 0.035	0.193 ± 0.208
125	*(Z)*-3-Hexen-1-ol benzoate	Fresh, green, leafy	2092	0.012 ± 0.048	n.d.	0.005 ± 0.018	0.006 ± 0.028
126	Hexadecanoic acid methyl ester	Oily, waxy, fatty, orris	2193	0.000 ± 0.003	n.d.	0.001 ± 0.004	0.000 ± 0.002
127	Jasmin lactone	Creamy, waxy, jasmin	2218	0.164 ± 0.090	0.381 ± 0.120	0.175 ± 0.082	0.143 ± 0.087
128	Dihydroactinidiolide	Ripe, apricot, fruity	2294	0.006 ± 0.010	0.008 ± 0.010	0.005 ± 0.009	0.067 ± 0.050
	Total esters			0.726 ± 0.427	1.038 ± 0.384	1.518 ± 0.872	2.633 ± 1.709
	Proportion			6.5% ± 1.8%	5.8% ± 1.0%	6.0% ± 2.7%	8.3% ± 2.9%
	Total			10.881 ± 5.562	17.728 ± 4.478	24.620 ± 7.055	29.703 ± 10.036

[a] The odor descriptions were from Flavor and Extracts Manufacturers Association (FEMA) database.
[b] n.d. means the compound was not detected in tea samples.

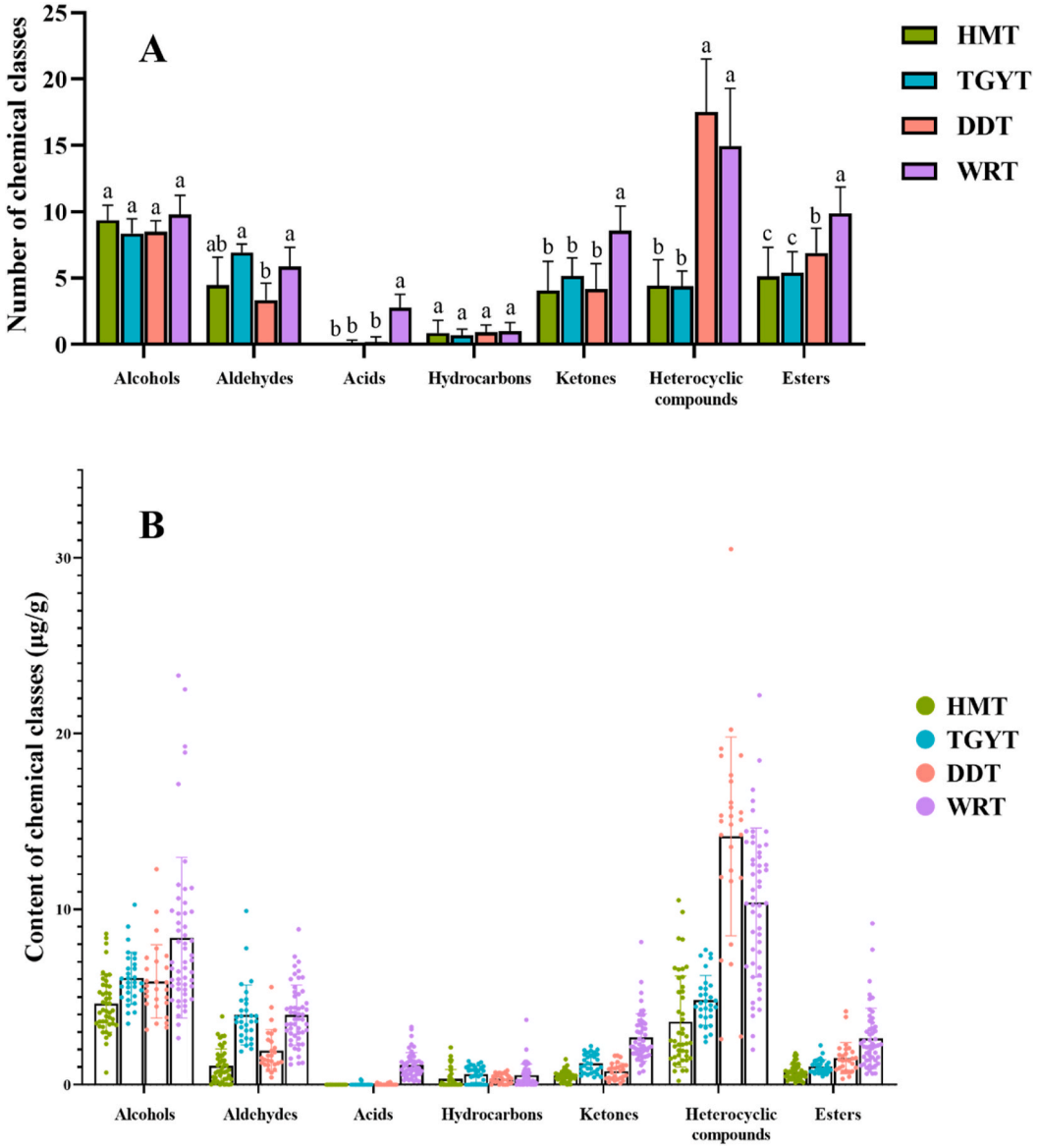

Fig. 2. The number (A) and content (B) of chemical classes in oolong tea, including HMT, TGYT, DDT, and WRT. Different letters of a–c indicate significantly different numbers of chemical classes ($P < 0.05$).

