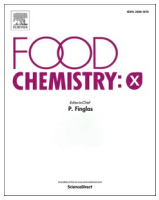
Food Chemistry: X 18 (2023) 100730

Contents lists available at ScienceDirect

Food Chemistry: X

journal homepage: www.sciencedirect.com/journal/food-chemistry-x



The aroma characteristics of oolong tea are jointly determined by processing mode and tea cultivars

Chang He [a], Jingtao Zhou [a], Yuchuan Li [a], De zhang [a], Bernard Ntezimana [a], Junyu Zhu [a], Xiaoyong Wang [a], Wenluan Xu [a], Xiaoju Wen [a], Yuqiong Chen [a,b,c], Zhi Yu [a,b,c], Yu Wang [a,b,c,*], Dejiang Ni [a,b,c,*]

[a] National Key Lab for Germplasm Innovation and Utilization of Horticultural Crops, College of Horticulture & Forestry Sciences, Huazhong Agricultural University, Wuhan, Hubei 430070, People's Republic of China
[b] Key Laboratory of Urban Agriculture in Central China, Ministry of Agriculture, Wuhan, Hubei 430070, People's Republic of China
[c] Hubei Hongshan Laboratory, Wuhan, 430070, People's Republic of China

ARTICLE INFO

Keywords:
Oolong tea
Aroma attribute
Tea processing mode
Tea cultivars
Molecular sensory technology

ABSTRACT

This study delved into the aroma characteristics of "Qingxiang" oolong tea, analyzing six different cultivars and their processing modes. The findings showed that both cultivars and processing modes have a significant impact on the oolong tea aroma system. The study identified 18 terpenoid volatiles (VTs), 11 amino-acid-derived volatiles (AADVs), 15 fatty-acid-derived volatiles (FADVs), 3 carotenoid-derived volatiles (CDVs), and 10 other compounds in oolong tea that differentiate it from green and black tea. The turn-over stage was found to be the primary processing stage for oolong tea aroma formation. Molecular sensory analysis revealed that the "fresh" odor attribute is the basis for its aroma, while "floral and fruity" fragrances are its aroma characteristics. The perception of oolong tea as "fresh" and "floral and fruity" is influenced by the interactions of its aroma components. These findings provide a new basis for breed improvement and process enhancement in oolong tea production.

Introduction

Oolong tea, which is predominantly produced in China, has gained worldwide recognition for its alluring fragrance of flowers and fruits and its numerous health benefits. The aroma of oolong tea is a crucial qualitative characteristic that directly influences its market value (Zeng, Zhou, Su, & Yang, 2020). The aroma characteristics of oolong tea were reported to be inextricably linked to the selection of cultivars and processing methods (Feng et al., 2019; Zeng, Zhou, Su, & Yang, 2020).

Early researchers investigated the aroma difference among oolong tea, black tea, and green tea based on commercially available teas, but they found it difficult to determine the cause of aroma variance due to their difference in origin, tenderness, cultivars, and processing. Specifically, 15 black teas, 27 green teas, 8 white teas, 27 oolong teas, and 9 Pu-erh teas were purchased in the market as the research subjects (K. Wang et al., 2011), and six volatile and two non-volatile components were identified using cluster analysis to distinguish the tea properties. In a recent study, researchers utilized the simultaneous distillation–extraction (SDE) method and gas chromatography/time-of-flight mass spectrometry (GC-TOF-MS) technology to analyze a total of 33 tea samples, including 9 black teas, 12 green teas, and 12 oolong teas obtained from the market. Through their analysis, the researchers were able to identify a total of 74 distinct differential chemicals present in the various tea samples. (Zhang, Zeng, Zhao, Kong, Lu, & Xu, 2013). In a recent study, researchers conducted an in-depth analysis of the aromatic components found in the six major types of tea—black tea, green tea, oolong tea, dark tea, yellow tea, and white tea—produced by "Longjing 43″. To achieve this, they utilized advanced technologies such as headspace-solid phase microextraction (HS-SPME), solid-phase extraction (SPE), solvent-assisted flavor evaporation (SAFE), and GC–MS (Feng et al., 2019). They aimed to explore the impact of each biological metabolic pathway corresponding to the six main tea processing models by comparing the proportion of substance content and *ACI* value (the proportion of OAV value of each substance). Their results

* Corresponding authors at: National Key Lab for Germplasm Innovation and Utilization of Horticultural Crops, College of Horticulture & Forestry Sciences, Huazhong Agricultural University, Wuhan, Hubei 430070, People's Republic of China.
E-mail addresses: catea37@mail.hzau.edu.cn (Y. Wang), nidj@mail.hzau.edu.cn (D. Ni).

https://doi.org/10.1016/j.fochx.2023.100730
Received 20 February 2023; Received in revised form 21 May 2023; Accepted 25 May 2023
Available online 29 May 2023

indicated that the oxidation of fatty acids may be the key pathway for the development of oolong tea's aroma.

Production experience demonstrates that oolong tea cultivars influence how the aroma develops (Guo, Schwab, Ho, Song, & Wan, 2022). When compared to other teas, oolong tea has a more robust floral and fruity aroma, which is the source of the "cultivar aroma" found in high-quality oolong tea and is associated with cultivar suitability (He et al., 2022). A distinctive economic characteristic of tea cultivars is their adaptability for processing (Zeng et al., 2020). In another study, researchers investigated the aroma changes of two tea varieties ("Zhenong 117", a unsuitable cultivar, and "Foshou", a suitable cultivar) during turning-over processing, and the ratio of terpenoid volatiles (VTs) to green leaf volatiles (GLVs) was shown to be one of the factors influencing cultivar suitability (Hu et al., 2018). However, only one suitable cultivar of oolong tea is limited and cannot fully demonstrate the effect of different suitable oolong tea cultivars on its aroma quality.

This study aims to analyze the unique aroma components of oolong tea by examining the connection between its processing method and aroma qualities that distinguish it from other teas, such as black tea and green tea. Additionally, the study will conduct an in-depth analysis of the relationship between oolong tea aroma characteristics and different tea cultivars, exploring the similarities and differences of aroma traits among various suitable cultivars. The research will utilize aroma profiling analysis, headspace solid-phase microextraction coupled to gas chromatography-mass spectrometry (GC–MS HS-SPME-GC–MS), and aroma extract dilution analysis combined with gas chromatography–olfactometry-mass spectrometry (AEDA-GC-O-MS) to investigate the aroma of oolong tea. Finally, the study will confirm the impact of the discovered aroma-active compounds on oolong tea aroma characteristics through aroma recombination and omission experiments. The findings of this study may provide valuable insights for further research on the suitability of tea cultivars. The results could offer fresh suggestions and techniques for enhancing tea tree cultivars, providing valuable insights for further research in this area.

Materials and methods

Tea manufacturing process

The experimental materials included the classic suitable cultivars (for oolong tea manufacturing) *Camellia sinensis (L.) O. Kuntze cv. Huangdan* (HD), *Camellia sinensis (L.) O. Kuntze cv. Tie guanyin* (TGY), and *Camellia sinensis (L.) O. Kuntze cv. Meizhan* (MZ), as well as the unsuitable cultivars *Camellia sinensis (L.) O. Kuntze cv. Fuding Dabaicha* (FD), *Camellia sinensis (L.) O. Kuntze cv. Yingshuang* (YS), and *Camellia sinensis (L.) O. Kuntze cv. Jiaming 1* (WNZ). One bud and two or three leaves were plucked in August for tea manufacturing using the "Qingxiang" Oolong tea processing method (Gui et al., 2015). All cultivars are harvested from the tea garden of Huazhong Agricultural University, located at an altitude of 49.8 m, a longitude of 114°22 min, and a latitude of 30°29 min. Briefly, the fresh leaves were first collected and then spread out over a water sieve for 2 h, followed by withering the leaves for 30 min in the sun (leaf temperature: 32 °C; light intensity: 24000Lx), then placing the leaves in a room to wither for another 2 h or until their moisture content reached 72%. Next, the withered leaves were turned over for 2 min (20 rpm), followed by further withering for 1.5 h, then a second turn-over for 4 min and withering for 3 h, and finally a third turn-over for 6 min and withering for 6 h or until the water content of the tea leaves reached 60% (Room temperature 26 ± 1°Celsius, air humidity 60 ± 5%). Subsequently, the samples were fixed at 220 °C using a 6CST-50 drum fixing machine (Zhejiang, China), immediately rolled for 10 min with a 6CR-25 rolling machine, dried at 110 °C (6CTH-60 drying machine) for 8 min, piled for 1 h, and finally dried at 80 °C until they were sufficiently dried. Meanwhile, we set up a control group where oolong tea was processed by maintaining the withered leaves indoors until the end of the third turn-over processing of the experimental group, and the remaining procedures were the same for the experimental group. The black tea was manufactured by using the broken black tea processing method (Mahanta & Baruah, 1992). Briefly, the fresh leaves were first collected and then spread out for 8 h over a water sieve until the water content of the tea leaves reached 60%. After breaking and cutting (6CRQ-16 CTC machine, Zhejiang, China), the tea leaves were sieved through a 40-mesh screen, followed by fermentation in a fermenter (33 °C, 95% humidity) for 2 h, drying at 110 °C (6CTH-60 type drying machine) for 10 min, piling for 1 h, and finally drying at 80 °C until they were sufficiently dried. The processing of green tea followed a previously reported method (Feng et al., 2019). Briefly, the fresh leaves were first gathered and then spread out for 2 h over a water sieve until their water content reached 72%, followed by fixing at 270 °C (6CST-50 fixing machine), chilling the tea leaves at room temperature for 1 h and rolling for 20 min (6CR-25 rolling machine). Subsequently, the tea leaves were dried at 110 °C (6CTH-60 drying machine) for 10 min, piled for 1 h, and finally dried at 80 °C until they were sufficiently dried. The samples were stored at −20 °C for further analysis. Fig. A.1 shows the flow chart of tea processing.

