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Aroma formation and transformation during production of Fenghuang Dancong oolong tea

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Abstract

Fenghuang Dancong tea is a type of semi-fermented oolong tea with unique floral and fruity flavours. However, little is known about aroma formation in Fenghuang Dancong tea during production. In the present work, headspace solid phase microextraction (HS-SPME) combined with gas chromatography mass spectroscopy (GC-MS) was used to extract, detect, and analyse aroma compounds in Fenghuang Dancong tea during the six stages of its production, namely fresh leaves, withering, rocking, de-enzyming, rolling, and baking. In addition, aroma compound changes were statistically validated, and sensory implications were discussed. In total, 131 aroma compounds were extracted and identified throughout production of Fenghuang Dancong tea. The content of alcohols and esters in fresh leaves, withering, and rocking stages was higher than that in de-enzyming, rolling, and baking stages. The content of aroma compounds such as phenylethanol, nerolidol, β-Ionone, and phenylacetaldehyde increased significantly throughout production of Fenghuang Dancong tea. These aroma compounds were the major contributors to the floral and fruity aroma of Fenghuang Dancong tea, and can be thus used as markers of aroma quality of Fenghuang tea during production. In addition, hierarchical cluster analysis (HCA) and principal component analysis (PCA) indicated that aroma compounds extracted during production of Fenghuang Dancong tea clustered into four distinct categories. The findings highlighted the potential of using aroma compounds as markers for aroma quality control during production of Fenghuang Dancong tea, with rocking and de-enzyming stages being identified as the key steps for aroma formation in Fenghuang Dancong tea.

Introduction

Tea is a well-appreciated beverage worldwide. Global tea annual consumption is approximately hundreds of billions of tons (Mao *et al*., 2018). Tea quality is mainly determined by its aroma and taste. It is widely known that tea taste is determined by non-volatile compounds, such as amino acids and polyphenols, among others, while tea aroma is determined by volatile aroma compounds (Zhang *et al*., 2019). Therefore, different teas have different aroma characteristics. Moreover, tea aroma is mainly affected by plant variety, origin, production, and processing technology. Tea aroma affects tea quality as well as plays an important role in sales prospects (Zhu *et al*., 2015).

Fenghuang Dancong tea is a type of semi-fermented oolong tea produced on Fenghuang Mountain in Chao'an County, Guangdong Province, China, where it has been cultivated for thousands of years. The fragrance of Fenghuang Dancong tea is elegant and clear, suggestive of flowers and fruits, being well appreciated by consumers (Chen *et al*., 2021a). Considerable progress has been made on the research of aroma compounds in Fenghuang Dancong tea. In a previous study, it was found that its main aroma compounds include linalool oxide I, linalool oxide II, linalool, dehydrolinalool, benzonitrile, geraniol, indole, nerolidol, and others (Li, 2022). Thus, research on the aroma profile of Fenghuang Dancong tea plays an important role in understanding its aroma characteristics.

It has been described that the aroma of Fenghuang Dancong tea mainly originates during tea production. Production of Fenghuang tea includes several stages, *i.e.*, withering, rocking, de-enzyming, rolling, and baking. The characteristic floral and fruity aroma compounds of Fenghuang Dancong tea

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mainly arise from enzymatic reactions and thermal transformations occurring during tea production. For instance, during the rocking stage, enzymatic reactions occur vigorously due to the intense collision between tea leaves, thereby promoting production of characteristic tea aroma. During the withering, de-enzyming, and baking stages, aroma compounds are also produced and transformed due to leaf dehydration and thermal chemical reactions. However, changes in characteristic aroma production and transformation in each stage of Fenghuang Dancong tea production have been insufficiently explored which hinders efficient quality control.

The analysis of aroma compounds in tea requires choosing a suitable method for tea aroma extraction. Commonly, simultaneous distillation extraction (SDE) is adopted for tea aroma extraction. However, it has many limitations, such as long extraction time, consumption of large volumes of samples and solvents, and easy decay and transformation of samples at high temperature, which limits its application in tea aroma extraction (Sheibani *et al*., 2016a). Headspace solid phase microextraction (HS-SPME) is a widely used volatile gas extraction method, which has the advantages of short extraction time, low extraction temperature, and low consumption of sample and solvent, having been applied to a variety of research studies exploring tea aroma (Tan *et al*., 2019; Chen *et al*, 2021b). Gas chromatography mass spectroscopy (GC-MS) is another commonly used method for aroma profile analysis. The identification and quantification of aroma compounds have the advantages of high stability, good accuracy, and strong repeatability, and are widely used in aroma profile analysis of tea (Liu *et al*., 2014). Therefore, the combined use of HS-SPME and GC-MS has become a commonly used method in tea aroma profiling.