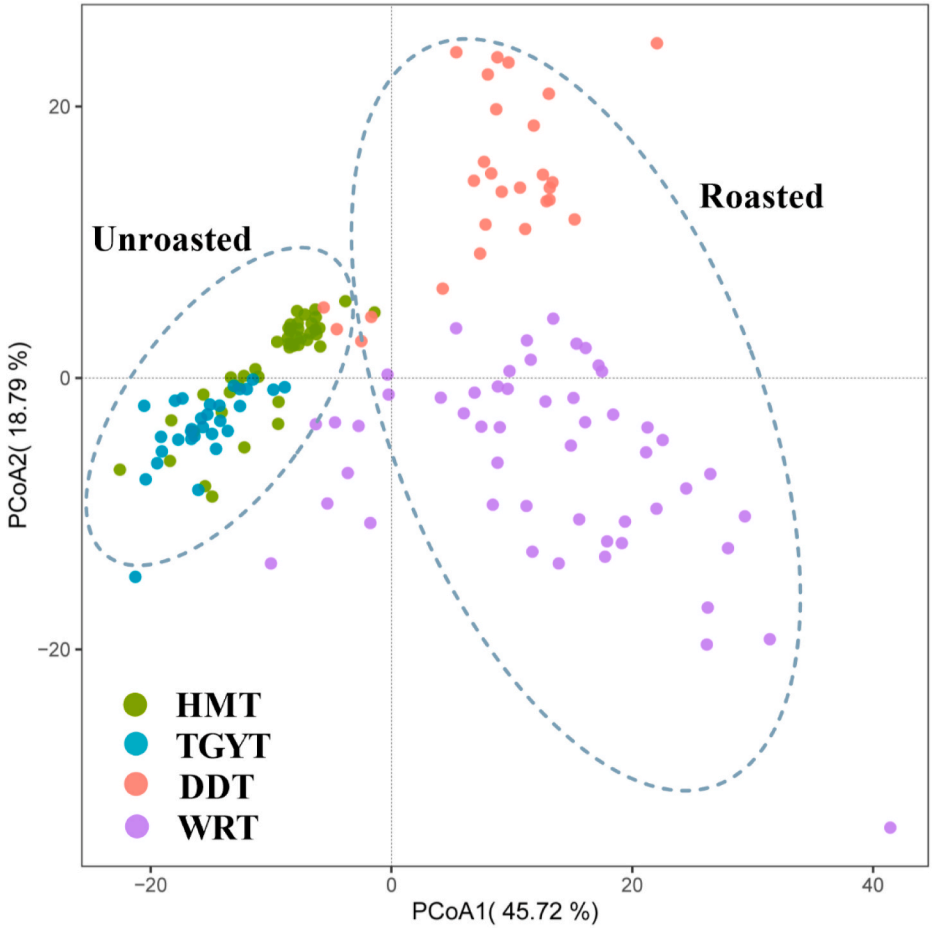

Fig. 3. Principal component analysis (PCA) of the volatile compounds in tea leaf samples from the typical oolong tea.

3.2.1. Alcohols

The main alcohol compounds in all oolong tea samples were linalool and its oxides, geraniol, hotrienol, phenylethyl alcohol, *(E)*-nerolidol (Table 1), which had typical floral and fruity aromas. Most of these alcohol compounds have been reported to exist in many other teas, such as white tea (Huang et al., 2021; Lin et al., 2021), yellow tea (Shi et al., 2021), and Pu-erh tea (Du et al., 2019). The alcohol compounds in the unroasted oolong teas accounted for the largest proportion (46.7% for HMT and 34.6% for TGYT). In roasted teas, lower proportion of alcohols were found. Similarly, as reported by Liu et al. (2022), after roasting, almost all the alcohol compounds except linalool oxide were reduced in WRT. Due to the lower boiling point, alcohols may evaporate during the high temperature roasting. On the contrary, higher linalool oxide content were found in the roasted samples, which may be related to the oxidation of linalool and its glycoside derivatives during the roasting process (Ho et al., 2015).