Chemicals

Hexanal (95%), (E)-3-hexen-1-ol, (97%), heptanal (95%), benzaldehyde (99%), 6-methyl-5-heptene-2-one (98%), octanal (98%), 2-ethyl-1-hexanol (99.5%), ocimene (mixture of isomers, 90%), (E)-2-octenal, (97%), nonanal (99.5%), (E,Z)-2,6-nonadienal, (94.5%), (E)-2-nonenal, (95%), methyl salicylate (99%), α-terpineol (95%), β-cyclocitral (80%), (Z)-geraniol (98%), phenethyl acetate (97%), citral (95%), indole (99%), hexanoic acid, hexyl ester (98%), geranylacetone (97%), β-ionone (97%), (Z,E)-α-farnesene (mixture of isomers, 90%), linalool (97%), (E)-nerolidol (85%), ethyl decanoate (internal standard, 99%), and *n*-alkanes (C3–C25) were purchased from Sigma-Aldrich (Milwaukee, USA). (E, E)-2,4-Hexadienal (95%), (Z)-2-heptenal, (95%), (E,E)-2,4-heptadienal, (90%), benzyl alcohol (98%), phenylethyl Alcohol (99.5%), butanoic acid, hexyl ester (98%), (E,E)-2,4-nonadienal, (85%), *cis*-3-hexenyl isovalerate (97%), (E)-2-decenal, (95%), (E,E)-2,4-decadienal, (90%), E-2-undecenal, (93%), *cis*-3-hexenyl *cis*-3-hexenoate (95%), *cis*-jasmone (98%), δ-decalactone (98%), 3-hexen-1-ol, benzoate, (Z)- (97%), benzoic acid, hexyl ester (98%) were purchased from Aladdin Biochemical Co., Ltd. (Shanghai, China). (E)-Hexanoic acid, 2-hexenyl ester, (97%) was purchased from Shanghai Macklin Biochemical Technology Co., Ltd. (Shanghai, China). *cis*-3-Hexenyl-α-methylbutyrate (97%) was obtained from TCI Development Co., Ltd. (Shanghai, China). Jasmine lactone (95%) was purchased from Bidepharm Co., Ltd. (Shanghai, China). Benzeneacetaldehyde (98%), linalool oxide pyranoside (mixture of isomers, 98%), geraniol (98%), α-ionone (90%), β-phenylethyl butyrate (98%), (E)-isoeugenol (98%), (E)-β-famesene (mixture of isomers, 80%), dihydroactindiolide (98%), was obtained from Shanghai yuanye Bio-Technology Co., Ltd (Shanghai, China). A SPME fiber manual sampling holder and polydimethylsiloxane/divinylbenzene (PDMS/DVB, 65 μm, 1 cm) microextraction fiber were purchased from Supelco (Bellefonte, PA, USA).

Aroma quality assessment by experts

The samples of green tea, black tea, and oolong tea from six different tea cultivars were evaluated and graded by skilled tea experts using a previous method (Feng et al., 2019). The aroma evaluation method involves taking 3 g (for black and green tea) or 5 g (for oolong tea), placing it in a corresponding cylindrical cup, adding boiling water at a tea-water ratio of 1:50, covering it, and timing it for 5 min. Afterward, the tea soup is quickly filtered out, and the aroma is evaluated by sniffing the cylindrical cup. The aroma evaluation and scoring standards are based on the guidelines provided in the national standard, which are listed in the Table A.11.

HS- SPME- GC–MS method

Based on the previously modified headspace-solid phase microextraction (HS-SPME) method(He et al., 2022), volatile compounds were directly extracted from each sample and identified by TRACE-DSQ-II GC–MS (Thermo Fisher Scientific, Waltham, MA, USA). Briefly, in a 20 mL headspace vial, each sample (1 g) crushed by liquid nitrogen was first brewed with 5 mL boiling water, followed by adding 200 μL (0.04 μL/100 mL) of ethyl decanoate as an internal standard (IS) and immediately sealing the vial. After fixing the polydimethylsiloxane/divinylbenzene (PDMS/DVB) fiber (Supelco, Bellefonte, PA, USA) on its upper end, the headspace vial was kept in a 60 ℃ water bath for 60 min to absorb the volatile gases, followed by taking the fiber out of the vial and instantly inserting it into the GC's entryway for aroma analysis. The fiber was desorbed for 5 min at 250 °C in between injections. The mass spectrometric identification was performed on the chromatographic detection system equipped with a DB-5MS column (30 m × 0.25 mm i.d. × 0.25 μm film thickness, Thermo Fisher) under the following conditions: GC oven temperature, 45 °C to 80 °C at 7 °C/min, up to 90 °C at 2 °C/min and hold for 2 min, up to 100 °C at 3 °C/min and hold for 2 min, up to 130 °C at 3 °C/min and hold for 2 min, up to 150 °C at 3 °C/min, and up to 230 °C at 10 °C/min and hold for 15 min; column oven temperature, 40 °C; GC, injection splitless; MS: full scan, *m/z* 45–400; EI mode, positive ion mode, 70 eV; source temperature, 280 °C; ion source temperature, 230 °C.

Identification and quantitation of volatiles

The actual retention index (RI) values of the tested samples were determined by adding *N*-alkane (c3-c25) standard to the headspace vial and following the same approach as described above. The corresponding reference information was analyzed based on the mass spectrometry fragmentation reference information and the RI value reference information in the NIST2017 database. The tested material was quantified simultaneously using the previously reported method (Cui et al., 2022) by quantitatively mixing the internal standard method with the adjustment factor.

The response factor (Rf) of each standard compound was calculated by equation (1):

$$f_i = \frac{f_{wi}}{f_{ws}} = \frac{m_i/A_i}{m_s/A_s} = \frac{A_S m_i}{A_i m_s} \quad (1)$$

where f_i is the response factor of standard compound i; m_s and mi, the injection weight of standard i and internal standard s, respectively; $A_{s,}$ the peak area of the internal standard s; $A_{i,}$ the peak area of compound i.

The concentration of volatiles was calculated by equation (2):

$$m_i = f_i \times A_S \times \frac{m_i}{A_i \times m_o} \times 1000 \quad (2)$$

where m_i is the concentration of identified volatiles, μg/kg; $m_{0,}$ the weight of the tea infusion, g; A_i, the peak area of identified volatiles; A_s and $m_{i,}$ the weight (μg) and peak area of the internal standard (ethyl caprate), respectively.

When comparing the relative content of volatile compounds of the three types of tea, the concentration was calculated with an Rf of 1. When comparing the actual content of aroma-active compounds of oolong tea, the concentration and Rf were calculated by equations (1) and (2).

SAFE analysis for oolong tea

Briefly, oolong tea (10 g) was brewed with 500 mL of boiling water for 10 min under continuous stirring, followed by filtering, cooling the infusion immediately to room temperature in running water, and then exposing the infusion to a SAFE system composed of a glass apparatus (Beijing Synthware Glass Co., Ltd., Beijing, China) and a vacuum pump as previously described (Engel, Bahr, & Schieberle, 1999). Volatiles were extracted from the tea under following conditions: water bath at 40 °C, condenser temperature at 40 °C, and high vacuum at 1 to 5 × 10–3 Pa. The SAFE distillate was thawed in running water and then extracted four times using 80 mL of distilled dichloromethane, followed by combining the organic phases, drying with anhydrous sodium sulfate, and concentration in a Vigreux column (60 cm) at 48 °C to about 10 mL and then to 0.5 mL with a gentle nitrogen stream. Finally, the concentrate was kept at − 80 °C for further analysis.

AEDA-GC-O-Ms

The potent odorants in the concentrated liquid were evaluated by AEDA-GC-O-MS as previously reported (Gui et al., 2015). Briefly, GC-O-MS consisted of an ODP-3 olfactory detection port (Gerstel, Germany), a TRACE 1300 chromatographic detection system (Thermo Fisher Scientific), and a ISQ 7000 mass spectrometry system (Thermo Fisher Scientific). The concentrate of oolong tea sample by SAFE was step diluted with dichloromethane 1:4 = 4^1, 1:16 = 4^2, 1:64 = 4^3, 1:256 = 4^4, 1:1024 = 4^5, and 1:4096 = 4^6. The flavor dilution (FD) factors of the odorants were determined by AEDA. Mass spectrometric identification was performed using the chromatographic detection system equipped with a TG-WAXMS A column (60 m × 0.25 mm i.d., 0.25 μm film thickness, Thermo Fisher Scientific). The aliquots (1 μL) of each sample were injected to the GC-O-MS in the splitless mode (S/SL: front and back) with a splitless time of 1 min, a split ratio of 33.3 at the time of split vent liberation. The other conditions were as follows: carrier gas, helium at 1 mL/min; injector temperature, 230 °C; GC oven temperature, 40 °C for 2 min, up to 230 °C at 5 °C/min and hold for 25 min; MS: full scan, *m/z* 45–400; EI mode, positive ion mode, 70 eV; mass spectrometer interface temperature, 260 °C; ion source temperature, 280 °C; temperature of olfactory detection port transmission line, 250 °C; temperature of olfactory detection port mixing chamber, 250 °C. Three skilled reviewers sniffed samples of oolong tea at different concentrations through the ODP-3 olfactory detection port and gave recognizable olfactory areas. Volatile compounds correlated to the olfactory areas were identified by direct comparison with the GC RI values, mass spectra, and those of the authentic specimens.