3.2.2. Aldehydes and acids

The main aldehyde compounds in oolong tea included β-cyclocitral, hexanal, *(E,E)*-2,4-heptadienal, benzaldehyde, and benzeneacetaldehyde (Table 1). The content of aldehydes in unroasted tea was higher than that in roasted tea (15.2% vs 10.8%). The possible reasons might be the evaporation of some low-boiling points aldehydes and the involvement into various reactions due to the presence of active carbonyl group. Adversely, the contents of a small amount of aldehydes with fruity and fatty odors, such as benzaldehyde and *(E,E)*-2,4-heptadienal, increased gradually, which was consistent with the results reported by Liu et al. (2022). These aldehydes may derive from the thermal degradation of lipids during the roasting. Five short chain acids, including hexanoic acid, heptanoic acid, *(E)*-3-hexenoic acid, 2-hexenoic acid, octanoic acid, were detected in this study. Notably, WRT had much higher amount of acid compounds than other teas (Table 1). Kuo et al. (2011) reported that long term storage facilitated the decarboxylation of long chain acids to form shorter chain products. In this study, most of HMT, DDT, and TGYT samples were collected in 2021, with relatively short storage time prior to determination, whereas WRT were all collected in 2020, which explained their higher acid contents.

3.2.3. Hydrocarbons

The hydrocarbon content in oolong tea was relatively low, with *(E)*-β-ocimene and α-farnesene as the most abundant ones (Table 1). Chen et al. (2013) found that the contents of main linear and branched hydrocarbons in freshly made tea leaves decreased significantly after 5 years of storage, which may due to the oxidation and degradation upon unsaturated bonds. Therefore, low amount of hydrocarbons in these finished tea samples, which have been stored for one to three years, were somehow reasonable. α-Farnesene, a sesquiterpene volatile compound with floral and woody flavors, was the common hydrocarbon detected in all four kinds of oolong tea. Its content in unroasted tea was much higher than that in roasted tea, reaching approximately 6–26 times of that in roasted oolong tea. In line with our results, Yang et al. (2021) also reported the extremely low content of α-farnesene in WRT with different roasting degrees, which may be related to the degradation of this compound during roasting.

3.2.4. Ketones

As showed in Table 1, 6-methyl-5-hepten-2-one, 3,5-octadien-2-one, β-ionone, *(Z)*-jasmone were the major ketones detected in all samples

studied. The content of ketones in WRT (2.692 μg/g) was significantly higher than that in other teas (0.514–1.217 μg/g). Ketones were derived from the degradation of carotenoids or unsaturated fatty acids, or the hydrolysis from their glycoside precursors (Ho et al., 2015; Zhu et al., 2015). They are also important intermediate products of Maillard reaction, playing critical roles in the formation of some downstream Maillard reaction products like pyrrolidones (Ho et al., 2015). Higher ketone content in WRT than those in other teas may be associated with the high contents of ketone precursors, or the more extensive roasting process.

3.2.5. Heterocyclic compounds

A number of heterocyclic compounds were detected in this study, mainly comprising pyrazines and pyrroles. Significantly higher amount of heterocyclic compounds was detected in the roasted teas than that in the unroasted ones. They are the most abundant volatile category in DDT and WRT (55.4% in DDT and 35.1% in WRT). Heterocyclic compounds are well known as the Maillard reaction products (Ho et al., 2015), therefore it is expected that higher amount of heterocyclic compounds would be detected in roasted teas. Conversely, indole, a common pyrrole with flowery note, demonstrated much higher amount in the unroasted teas. Yang et al. (2021) also reported the low content of indole in WRT, and it decreased with the increase of roasting degree. Indole is mainly derived from tryptophan by indole-lyase (Ho et al., 2015), while the roasting may inactivate this enzyme and terminate the conversion.

3.2.6. Esters

The ester compounds in the four oolong tea types mainly included methyl phenylacetate, methyl salicylate, jasmine lactone, and caproic acid hexenyl ester, which contributed floral and fruity flavors to oolong teas (Table 1). The total contents of esters ranged from 5.8% to 8.3% in the four oolong tea types. No significant difference in esters among all samples. It may be due to the fact that esters had relatively higher boiling points and were more stable during roasting (Ho et al., 2015). The only exception was methyl salicylate, which was significantly higher in roasted tea than that in unroasted tea. Liu et al. (2022) also reported an increasing trend of this compound during tea roasting.

3.3. Analysis of characteristic volatile compounds of different types of oolong tea

In order to further explore the characteristic volatile compounds of each type of oolong tea, four PLS-DA models based on different grouping methods were established (Figs. 4–7). The details regarding the approach and the results were discussed in the following sections.

3.3.1. The characteristic volatile compounds of HMT

For the identification of the characteristic volatile compounds of HMT, all tea samples were first divided into HMT group and control group that included all the rest tea samples. Then, based on this grouping method, PLS-DA model was constructed. Thereafter, the contribution of each volatile compound in distinguishing the two groups was evaluated according to its VIP value obtained from PLS-DA model.

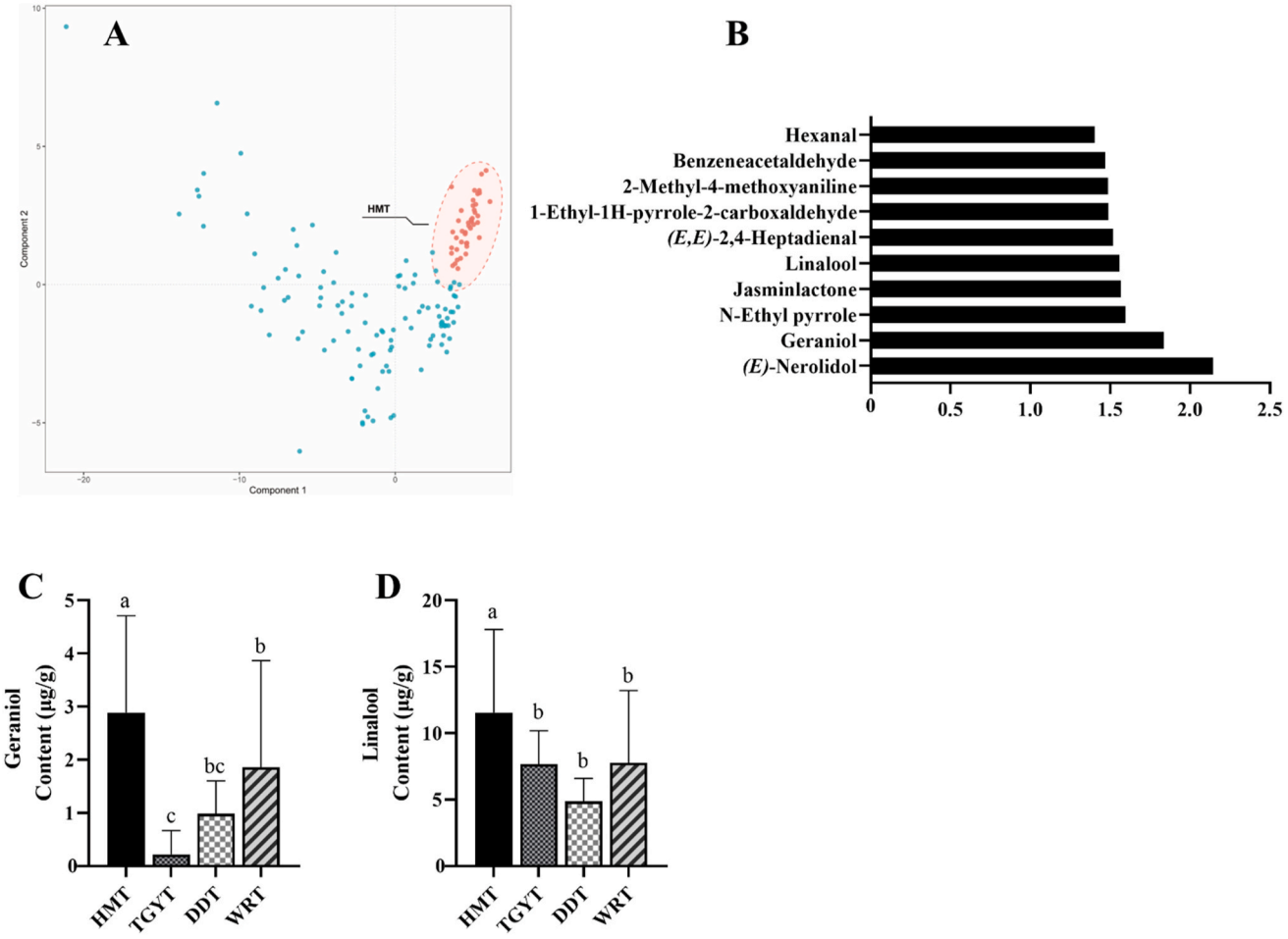

Fig. 4. Comparison of the volatile compounds in HMT and other types of oolong tea by using PLS-DA analysis (A); volatile compounds with the top 10 highest VIP values in PLS-DA modeling (B); the concentrations of geraniol (C) and linalool (D) in the four types of oolong tea. Different letters of a–c indicate significantly different concentrations ($P < 0.05$).

Finally, the volatiles with high VIP value and significantly higher content in HMT group were considered as the characteristic volatile compounds of HMT. As demonstrated in Fig. 4A, clear separation between HMT and the control was obtained from PLS-DA modeling. The top 10 volatiles with the highest VIP values, including *(E)*-nerolidol, geraniol, linalool, benzeneacetaldehyde, etc., were listed in Fig. 4B. Among them, the contents of linalool and geraniol in HMT were higher than those in the rest teas (Fig. 4C and D). Due to the potent floral, fruity and sweet scent, linalool and geraniol were have been reported as the key aromatic compounds responsible for the unique odor of oolong tea (Lücker et al., 2001; Nagegowda et al., 2008). Adversely, as reported by Guo et al. (2021a,b), linalool and geraniol in WRT decreased with the process of fermentation and roasting. Therefore, the processing of tea may have negative effect for the reservation of these two compounds. As the light processing tea type, it is reasonable to detect relatively higher amount of linalool and geraniol in HMT as shown in Fig. 4C and D.