Aroma profile analysis of oolong tea

The samples of oolong tea from six cultivars were evaluated by a panel of 10 skilled and trained assessors in a quantitative descriptive manner. Based on their discussion and sniffing of the samples, assessors recognized the aroma attributes of the oolong tea as fruity, floral, sweet, and fresh. Coffee Aroma Kit 100 (SCENTONE, USA) provided the aroma indicator for the smell training, with rose, jasmine, and chrysanthemum aroma simulants as floral indication; typical fruitmix aroma simulants as fruity indication; caramel aroma simulants as sweet indication; cucumber aroma simulants as fresh indication. The aroma indicators with progressively higher dilution concentrations were set up based on group discussion and sniffing with the odor concentration corresponding to 1 to 9 points: 8 to 9, strong; 5 to 7, moderate; 3 to 5, clear; 1 to 2, ambiguous.

The evaluation was carried out using a previous method (He et al., 2022). Briefly, in a gaiwan (110 mL, including a bowl and a lid), each sample (5 g) was brewed with boiling water for 1 min, followed by removing the lid, and assessing the aroma of each sample for the first time. Two min later, the tea soup was poured out, and the samples left in the gaiwan were brewed again with boiling water for 2 min, followed by removing the lid, and assessing the aroma of each sample for the second time. One min later, the tea soup was poured out, and the samples left in the gaiwan were steeped in boiling water for three min, followed by removing the lid again, and assessing the aroma for the third time. The aroma results were based on the three evaluation results, with more

weight for the second evaluation result when using the nine-point scale for evaluation (see Table 1).

Odor activity value (OAV)

OAV is the ratio of the actual concentration of a compound in a tea infusion to its odor threshold (OT) in water. Compounds contributing to aroma characteristics have an OAV greater than 1 (Cui et al., 2022). In this study, the OAVs of aroma components in the samples of oolong tea were calculated and shown in Table A.4.

Aroma recombination and omission experiments

Aroma-active compounds identified with GC–O (FD $\geq$ 4) and OAV (OAV $\geq$ 1) methods were used for aroma recombination and omission experiments (Table 2). The blend essential oils were mixtures of the aroma-active compounds with quantitative amounts in each sample. Based on previously described procedures (Du, Wang, Li, Xiao, Li, & Xu, 2013), the "volatile-free" tea matrix was produced by repeatedly leaching the tea leaves of the relevant samples with boiling water and rotary evaporation to eliminate scent. The aroma recombination system of each oolong tea sample contained 1 g of unflavored tea matrix and blend essential oils, and in a 20 mL headspace bottle, 5 mL of boiling water was poured into the aroma recombination system, followed by leaching the bottle in a 60 °C water bath for 10 min. Meanwhile, the equivalent control samples were also obtained using the same technique for the real samples of the same quality. Reviewers assessed the recombinant samples and matched control samples as mentioned above in *Aroma profile analysis of oolong tea*. To demonstrate the impact of each compound on the aroma attributes more clearly, one compound was removed from the aroma-active compounds of the HD recombinant sample at a time to create the omission samples, with the initial sample as a control. The reviewers evaluated how closely the experimental (omission) samples matched the control samples using the previous nine-point scale (Liu, Cheng, Zhang, Deng, Chen, & Xu, 2012). The assigned scores ranged from 1 (extremely similar) to 9 (extremely different). Lastly, they also discussed the fragrance alterations in experimental samples.

Validation experiments on effects of volatile compounds on aroma properties

The aroma recombination systems of HD and WNZ were used to confirm the effects of volatile compounds on the "fruity, floral", and "fresh" odor in two groups (contributors to the "fruity, floral" (Group (1) and "fresh" (Group (2) aroma attributes) as described by the aroma of a single volatile compound at the concentration of each sample in the results of aroma recombination and omission experiments (Table 2). Specifically, in order to obtain samples HD-F and HD-Q, the Group (1) and Group (2) compounds in the HD recombination system were completely removed, using the original HD recombination system without treatment as a control sample (HD-R). Next, the sample WNZ-R + HD-F was prepared by adding Group (1) compounds from the HD recombination system to the WNZ recombination system, using the original WNZ recombination system without treatment as a control sample (WNZ-R). Finally, aroma profile analysis was performed for samples HD-R, HD-Q, HD-F, WNZ-R, and WNZ-R + HD-Q with identified aroma attribute scores as described in *Aroma Profile Analysis of oolong tea*.

Statistical analysis

The statistical analyses were performed with one-way analysis of variance (ANOVA) followed by Student *t*-test using SAS JMP Statistical Discovery Pro 16 software (SAS, Cary, NC, USA). R 4.1.3 were used for cluster analysis, (Paired) Wilcoxon Signed-Rank Test, Kruskal-Wallis Test, Spearman's correlation analysis, PCA analysis, PCoA analysis, Adonis test and plotting Figs. 1–4. A p-value < 0.05 was defined as statistically significant difference.

Results and discussion

Characteristics of aroma compounds under different processing modes

In Fig. 1 and Table A.1, the expert sensory review results of the three teas in this study were consistent with the typical quality traits of oolong tea, black tea, and green tea, with a chestnut aroma for green tea, a high fresh aroma for black tea, and a stronger flowery and fruity aroma for oolong tea than green tea or black tea. In Fig. 1C, PCoA analysis of the GC–MS results revealed significant differences in the distribution of aroma components of oolong tea, black tea, and green tea (Bray Curtis, Adonis: R2 = 0.8453, P = 0.001). Total aroma content showed no obvious difference between oolong and black tea, but was significantly lower in green tea than in oolong or black tea. Specifically, terpenoid volatiles (VTs) are the dominant category in the aroma, with the highest percentage for green tea (51.98 ± 1.43%), followed by black tea (40.4

Table 1
Identification of potent odorants (FD Factor $\geq 4^4$) in oolong tea.

Name	FD factor (4^n)						description	P of Kruskal. Test
	TGY	HD	MZ	WNZ	FD	YS		
6-methyl-5-Hepten-2-one	4	4	4	4	5	4	light floral	0.317
(E)-3-Hexen-1-ol	5	5	5	4	2	5	light floral	0.121
(Z)-Furan linalool oxide	5	5	6	5	5	4	fruity	0.197
(E, E)-2,4-Heptadienal	5	4	5	5	1	0	light floral	0.246
Benzaldehyde	6	5	5	0	5	4	fruity	0.105
Linalool	6	5	5	5	5	4	fruity and floral	0.197
Benzeneacetaldehyde	5	5	4	5	5	5	sweet	1.000
Linalool oxide pyranoid	5	5	5	6	5	5	sweet	0.317
Methyl salicylate	5	5	4	0	5	5	floral	0.796
Geraniol	4	6	5	3	4	5	light fruity	0.261
Benzyl alcohol	5	4	6	5	5	2	fresh	0.487
Phenylethyl Alcohol	5	4	4	4	4	4	sweet	0.317
(E)-Nerolidol	5	5	5	2	5	2	fruity and floral	0.114
(Z)-3-Hexenyl benzoate	3	4	5	1	3	4	green	0.261
Jasmine lactone	4	5	5	3	3	2	fruity	0.046*
(E)-Isoeugenol	6	5	5	4	5	4	floral	0.099
Indole	5	6	5	4	3	3	strong floral	0.043*

Note: The statistical analyses were performed with Kruskal-Wallis Test between the FD factors of suitable cultivars (HD, TGY, MZ) and unsuitable cultivars (FD, YS, WNZ). "*": P values < 0.05.

Table 2
Aroma-active compounds (OAV greater than 1 and FD Factor $\geq 4^4$) utilized in aroma recombination and omission experiments of oolong tea samples.

Name	Type	RF	OT (ug/L)	Content of HD (μg/kg)	Content of WNZ (μg/kg)	P value (Wilcox. Test)	Differences (Wilcox. Test)	Description after Elimination
(E)-3-Hexen-1-ol		0.625	110[a]	11.208	3.162	0.181		fresh, more floral, citrus, sweet
Heptanal	Group (2)	3.657	2.8[b]	16.736	11.893	0.018	*	fresh, more floral, melon
Benzaldehyde	Group (1)	3.344	750.89[b]	54.765	77.738	0.021	*	fresh, floral and almond
6-methyl-5-Hepten-2-one	Group (1)	0.247	68[b]	17.757	15.902	0.021	*	fresher
(E, E)-2,4-Heptadienal	Group (2)	0.656	56[e]	21.887	14.507	0.020	*	fresh, more citrus, cucumber
Benzeneacetaldehyde	Group (1)	3.330	0.0063	107.986	94.716	0.020	*	fresh, and more melon
(E)-2-Octenal	Group (2)	0.397	4[f]	8.953	4.274	0.020	*	more fresh
(E, Z)-2,6-Nonadienal		0.818	0.03[d]	2.618	2.608	0.060		more fresh and sweet
(E)-2-Nonenal	Group (2)	0.374	0.4[d]	6.149	2.677	0.020	*	more fresh and sweet
Methyl salicylate	Group (1)	0.977	40[f]	19.956	33.262	0.020	*	fresh and melon
(E, E)-2,4-Nonadienal	Group (2)	0.531	0.16[d]	5.655	3.474	0.020	*	floral, more floral
β-Cyclocitral	Group (1)	0.427	5[b]	33.507	28.642	0.020	*	fresh, melon and chrysanthemum
Geraniol		0.797	3.2[d]	16.951	13.020	0.069		melon and floral
(E)-2-Decenal		0.572	0.3[i]	45.204	15.408	0.181		floral and fresh
Indole	Group (1)	3.044	11[f]	3530.936	1255.118	0.020	*	more fruity and sweet
(E, E)-2,4-Decadienal	Group (2)	0.722	0.2[i]	15.006	6.0438	0.021	*	fresh, floral and fruity
(E)-isoeugenol	Group (1)	2.025	6 [l]	304.421	485.592	0.020	*	floral and fruity,more citrus
β-Ionone	Group (1)	0.574	0.007[b]	69.069	68.502	0.020	*	fresh,more citrus, sweet
Jasmine lactone	Group (1)	1.650	120[f]	340.781	5.448	0.020	*	more fresh, citrus and melon
(E)-Nerolidol	Group (1)	0.227	250[j]	54.675	63.802	0.021	*	fresh, floral
Linalool	Group (1)	0.426	0.6[f]	87.863	43.447	0.021	*	floral and melon

Note: RF, the response factors were obtained in *Identification and quantitation of volatiles* method. Content of HD and WNZ, average of quantitation of volatiles in oolong tea were obtained by *HS–SPME–GC–MS method* and *Identification and quantitation of volatiles*. P value, the paired Wilcoxon Test was used to evaluate the differences between original system and omission system. Differences, "*": p value < 0.05. OT, odor threshold (in water), OT annotation from the references, [a] (Bonneau, Boulanger, Lebrun, Maraval, & Gunata, 2016), [b] (Feng et al., 2022), [c] (Teranishi, Buttery, & Guadagni, 1974), [d] (Schuh & Schieberle, 2006), [e] (Zhu et al., 2015), [f] (J. Wang et al., 2022), [g] (Xie et al., 2023), [h] (Guo, Ho, Wan, Zhu, Liu, & Wen, 2021), [i](Liu et al., 2023), [j] (Yao et al., 2023), [k] (Dubrow, Forero, & Peterson, 2022), [l] (Del Barrio Galán, Bueno-Herrera, De La Cuesta, & Pérez-Magariño, 2022).