3.3.2. The characteristic volatile compounds of TGYT

Following the same approach, the PLS-DA model for TGYT was constructed. The resulting scatter plots and volatiles with high VIP values were demonstrated in Fig. 5A and B. Of the compounds with high VIP values, *(E)*-nerolidol, jasmine lactone, benzeneacetaldehyde and 4-methylbenzaldehyde oxime in TGYT were higher than those in the rest teas (Fig. 5C–F). In the "turn over" step of TGYT, tea leaves are continuously inverted and repeatedly kneaded, which induce the mechanical damage of tea leaves and promote the degradation of lipids to form jasmine lactone (Zeng et al., 2018). Due to the strong fatty-fruity peach and apricot flavor, jasmine lactone was reported to play an important role in the overall aroma of unroasted tea (Zeng et al., 2018). At high temperature of roasting, jasmine lactone could be converted to other jasmonic acid derivatives (Katsuno et al., 2014). Regarding *(E)*-nerolidol and benzeneacetaldehyde, Xu et al. (2018) also identified these two compounds as the key aroma compounds in fresh shoots of TGYT. Both *(E)*-nerolidol, a volatile sesquiterpene that provides a typical floral and fresh flavor (Zhu et al., 2018), and benzeneacetaldehyde, an aldehyde with honey-like, sweet, rose, green, grassy aroma, are sensitive to high temperature. For example, it was reported that following the roasting process of WRT, the content of benzeneacetaldehyde decreased significantly (Liu et al., 2022). The formation of *(E)*-nerolidol was closely related to the presence of *(E)*-nerolidol synthase (Zhou et al., 2017). This enzyme was highly expressed during the turn over and fermentation stages of oolong tea. Due to the generally higher fermentation degree of TGYT than HMT (Chen et al., 2013), higher amount of *(E)*-nerolidol and benzeneacetaldehyde were therefore expected.

3.3.3. The characteristic volatile compounds of DDT

The characteristic volatile compounds in DDT were also investigated by using PLS-DA modeling. The resulting scatter plot and volatiles with high VIP values were shown in Fig. 6A and B. Due to the high VIP values and significantly higher contents, seven volatiles, including N-ethyl pyrrole, 3-(hydroxy methyl) pyridine, 4-pyridylcarbinol, 1-methyl pyrrole-2-carboxaldehyde, 2-ethyl-3,5-dimethyl pyrazine, 4-amino-2,3-xylenol and 4,6-dimethyl pyrimidine, were recognized as the

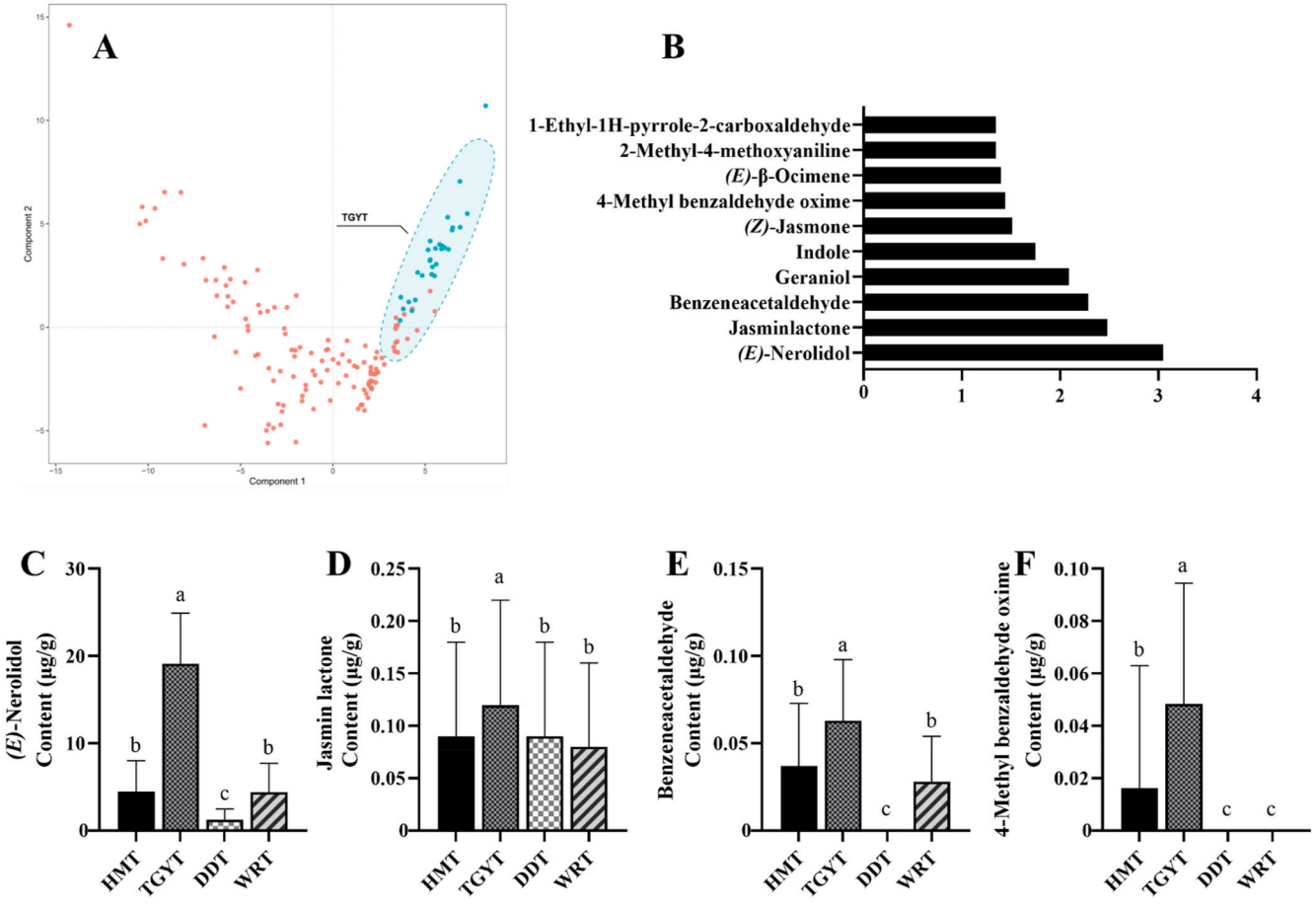

Fig. 5. Comparison of the volatile compounds in TGYT and other types of oolong tea by using PLS-DA analysis (A); volatile compounds with the top 10 highest VIP values in PLS-DA modeling (B); the concentrations of *(E)*-nerolidol (C), jasmin lactone (D), benzeneacetaldehyde (E), and 4-methyl benzaldehyde oxime (F) in the four types of oolong tea. The concentration of 4-methyl benzaldehyde oxime was calculated based on 2-octanol. Different letters of a–c indicate significantly different concentrations ($P < 0.05$).