± 0.75%) and oolong tea (34.3 ± 1.84%) (Fig. 1B and Table A.2). Meanwhile, oolong tea has the largest percentage of fatty-acid-derived volatiles (FADVs) in its aroma (24.21 ± 1.72%), followed by black tea (13.33 ± 0.98%), and green tea (2.13 ± 0.12%). Compared with green tea (10.89 ± 0.64%), oolong tea (27.21 ± 1.8%) and black tea (30.14 ± 0.79%) have a higher percentage of amino-acid-derived volatiles (AADVs) in their aroma. These data indicated that oolong tea differs from the other two teas in the aroma qualities due to compositional variances between various components.

In Fig. 1D and Supplemental files 2, further analysis of variations in aroma compounds across different categories revealed 18 distinct VTs compounds in various teas, with 10 compounds significantly greater in oolong tea than in green tea ((E)-β-ocimene, α-farnesene, and so on) and 6 compounds considerably higher in oolong tea than in black tea (β-ocimene, (E)-β-ocimene, (E)-4,8-dimethylnona-1,3,7-triene, (Z,E)-α-farnesene, α-farnesene, and (E)-nerolidol). Oolong tea had been reported to produce the following compounds during the turn-over stage or adversity coercion: (E)-ocimene, α-farnesene, and (E)-nerolidol (Chen et al., 2021; Gui et al., 2015; Zeng et al., 2017). Furthermore, (E)-β-ocimene, (E)-farnesene, and (E)-4,8-dimethylnona-1,3,7-triene were considered to contribute to the plant's defensive system against adversity, and the components of oolong tea's aroma compounds include (E)-nerolidol and α-farnesene (Farré-Armengol, Filella, Llusià, & Peñuelas, 2017; Guo et al., 2022; Hegde et al., 2012; Jing et al., 2021). On the other hand, oolong tea was much lower than black tea in the content of linalool and its oxides, geraniol, and other fragrance compounds with floral and sweet aroma. According to studies, linalool and its glycosides were present in fresh leaves in a binding form and released in significant amounts as a result of glycoside hydrolysis during the rolling and fermenting processes of black tea, endowing the beverage with a distinctive scent. However, the position of the hydrolase and these bound glycosides was shown to differ in cells. Oolong tea undergoes little cell wall damage during the turn-over stage, and the generation of these aromatic compounds is not primarily based on hydrolysis but on de novo synthesis (Cui et al., 2016; Gui et al., 2015).

In Fig. 1D and Supplemental files 2, there were 11 unique chemicals for AADVs from various teas, and oolong tea was significantly higher than green tea in ten AADVs and black tea in five AADVs (indole, β-Phenylethyl butyrate, (Z)-3-Hexen-1-ol benzoate, benzoic acid, and hexyl ester). Indole was reported to be one of the characteristic components of oolong tea (Gui et al., 2015; Guo et al., 2022). The rolling stage of black tea processing had been claimed to stop the production of indole due to destruction of cells, inferring that the indole in black tea is mostly derived from a slight amount of accumulation during the

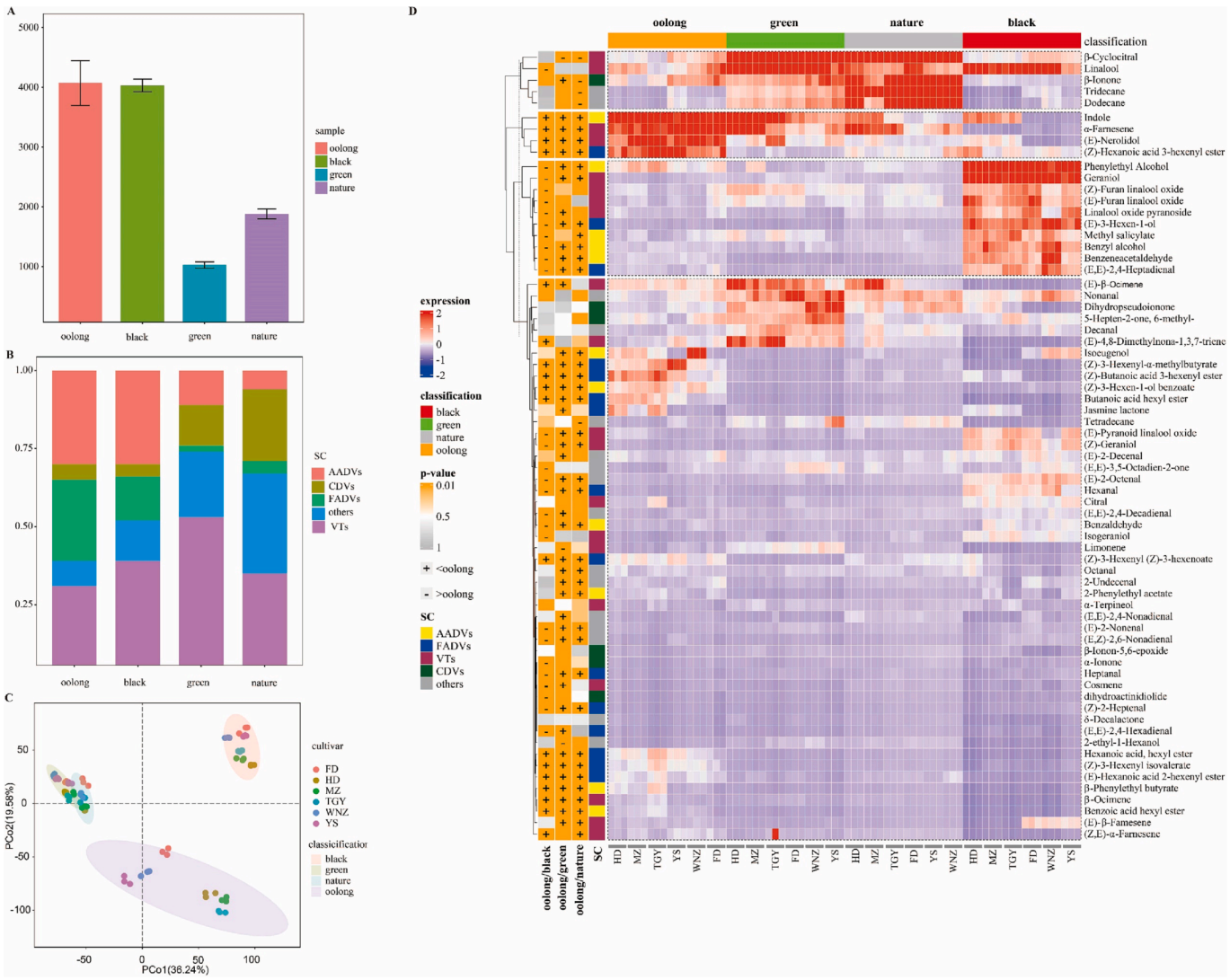


Fig. 1. Relative quantitation and statistical analysis of volatiles in different processing modes. (A) GC–MS-based bar graph of total aroma of different tea samples. (B) Classification stacking diagram of aroma compounds in different tea samples based on GC–MS. (C) Principal Coordinate Analysis (PCoA) score plot of aroma compound content of different tea samples based on GC–MS. (D) Heat map of aroma compound content of different tea samples based on GC–MS. SC: Classification of aroma compounds according to metabolic pathways. Expression: z-score normalized and centralized numerical values of relative content of aroma compounds. Classification: different types of tea samples are grouped, and "nature" was an oolong tea sample without turn-over stage. P-value: The p value of the paired Wilcoxon signed-rank test based on the Type grouping, "+": The content of aroma compounds in the paired Wilcoxon signed-rank test is significantly ($p < 0.05$) less than that of the oolong tea sample; "-": In the paired Wilcoxon signed-rank test, the content of aroma compounds in the group was significantly ($p < 0.05$) greater than that of the oolong tea samples.

withering stage, while the indole in oolong tea mainly comes from a larger amount of accumulation during the turn-over stage. (Zeng et al., 2016). Benzaldehyde, benzyl alcohol, benzeneacetaldehyde, and phenylethyl alcohol were compounds with floral and fruity aromas. Meanwhile, benzyl alcohol and phenylethyl alcohol are also reported to be glycoside bound volatiles present in fresh leaves. The generation of a significant amount of glycosidically bound volatiles (GBVs) due to the reaction between β-primeverosidase and the substrate (Cui et al., 2016; Liang et al., 2022). Additionally, phenylalanine breakdown has been suggested to be responsible for the large changes of benzaldehyde and benzeneacetaldehyde in black tea during fermentation (Liu et al., 2023). However, these GBVs did not change much in the turn-over process of oolong tea, indicating no occurrence of hydrolysis of GBVs during oolong tea processing (Cui et al., 2016), and benzyl alcohol and phenylethyl alcohol were generated by de novo synthesis. The scent of methyl salicylate is like wintergreen oil (Ho, Zheng, & Li, 2015), and it was reported to be generated in significant amounts throughout the fermentation process as a key indicator of fermentation degree (L.-F. Wang, Lee, Chung, Baik, So, & Park, 2008). As a result, this compound had a significantly higher content in black tea than in oolong tea or green tea.