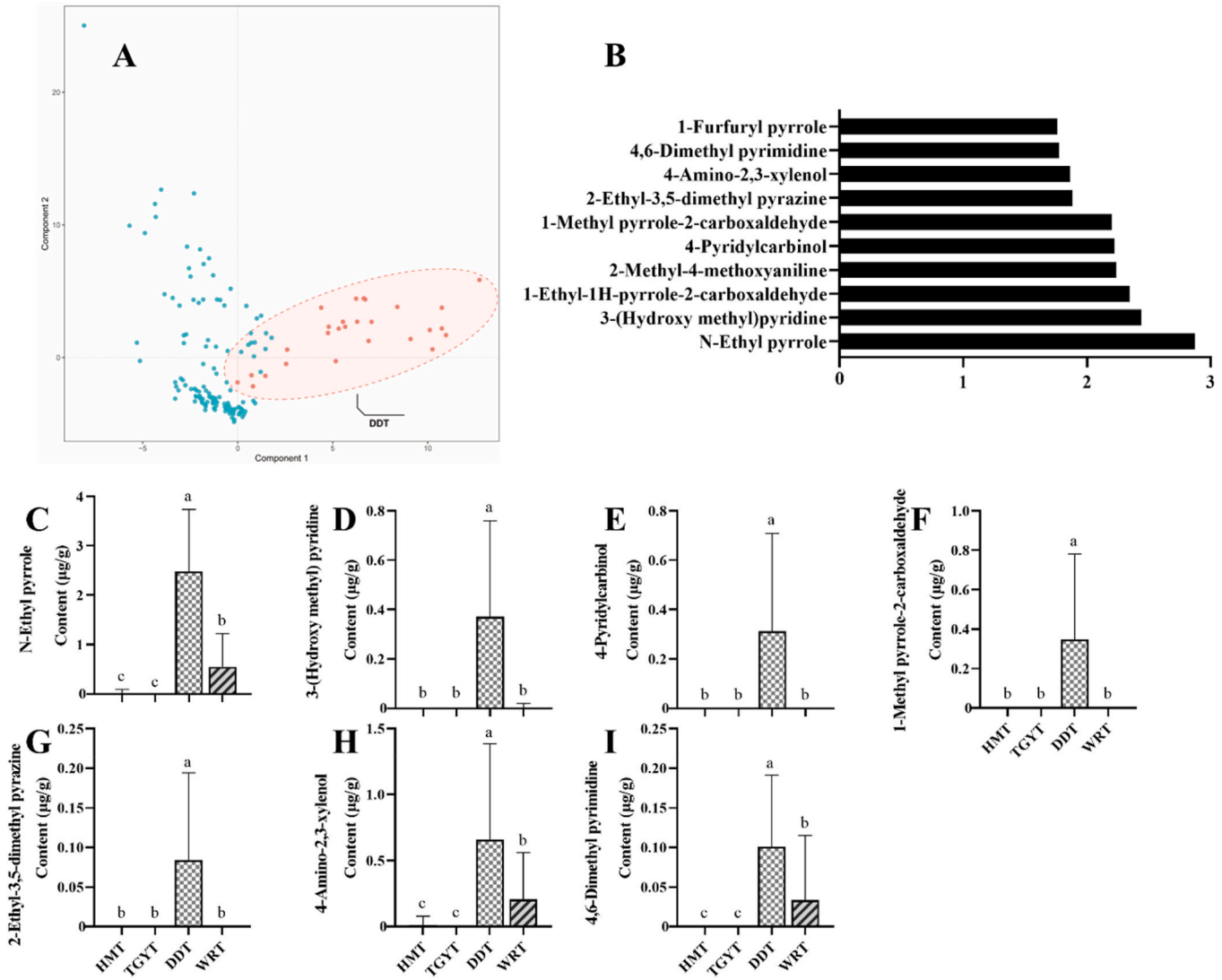

Fig. 6. Comparison of the volatile compounds in DDT and other types of oolong tea by using PLS-DA analysis (A); volatile compounds with the top 10 highest VIP values in PLS-DA modeling (B); the concentrations of N-ethyl pyrrole (C), 3-(hydroxy methyl) pyridine (D), 4-pyridylcarbinol (E), 1-methyl pyrrole-2-carboxaldehyde (F), 2-ethyl-3,5-dimethyl pyrazine (G), 4-amino-2,3-xylenol (H), and 4,6-dimethyl pyrimidine (I) in the four types of oolong tea. The concentration of N-ethyl pyrrole, 3-(hydroxy methyl) pyridine, 4-pyridylcarbinol, 1-methyl pyrrole-2-carboxaldehyde, 4-amino-2,3-xylenol, and 4,6-dimethyl pyrimidine were calculated based on 2-octanol. Different letters of a–c indicate significantly different concentrations ($P < 0.05$).

characteristic volatile compounds of DDT (Fig. 6C–I). Different from TGYT and HMT, the characteristic volatile compounds in DDT mainly showed a roasted scent. Except for 4-amino-2,3-xylenol, most of these compounds belong to pyrroles, pyrazines and pyridines, which were derived from Maillard reaction during the roasting process. Due to the low odor threshold, these compounds were also considered as the key aroma compounds of several roasted teas. Taking 2-ethyl-3,5-dimethyl pyrazine as an example, it was recognized as important aromatic-active compound in roasted Qingxin oolong tea (Lan et al., 2022). Similar to the "turn over" step in TGYT processing, there is a so-called "cloth ball-rolling" process for DDT to make it into spherical or semi-spherical shapes (Su et al., 2021). This tea leaves tissue injury process may promote the release of some glycoside bounded compounds, such as linalool oxide and phenethyl alcohol (Hu et al., 2018).

3.3.4. *The characteristic volatile compounds of WRT*

Interestingly, WRT demonstrated quite different volatile profiles from those of DDT, as revealed by PLS-DA modeling (Fig. 7A and B). The characteristic volatile compounds in WRT were mainly comprised of ketones, aldehydes, alcohols, such as 2,2,6-trimethyl cyclohexan-1-one, hexanoic acid, benzaldehyde, benzyl alcohol, β-cyclocitral, *(E)*-β-ionone, α-ionone and octanoic acid (Fig. 7B). Despite the higher variations among the collected 52 WRT as aforementioned, they commonly presented higher concentrations of these compounds as compared with other teas (Fig. 7C–J). WRT are frequently associated with fruity and floral notes. Indeed, the odor attributes of these volatile compounds are fruity and floral notes. Taken benzaldehyde as an example, it is the most abundant aldehyde in WRT, accounting for 35.7% of the total content of aldehyde compounds. It demonstrates fruity, sweet and sharp flavors. The high content of benzaldehyde in WRT was also observed by Yang et al. (2021). The high amount of fruity and floral ketones, aldehydes and alcohols may associate with the unique geographical and environmental condition in Wuyi Mountain area (Zeng et al., 2020). It is well known that tea grown and made in Wuyi Mountain area has premium quality due to the altitudinal, climatical and soil factors, which may facilitate the formation of these fruity and floral compounds (Ng et al., 2018). Notably, the typical Maillard reaction products, such as pyrroles, pyrazines and pyridines, were not recognized as the most characteristic volatile compounds in WRT, suggesting an overwhelming contribution of fruity and floral compounds to the overall aroma of WRT.