FADVs from the various types of teas were seen to contain 15 distinct chemicals (Fig. 1D and Supplemental files 2). Oolong tea was significantly higher than green tea in the 15 different FADVs compounds and higher than black tea in 8 different FADVs containing the C6 moiety of leaf alcohol esters(such as hexanoic acid hexyl ester). The compound (Z)-butanoic acid 3-hexenyl ester, which has a floral and fruity scent, had been reported to be released when plants are exposed to biotic stress (Mu et al., 2012). (Z)-hexanoic acid 3-hexenyl ester has a pronounced fruity scent and was transformed by *cis*-3-hexenyl alcohol (Yang, Baldermann, & Watanabe, 2013). In addition, jasmine lactone, which was generated from scratch in the oolong tea turn-over process as one of the identified characteristic scent compounds of oolong tea, has a fruity aroma (Gui et al., 2015).

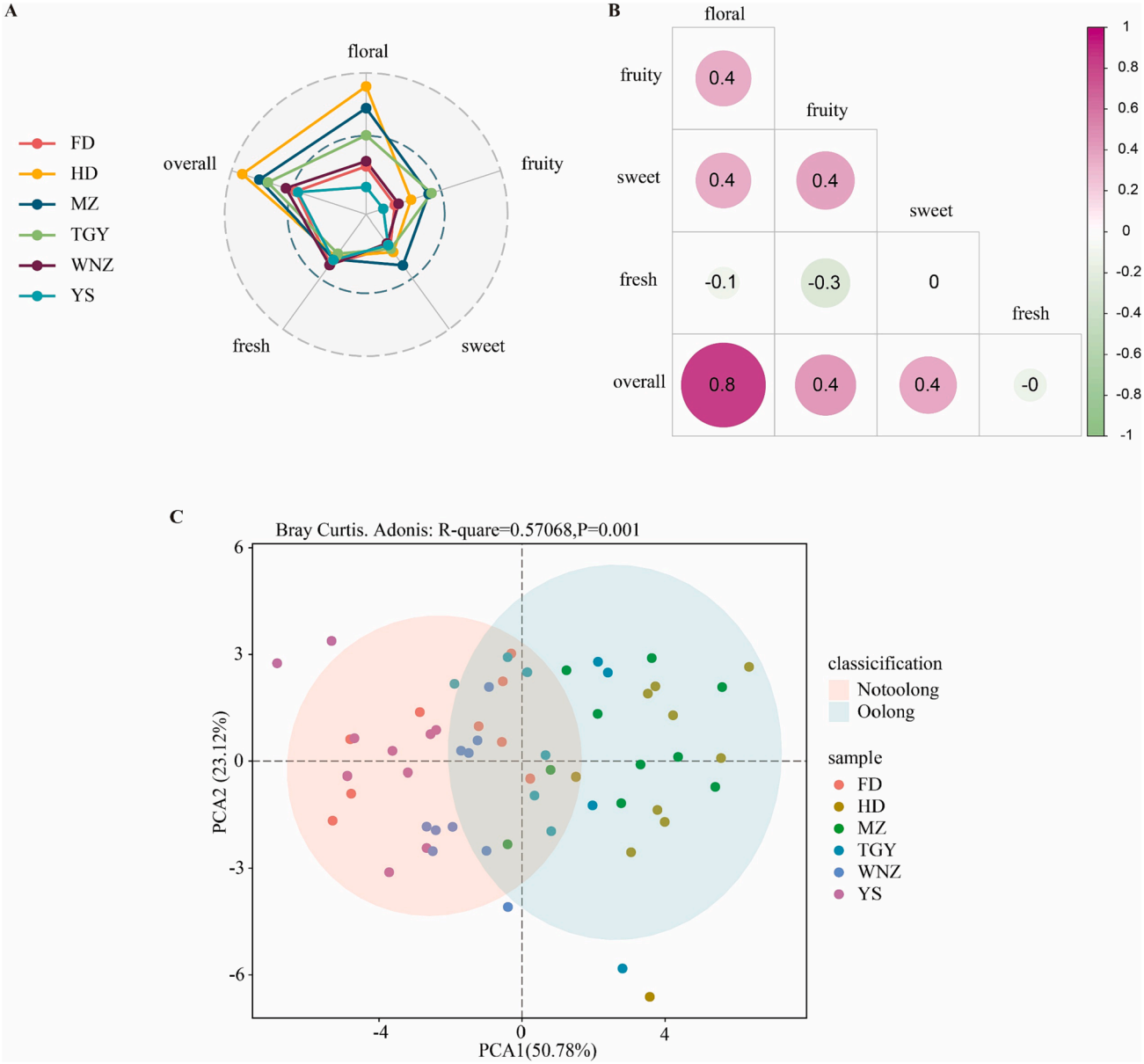


Fig. 2. Aroma profile analysis and statistical analysis of oolong tea soups from different cultivars. (A) Radar chart for quantitative description of tea soup aroma attributes for six oolong tea samples. The gray dotted line in the outer circle represents 9 points; the indigo blue dotted line in the inner circle, 5 points; the center of the radar chart, 1 point. (B) Heatmap of correlations between tea soup aroma attributes and overall evaluation for six oolong tea samples. The size of the circle and the shade of color both indicate the magnitude of the correlation. (C) The PCA analysis score plot of the aroma attribute evaluation scores of the tea soups of the six oolong tea samples. Dots: the distribution of scores of the samples in the Aroma Profile Analysis experiment on the PCA score map. Classification: suitability grouping of oolong tea samples; Notoolong: samples of unsuitable oolong tea cultivars; Oolong: samples of suitable oolong tea cultivars. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Three distinct compounds (dihydroactinidiolide, α-ionone, and β-ionone) were present in various tea carotenoid-derived volatiles (CDVs) (Fig. 1D). Oolong tea had a much greater β-ionone content than green tea, but a significantly lower β-ionone and dihydroactinidiolide content than black tea. Carotenoid cleavage enzymes (CCDs) were reported to produce β-ionone during tea enzymatic reaction, especially during fixation of oolong tea and green tea as well as fermentation of black tea. Different carotenoid precursor molecules may be the root of variations in carotenoid derivatives, such as β-ionone, which are subject to enzyme reactions (Yang et al., 2013). In the category of others, ten distinct compounds were discovered, but they were all low in content (Fig. 1D). Oolong tea contains octanal, (E)-2-octenal, (E, Z)-2,6-nonadienal, (E)-2, (E, E)-2,4-nonenal, (E)-2-decenal, (E,E)-2,4-decadienal, and 2-undecenal, whose levels were all significantly greater in oolong tea than in green tea.

Turn-over stage was an essential process for oolong tea to form its aroma characteristics, and turning-over and withering are used alternately throughout oolong tea processing. In this study, the processed product with turn-over treatment served as the experimental group, while the product without turn-over treatment served as the control group. The total aroma amount was significantly higher in the experimental group than in the control group (Supplementary B.1). In Fig. 1, PCoA analysis of the GC–MS data showed a significant difference between the experimental and control groups. Specifically, the experimental group was significantly higher than the control group in the contents of FADVs, AADVs, and VTs, indicating the occurrence of several chemical processes during the turn-over process of oolong tea, such as lipid oxidation, amino acid metabolism, terpene synthesis, and

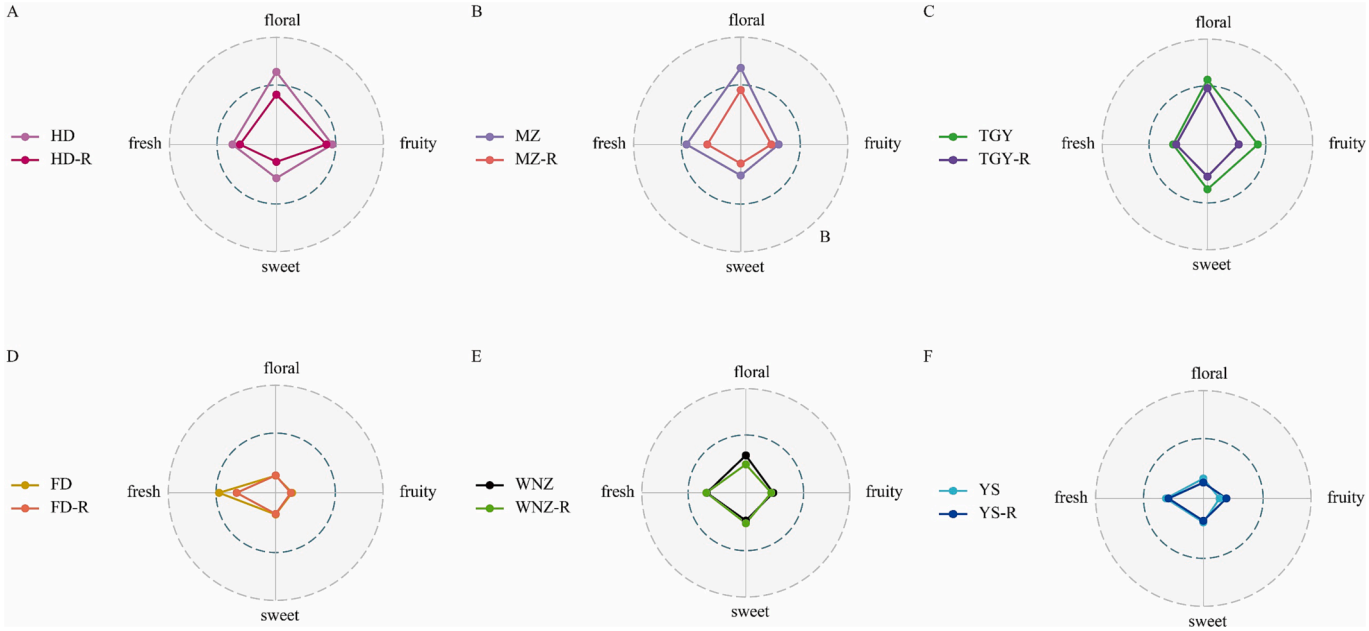


Fig. 3. Radar plots of scores on aroma attributes between the reconstituted system and actual samples of six oolong tea samples. Note: The gray dotted line in the outer circle represents 9 points; the indigo blue dotted line in the inner circle, 5 points; the center of the radar chart, 1 point. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

conversion, enabling the formation of oolong tea aroma characteristics.