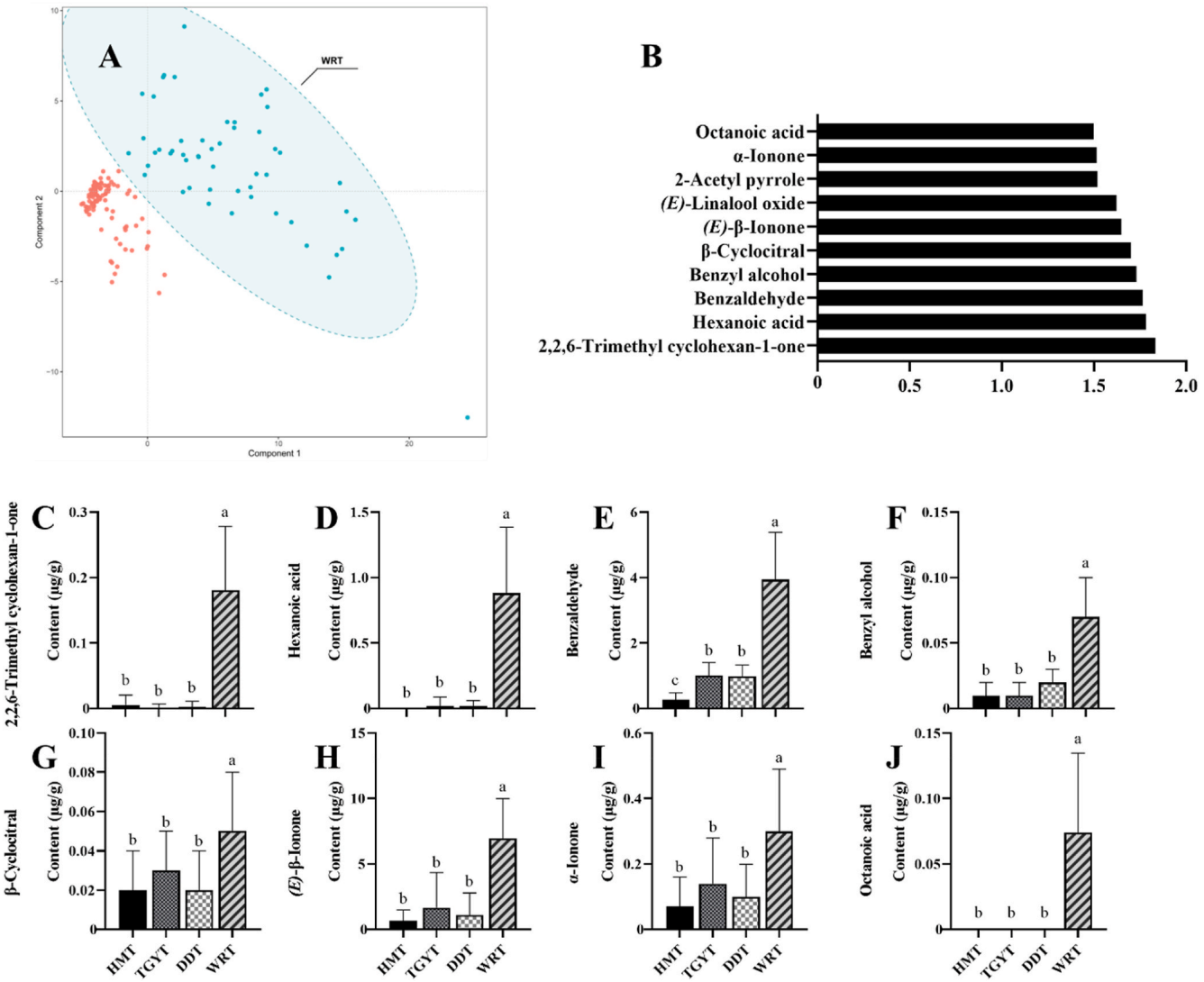

Fig. 7. Comparison of the volatile compounds in WRT and other types of oolong tea by using PLS-DA analysis (A); volatile compounds with the top 10 highest VIP values in PLS-DA modeling (B); the concentrations of 2,2,6-trimethyl cyclohexan-1-one (C), hexanoic acid (D), benzaldehyde (E), benzyl alcohol (F), β-cyclocitral (G), *(E)*-β-ionone (H), α-ionone (I), and octanoic acid (J) in the four types of oolong tea. The concentration of 2,2,6-trimethyl cyclohexan-1-one, hexanoic acid and octanoic acid were calculated based on 2-octanol. Different letters of a–c indicate significantly different concentrations ($P < 0.05$).

4. Conclusions

In this study, the volatile fingerprints, including the sensory profiles and volatile compositions, of four typical oolong teas were characterized comprehensively. A total of 128 volatile compounds were identified in the four types of oolong tea, with alcohols and heterocyclic compounds as the most abundant volatile categories. Distinctively different volatile profiles of roasted and unroasted oolong teas were demonstrated in this study. It was also found that the roasting process largely increase the variations of volatiles in tea. Furthermore, the characteristic volatile compounds of each oolong tea type were investigated in detail. Further studies, such as aroma recombination test, are required to further confirm their contribution to the overall aroma of tea. Nevertheless, these results provide useful information for better understanding the flavor of these four popular teas.

CRediT authorship contribution statement

Daoliang Wang: Conceptualization, Methodology, Validation, Investigation, Formal analysis, Writing – original draft, Visualization. **Zhibin Liu:** Methodology, Validation, Writing – review & editing, Supervision. **Wensong Chen:** Methodology, Investigation. **Xiaoye Lan:** Visualization, Investigation. **Sijia Zhan:** Validation, Investigation. **Yaqian Sun:** Formal analysis, Data curation. **Weiying Su:** Formal analysis, Validation. **Chih-Cheng Lin:** Sample collection, Investigation. **Li Ni:** Methodology, Validation, Writing – review & editing, Supervision, Project administration.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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