Aroma profile analysis of oolong tea soups

Tea cultivars have long been considered as the primary element influencing the formation of oolong tea aroma (Zeng et al., 2020). The aroma profile analysis results of oolong tea soups are shown in Fig. 2 and Table A.5. In floral odor scores (Table A.5), the six different oolong tea cultivars were in the sequence of HD (8.15 ± 0.26) > MZ (6.76 ± 0.45) > TGY (5.03 ± 0.63) > WNZ (3.38 ± 0.17) > FD (3.05 ± 0.47) > YS (1.74 ± 0.21). In fruity odor scores, TGY (4.37 ± 0.67) and MZ (4.18 ± 0.47) had a higher score, followed by HD (3.01 ± 0.53), WNZ (2.17 ± 0.43), FD (1.92 ± 0.36), and YS (1.15 ± 0.21). In sweet odor scores, MZ was significantly higher than the others. In fresh odor scores, the six cultivars showed no significant difference from one another. The overall score was in the order of HD (8.29 ± 0.18) > MZ (7.15 ± 0.38) > TGY (6.58 ± 0.11) > WNZ (5.37 ± 0.21) > FD (4.71 ± 0.25) > YS (4.56 ± 0.3).

Additionally, the suitability of the six tea cultivars was investigated by further analyzing their aroma variations. Obviously, in terms of flowery and fruity scent qualities, the suitable oolong tea cultivars (HD, MZ, TGY) clearly outperformed the unsuitable cultivars (WNZ, YS, FD), but they showed no significant difference in fresh odor. A PCA score-plot is shown in Fig. 2C, and the scores of products from suitable cultivars are distributed on the positive semi-axis of the x-axis (PCA1), but it is the opposite for the unsuitable varieties. These observations demonstrated that the scores of each fragrance attribute could significantly separate the oolong tea product groupings (Bray Curtis, Adonis: $R^2 = 0.5707$, $P = 0.001$).

The relationship between various fragrance qualities and overall evaluation was investigated to demonstrate the significance of aroma attributes (Fig. 2B and Table A.6). The overall evaluation of oolong tea was seen to have a substantial association with floral odor ($P = 0.832$), a weak correlation with fruity odor ($P = 0.431$), a limited correlation with sweet odor ($P = 0.379$), and no correlation with fresh odor ($P < 0.3$). This suggested that the overall quality evaluation of oolong tea is not affected by fresh odor, but by floral, fruity, and sweet odors.

Aroma content of oolong tea by HS-SPME-GC–MS

The GC–MS data demonstrated that the content of volatile compounds varied in the oolong tea samples of various cultivars (Table A.7). Through suitability grouping, 47 differential compounds were discovered, including 12 VTs ((E)-nerolidol, citral, α-terpineol and so on), 8 AADVs (benzaldehyde, benzyl alcohol, indole and so on), 13 FADVs (hexanal, (E)-3-hexen-1-ol, heptanal, jasmine lactone and so on), 2 CDVs (dihydropseudoionone, 6-methyl-5-hepten-2-one), and 12 other compounds (octanal, 2-ethyl-1-hexanol, (E)-2-octenal, (E)-2-decenal, and so on).

Characteristic aroma compounds of oolong tea by AEDA-GC-O-MS

Additionally, the variations in aroma compounds in six oolong tea samples were investigated by AEDA-GC-O-MS. Among the 34 peaks (compounds), 17 compounds with a FD factor above or equal to 4^4 (FD $\geq 4^4$) were discovered and identified by sniffing (Table 1 and Table A.3). For HD and MZ samples, all the 17 compounds had a high FD factor (FD $\geq 4^4$). For TGY samples, except for (Z)-3-Hexenyl benzoate, all the other 16 compounds had a high FD factor (FD $\geq 4^4$). Meanwhile, several compounds were detected in the other three varieties of oolong tea with a FD factor $< 4^4$. Specifically, the compounds with a FD factor $< 4^4$ were benzaldehyde, methyl salicylate, (Z)-3-hexenyl benzoate, (E)-nerolidol, geraniol, and jasmine lactone in WNZ; (E)-3-hexen-1-ol, (E, E)-2,4-heptadienal, (Z)-3-hexenyl benzoate, jasmine lactone, and indole in FD; (E, E)-2,4-heptadienal, benzyl alcohol, (E)-nerolidol, jasmine lactone, and indole in YS. In the grouping (different suitability) test (Table 1), only jasmine lactone and indole showed a significant difference (Kruskal-Wallis test $P < 0.05$). Indole, jasmine lactone, β-ionone, and linalool have recently been shown to be strong (FD $\geq 4^4$) odor components in oolong tea (*Camellia sinensis* var. *Jinxuan*) as detected by AEDA-GC-O-MS (Gui et al., 2015). The substances with floral and fruity aromas (such as benzaldehyde, geraniol, (E)-nerolidol, jasmine lactone, indole) had a higher FD factor in suitable oolong tea cultivars, but they were not always the substances with a high FD factor in unsuitable varieties, suggesting that these compounds had a direct impact on the differences in scent recognition between tea cultivars. In this

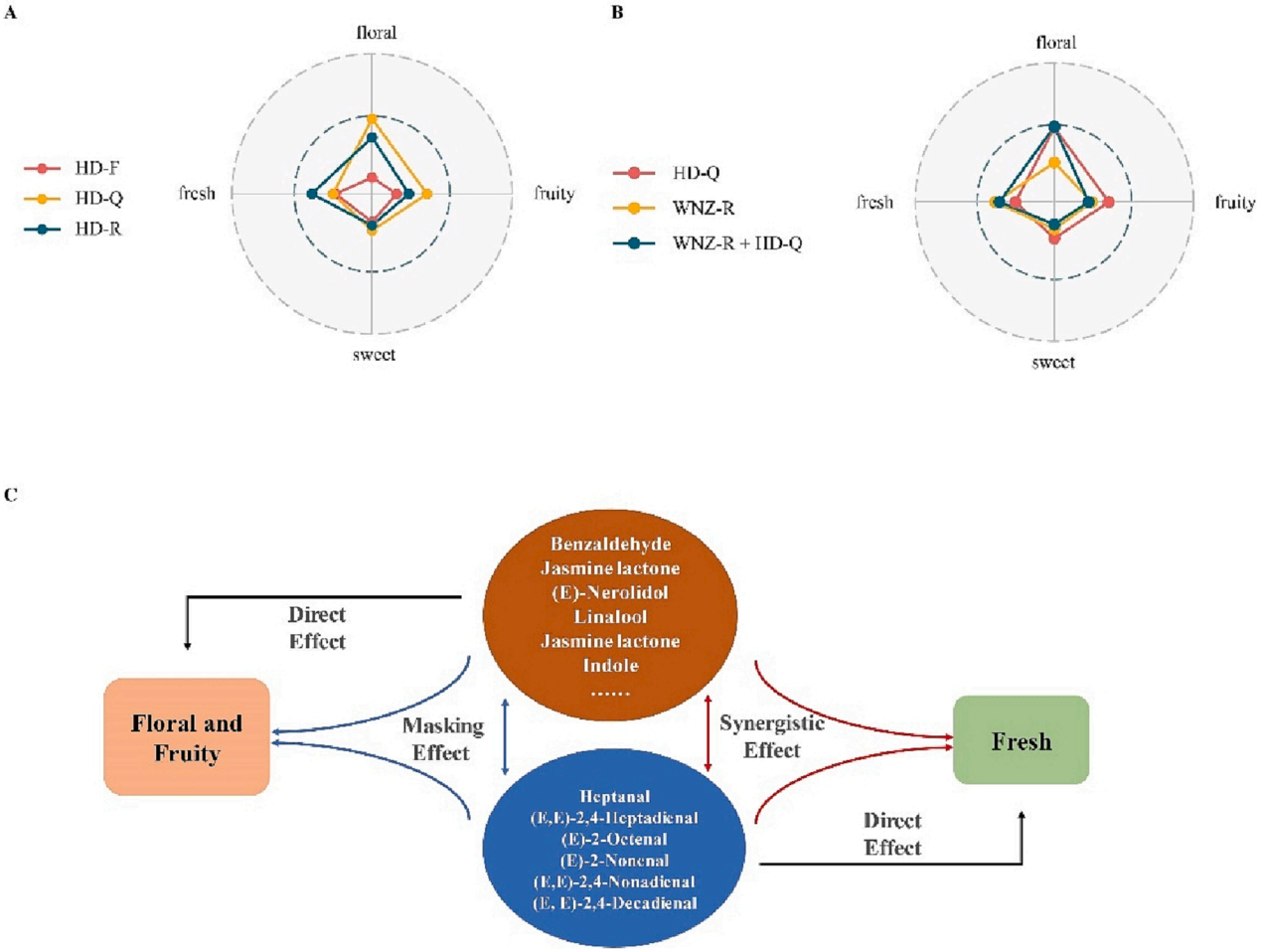


Fig. 4. Radar chart for the scores of samples in the experiments to validate the effects of volatile compounds on aroma properties. (A) The first experiment in the HD recombination system. (B) The second experiment in the WNZ recombination system. The gray dotted line in the outer circle represents 9 points; the indigo blue dotted line in the inner circle, 5 points; the center of the radar map, 1 point. (C) Schematic diagram for the relationship between aroma compound interactions and sensory attributes. The compounds in the red area in the middle of the image was defined as Group (1), and the compounds in the blue area in the middle of the image was defined as Group (2). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

investigation, the compounds (6-methyl-5-hepten-2-one, linalool, (Z)-furan linalool oxide, linalool pyranoid oxide, benzeneacetaldehyde, phenylethyl alcohol, and (E)-isoeugenol) were found to have a high FD factor (FD $\geq 4^4$) in all cultivars, indicating that they were stable between cultivars and belonged to the distinctive aroma components of oolong tea.

Overall, indole, benzaldehyde, methyl salicylate, (Z)-3-hexenyl benzoate, (E)-nerolidol, geraniol, jasmine lactone, 3-hexen-1-ol, (E, E)-2,4-heptadienal, and benzyl alcohol were the primary compounds to induce scent variances in the various oolong tea cultivars, while 6-methyl-5-hepten-2-one, linalool, (Z)-furan linalool oxide, linalool oxide pyranoid, benzeneacetaldehyde, phenylethyl alcohol, (E)-isoeugenol were the substances to constitute similar aroma attributes among cultivars.

Aroma recombination

Fig. 3 shows the scores of aroma attributes for both the original samples and reconstructed samples evaluated by the reviewers, and a strong correlation could be observed between them based on the scores of the four aroma attributes (fresh, floral, fruity, and sweet) for each cultivar: MZ (0.893), TGY (0.852), HD (0.881), YS (0.848), WNZ (0.661), and FD (0.986) (Table A.9), suggesting that the recombination system can restore the aroma state of the sample itself, and the selected aroma active compounds basically constitute the aroma characteristics of oolong tea. In Table A.8, consistent with the scores of the original samples, the reconstituted samples of suitable oolong tea cultivars (HD, MZ, TGY) had higher scores in floral and fruity odor characteristics, in contrast to less pronounced flower and fruit fragrances for the reconstituted samples of unsuitable oolong tea cultivars (WNZ, YS, FD), but they also exhibited obvious fresh odor. Collectively, the six cultivars had no obvious difference in the scores of fresh odors, indicating that the floral and fruity aromas obscured the fresh odor, endowing the samples from suitable cultivars mainly with the fragrance of flowers and fruits.

Aroma omission

We chose the HD recombinant samples for aroma omission tests to characterize the contribution of various components to overall aroma. Table 2 displays the statistical results for the effect of each component deletion on the original recombinant system, and 17 substances were found to significantly (Wilcox. Test. $P < 0.05$) impact the recombinant system after deletion. Among them, benzaldehyde, 6-methyl-5-hepten-2-one, benzeneacetaldehyde, methyl salicylate, β-cyclocitral, (E)-isoeugenol, β-ionone, and (E)-nerolidol were described as having floral or fruity aroma, but their omission systems enhanced the characteristics of fresh odor. As single compounds, heptanal, (E, E)-2,4-hexadienal, (E, E)-2,4-heptadienal, (E)-2-octenal, (E)-2-nonenal, (E, E)-2,4-nonadienal,

and (E, E)-2,4-decadienal had green or fresh odor, but their absence was described as enhancing fresh odor while exhibiting floral odor. Additionally, linalool and indole had a floral or fruity aroma and a slight sweet aroma on their own, but their omission systems were shown to decrease fresh odor and increase sweet odor. Moreover, the omission system of jasmine lactone was described as enhancing fresh odor and stimulation.

Interactions between odor compounds

Based on the above results, the substances contributing to floral and fruity aromas were classified as Group (1) and those contributing to fresh aroma were classified as Group (2) for verification experiments (Fig. 4 and Table A.10), thus allowing us to further explore the relationship between aroma attributes and components in oolong tea. In Table A.10, the Group1compounds were shown to significantly improve the floral odor and fresh odor, as indicated by the deletion experiment in the HD recombination system, where the scores of floral and fresh odors were significantly lower in HD-F than in the control HD-R. When compared with the control HD-R, HD-Q was significantly higher in scores of floral and fruity odors, but significantly lower in the score for fresh odor, demonstrating that Group (2) aroma compounds could increase the intensity of fresh odor and lessen the intensity of floral and fruity odors in the system. Moreover, the floral odor intensity was seen to be noticeably higher in WNZ-R + HD-Q (4.88) (group (1) was directly added to WNZ reconstituted samples) than in the control WNZ-R (2.55), suggesting that group (1) may greatly increase the floral odor intensity of WNZ reconstituted samples. In addition to demonstrating the contributions of aroma-active compounds in HD samples to the qualities of floral, fruity, and fresh scents, the results also suggested the possibility that these compounds may interact with one another, i.e., the compounds contributing to fresh odor may mask the strength capabilities of the compounds contributing to floral and fruity odors, while the compounds contributing to floral and fruity odors may work in concert with the compounds contributing to fresh odor to enhance the intensity of fresh aroma attribute in the system. In recent years, the research on the interaction between odorant compounds and human perception has become a hot topic. The present study revealed for the first time the influence of the interaction of aroma components on the perception of fresh, floral, and fruity aromas of oolong tea, providing an important clue for further related research.

Conclusions

The present study sheds light on the differences in aroma composition between oolong, black, and green tea owing to their distinct processing technologies. The aroma composition of Qingxiang oolong tea was investigated for the first-time using tea cultivars with varying suitability. Our findings revealed that the aroma of Qingxiang oolong tea was characterized by numerous interactive odor compounds, with fresh odor compounds forming the foundation and floral and fruity odor compounds as distinct features. So, the aroma system of oolong tea was significantly influenced by processing and cultivars. Notably, the interaction between the molecules that constitute the aroma complex was identified as a crucial component of the system. This study provided a data foundation for quality control and assessment of Qingxiang Oolong tea in the industry. Furthermore, our analysis of tea products using the suitability of tea tree cultivars as the foundation offers valuable insights and approaches for enhancing tea tree cultivars and tea processing technology.

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.

Acknowledgements

The authors appreciate the financial supporting provided by The National Key R & D Program of China (Project No. 2021YFD1000401) and National Natural Science Foundation of China (32272769).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2023.100730.

References

Bonneau, A., Boulanger, R., Lebrun, M., Maraval, I., & Gunata, Z. (2016). Aroma compounds in fresh and dried mango fruit (Mangifera indicaL. cv. Kent): Impact of drying on volatile composition. *International Journal of Food Science & Technology, 51*(3), 789–800. https://doi.org/10.1111/ijfs.13038

Chen, S., Xie, P., Li, Y., Wang, X., Liu, H., Wang, S., … Yu, X. (2021). New Insights into Stress-Induced β-Ocimene Biosynthesis in Tea (Camellia sinensis) Leaves during Oolong Tea Processing. *Journal of Agricultural and Food Chemistry, 69*(39), 11656–11664. https://doi.org/10.1021/acs.jafc.1c04378

Cui, J., Katsuno, T., Totsuka, K., Ohnishi, T., Takemoto, H., Mase, N., … Tong, H. (2016). Characteristic Fluctuations in Glycosidically Bound Volatiles during Tea Processing and Identification of Their Unstable Derivatives. *Journal of Agricultural and Food Chemistry, 64*(5), 1151–1157. https://doi.org/10.1021/acs.jafc.5b05072

Cui, J., Zhai, X., Guo, D., Du, W., Gao, T., Zhou, J., … Song, C. (2022). Characterization of Key Odorants in Xinyang Maojian Green Tea and Their Changes During the Manufacturing Process. *Journal of Agricultural and Food Chemistry, 70*(1), 279–288. https://doi.org/10.1021/acs.jafc.1c06473

Del Barrio Galán, R., Bueno-Herrera, M., De La Cuesta, P. L., & Pérez-Magariño, S. (2022). Volatile composition of Spanish red wines: Effect of origin and aging time. *European Food Research and Technology, 248*(7), 1903–1916. https://doi.org/10.1007/s00217-022-04014-x

Du, L., Wang, C., Li, J., Xiao, D., Li, C., & Xu, Y. (2013). Optimization of Headspace Solid-Phase Microextraction Coupled with Gas Chromatography-Mass Spectrometry for Detecting Methoxyphenolic Compounds in Pu-erh Tea. *Journal of Agricultural and Food Chemistry, 61*(3), 561–568. https://doi.org/10.1021/jf304470k

Dubrow, G. A., Forero, D. P., & Peterson, D. G. (2022). Identification of volatile compounds correlated with consumer acceptability of strawberry preserves: Untargeted GC–MS analysis. *Food Chemistry, 378*, Article 132043. https://doi.org/10.1016/j.foodchem.2022.132043

Engel, W., Bahr, W., & Schieberle, P. (1999). Solvent assisted flavour evaporation - a new and versatile technique for the careful and direct isolation of aroma compounds from complex food matrices. *European Food Research and Technology, 209*(3–4), 237–241. https://doi.org/10.1007/s002170050486

Farré-Armengol, G., Filella, I., Llusià, J., & Peñuelas, J. (2017). β-Ocimene, a Key Floral and Foliar Volatile Involved in Multiple Interactions between Plants and Other Organisms. *Molecules, 22*(7), 1148. https://doi.org/10.3390/molecules22071148

Feng, T., Sun, J., Wang, K., Song, S., Chen, D., Zhuang, H., … Ho, C.-T. (2022). Variation in Volatile Compounds of Raw Pu-Erh Tea upon Steeping Process by Gas Chromatography-Ion Mobility Spectrometry and Characterization of the Aroma-Active Compounds in Tea Infusion Using Gas Chromatography–Olfactometry–Mass Spectrometry. *Journal of Agricultural and Food Chemistry*. https://doi.org/10.1021/acs.jafc.2c04342

Feng, Z., Li, Y., Li, M., Wang, Y., Zhang, L., Wan, X., & Yang, X. (2019). Tea aroma formation from six model manufacturing processes. *Food Chemistry, 285*, 347–354. https://doi.org/10.1016/j.foodchem.2019.01.174

Gui, J., Fu, X., Zhou, Y., Katsuno, T., Mei, X., Deng, R., … Yang, Z. (2015). Does Enzymatic Hydrolysis of Glycosidically Bound Volatile Compounds Really Contribute to the Formation of Volatile Compounds During the Oolong Tea Manufacturing Process? *Journal of Agricultural and Food Chemistry, 63*(31), 6905–6914. https://doi.org/10.1021/acs.jafc.5b02741

Guo, X., Ho, C.-T., Wan, X., Zhu, H., Liu, Q., & Wen, Z. (2021). Changes of volatile compounds and odor profiles in Wuyi rock tea during processing. *Food Chemistry, 341*, Article 128230. https://doi.org/10.1016/j.foodchem.2020.128230

Guo, X., Schwab, W., Ho, C.-T., Song, C., & Wan, X. (2022). Characterization of the aroma profiles of oolong tea made from three tea cultivars by both GC–MS and GC-IMS. *Food Chemistry, 376*, Article 131933. https://doi.org/10.1016/j.foodchem.2021.131933

He, C., Li, Y., Zhou, J., Yu, X., Zhang, D., Chen, Y., … Yu, Z. (2022). Study on the Suitability of Tea Cultivars for Processing Oolong Tea from the Perspective of Aroma Based on Olfactory Sensory, Electronic Nose, and GC-MS Data Correlation Analysis. *Foods, 11*(18), 2880. https://doi.org/10.3390/foods11182880

Hegde, M., Oliveira, J. N., da Costa, J. G., Loza-Reyes, E., Bleicher, E., Santana, A. E. G., … Birkett, M. A. (2012). Aphid antixenosis in cotton is activated by the natural plant defence elicitor cis-jasmone. *Phytochemistry, 78*, 81–88. https://doi.org/10.1016/j.phytochem.2012.03.004

Ho, C.-T., Zheng, X., & Li, S. (2015). Tea aroma formation. *Food Science and Human Wellness, 4*(1), 9–27. https://doi.org/10.1016/j.fshw.2015.04.001

Hu, C.-J., Li, D., Ma, Y.-X., Zhang, W., Lin, C., Zheng, X.-Q., … Lu, J.-L. (2018). Formation mechanism of the oolong tea characteristic aroma during bruising and withering treatment. *Food Chemistry, 269*, 202–211. https://doi.org/10.1016/j.foodchem.2018.07.016

Jing, T., Du, W., Gao, T., Wu, Y., Zhang, N., Zhao, M., … Song, C. (2021). Herbivore-induced DMNT catalyzed by CYP82D47 plays an important role in the induction of JA -dependent herbivore resistance of neighboring tea plants. *Plant, Cell & Environment, 44*(4), 1178–1191. https://doi.org/10.1111/pce.13861

Liang, S., Wang, F., Granato, D., Zhong, X., Xiao, A.-F., Ye, Q., … Xu, Y.-Q. (2022). Effect of β-glucosidase on the aroma of liquid-fermented black tea juice as an ingredient for tea-based beverages. *Food Chemistry, 134201*. https://doi.org/10.1016/j.foodchem.2022.134201

Liu, C., Cheng, Y., Zhang, H., Deng, X., Chen, F., & Xu, J. (2012). Volatile Constituents of Wild Citrus Mangshanyegan (Citrus nobilis Lauriro) Peel Oil. *Journal of Agricultural and Food Chemistry, 60*(10), 2617–2628. https://doi.org/10.1021/jf2039197

Liu, P.-P., Feng, L., Xu, Y.-Q., Zheng, L., Yin, P., Ye, F., … Zheng, P.-C. (2023). Characterization of stale odor in green tea formed during storage: Unraveling improvements arising from reprocessing by baking. *LWT, 174*, Article 114458. https://doi.org/10.1016/j.lwt.2023.114458

Liu, Y., Chen, Q., Liu, D., Yang, L., Hu, W., Kuang, L., … Liu, Y. (2023). Multi-omics and enzyme activity analysis of flavour substances formation: Major metabolic pathways alteration during Congou black tea processing. *Food Chemistry, 403*, Article 134263. https://doi.org/10.1016/j.foodchem.2022.134263

Mahanta, P. K., & Baruah, H. K. (1992). Theaflavin pigment formation and polyphenol oxidase activity as criteria of fermentation in orthodox and CTC teas. *Journal of Agricultural and Food Chemistry, 40*(5), 860–863. https://doi.org/10.1021/jf00017a032

Mu, D., Cui, L., Ge, J., Wang, M.-X., Liu, L.-F., Yu, X.-P., … Han, B.-Y. (2012). Behavioral responses for evaluating the attractiveness of specific tea shoot volatiles to the tea green leafhopper. *Empoaca vitis. Insect Science, 19*(2), 229–238. https://doi.org/10.1111/j.1744-7917.2011.01476.x

Schuh, C., & Schieberle, P. (2006). Characterization of the Key Aroma Compounds in the Beverage Prepared from Darjeeling Black Tea: Quantitative Differences between Tea Leaves and Infusion. *Journal of Agricultural and Food Chemistry, 54*(3), 916–924. https://doi.org/10.1021/jf052495n

Teranishi, R., Buttery, R. G., & Guadagni, D. G. (1974). ODOR QUALITY AND CHEMICAL STRUCTURE IN FRUIT AND VEGETABLE FLAVORS. *Annals of the New York Academy of Sciences, 237*(1 Odors), 209-216. https://doi.org/10.1111/j.1749-6632.1974.tb49855.x.

Wang, J., Li, M., Wang, H., Huang, W., Li, F., Wang, L., … Wan, X. (2022). Decoding the Specific Roasty Aroma Wuyi Rock Tea (Camellia sinensis: Dahongpao) by the Sensomics Approach. *Journal of Agricultural and Food Chemistry*. https://doi.org/10.1021/acs.jafc.2c02249

Wang, K., Liu, F., Liu, Z., Huang, J., Xu, Z., Li, Y., … Yang, X. (2011). Comparison of catechins and volatile compounds among different types of tea using high performance liquid chromatograph and gas chromatograph mass spectrometer. *International Journal of Food Science and Technology, 46*(7), 1406–1412. https://doi.org/10.1111/j.1365-2621.2011.02629.x

Wang, L.-F., Lee, J.-Y., Chung, J.-O., Baik, J.-H., So, S., & Park, S.-K. (2008). Discrimination of teas with different degrees of fermentation by SPME–GC analysis of the characteristic volatile flavour compounds. *Food Chemistry, 109*(1), 196–206. https://doi.org/10.1016/j.foodchem.2007.12.054

Xie, S., Wu, G., Ren, R., Xie, R., Yin, H., Chen, H., … Ge, M. (2023). Transcriptomic and metabolic analyses reveal differences in monoterpene profiles and the underlying molecular mechanisms in six grape varieties with different flavors. *LWT, 174*, Article 114442. https://doi.org/10.1016/j.lwt.2023.114442

Yang, Z., Baldermann, S., & Watanabe, N. (2013). Recent studies of the volatile compounds in tea. *Food Research International, 53*(2), 585–599. https://doi.org/10.1016/j.foodres.2013.02.011

Yao, H., Su, H., Ma, J., Zheng, J., He, W., Wu, C., … Zhou, Q. (2023). Widely targeted volatileomics analysis reveals the typical aroma formation of Xinyang black tea during fermentation. *Food Research International, 164*, Article 112387. https://doi.org/10.1016/j.foodres.2022.112387

Zeng, L., Liao, Y., Li, J., Zhou, Y., Tang, J., Dong, F., & Yang, Z. (2017). α-Farnesene and ocimene induce metabolite changes by volatile signaling in neighboring tea (Camellia sinensis) plants. *Plant Science, 264*, 29–36. https://doi.org/10.1016/j.plantsci.2017.08.005

Zeng, L., Zhou, X., Su, X., & Yang, Z. (2020). Chinese oolong tea: An aromatic beverage produced under multiple stresses. *Trends in Food Science & Technology, 106*, 242–253. https://doi.org/10.1016/j.tifs.2020.10.001

Zeng, L., Zhou, Y., Gui, J., Fu, X., Mei, X., Zhen, Y., … Yang, Z. (2016). Formation of Volatile Tea Constituent Indole During the Oolong Tea Manufacturing Process. *Journal of Agricultural and Food Chemistry, 64*(24), 5011–5019. https://doi.org/10.1021/acs.jafc.6b01742

Zhang, L., Zeng, Z., Zhao, C., Kong, H., Lu, X., & Xu, G. (2013). A comparative study of volatile components in green, oolong and black teas by using comprehensive two-dimensional gas chromatography–time-of-flight mass spectrometry and multivariate data analysis. *Journal of Chromatography A, 1313*, 245–252. https://doi.org/10.1016/j.chroma.2013.06.022

Zhu, J., Chen, F., Wang, L., Niu, Y., Yu, D., Shu, C., … Xiao, Z. (2015). Comparison of Aroma-Active Volatiles in Oolong Tea Infusions Using GC–Olfactometry, GC–FPD, and GC–MS. *Journal of Agricultural and Food Chemistry, 63*(34), 7499–7510. https://doi.org/10.1021/acs.jafc.5b02358