To maximise the extraction of useful information from a large amount of data on aroma compounds, the application of chemometrics methods, such as hierarchical cluster analysis (HCA) or principal component analysis (PCA), is particularly important. At present, the combined use of instrumental analysis and chemometrics has been applied in the aroma identification of different varieties of tea. However, it has not yet been applied to the analysis of aroma compounds during production of Fenghuang Dancong tea.

Therefore, the aim of the present work was to apply HS-SPME combined with GC-MS in the extraction, analysis, and identification of aroma compounds in Fenghuang Dancong tea during production, as well as to explore the changes in the content of important aroma compounds. The present work would provide a theoretical reference for assessing the quality of Fenghuang Dancong tea during production and processing.

Materials and methods

Sample materials

Fresh leaves (one bud and two leaves) were obtained on April 15th, 2021 from the Fenghuang Dancong tea plants grown on the Fenghuang Mountain, Chao'an County, Guangdong Province, China. Plants were assessed by experts, being identified as a Zhilanxiang type of Fenghuang Dancong tea plant. Fresh leaves were prepared in strict accordance with the traditional processing method of Fenghuang Dancong tea, whose steps included: (i) collection of fresh leaves; (ii) sun-withering at 25°C for 1 h; (iii) rocking for 6 h; (iv) de-enzyming at 220°C for 4 min; (v) rolling at 50 rpm for 8 min; and (vi) baking at 80°C for 2 h. Samples were collected during each production step, totalling six steps. In order to ensure the accuracy of the experiment, four biological replicates were performed in each step. After rapid freeze-drying, samples were refrigerated at 4°C for further analysis.

HS-SPME extraction of tea aroma compounds

Briefly, 1.0 g of samples was placed in a 20-mL sample vial, followed by the addition of 5 mL of boiling ultra-pure water and 5.0 μL of 100 ng/μL ethyl decanoate solution (in hexane) as the internal standard. Vials were then sealed with a sample bottle cap with Teflon/silica gel spacer, and then placed in a water bath at 65°C for 5 min. The manual sampler equipped with an aged PDMS/DVB/Carboxen extraction head was inserted into the vials, and after 60 min, this was removed. HS-SPME extraction was conducted at 65°C, and the column was immediately inserted into the sample inlet of the GC system for desorption for 5 min. Data were collected simultaneously.

GC-MS analysis

For GC-MS analysis, the TRACE ISQ™ (Thermo Scientific) was used based on the following conditions: GC column, TG-5MS (30 m × 0.25 mm × 0.25 μm); injection port temperature, 250°C; carrier gas, high-purity helium (purity > 99.9999%); flow rate, 1 mL/min; no split injection. Temperature change was employed as follows: 60°C for 1 min; then increased to 230°C at the rate of 10°C/min, and maintained for 10 min; and then increased to 300°C at the rate of 40°C/min.

MS analysis was conducted using an electron ionisation source based on the following conditions: ion source temperature, 250°C; MS transmission line temperature, 250°C; scanning time, 0.3 s; and mass scanning range, 40 - 400 AMU.

Standards

Standards for linalool, linalool oxides, dehydrolinalool, nerolidol, geraniol, *(Z)*-3-Hexen-1-ol, phenylacetaldehyde, phenylethyl alcohol, *α*-terpineol, *β*-ionone, *α*-farnesene, *cis*-3-hexenyl benzoate, phytol, indole, and benzonitrile were purchased from ANPEL Laboratory Technologies (Shanghai, China).

Data analysis

The National Institute of Standards and Technology (NIST) database was used to retrieve data obtained from GC-MS analysis, and compounds with a matching degree greater than 70 were considered. The retention index (RI) of each component was calculated based on retention time of the component and the retention time of adjacent *n*-alkanes (C7-C40, ANPEL Laboratory Technologies, Shanghai, China). In addition, the RI of each chromatographic peak was compared with RIs available in the literature (Guo *et al*., 2018; Liu *et al*., 2018). Important aroma compounds were confirmed based on chemical standards. The peak area normalisation method was used for performing quantitative analysis of aroma compounds. Significant differences among tea compounds were analysed using SPSS software. HCA was performed using SPSS version 22 (IBM, USA), and PCA was conducted using SIMCA 14.1(Umetrics, Sweden).

Results and discussion

GC-MS results

GC-MS results of volatile compounds in 24 Fenghuang Dancong tea samples collected in six different production stages are shown in Table 1. A total of 131 volatile aroma compounds were detected and segregated by production step: 64 compounds in fresh leaves, 90 in sun-withering, 104 in rocking, 77 in de-enzyming, 74 in rolling, and 78 in baking. Among these volatile compounds, 40 esters, 27 alcohols, 22 alkanes, 14 alkenes, nine ketones, four acids, three aldehydes, and 12 other substances were identified. Furthermore, 43 aroma compounds were found to be commonly shared among samples from all six production steps. Among shared aroma compounds, those with higher relative contents were phenylacetaldehyde, linalool, linalool oxide I, indole, geraniol, nerolidol, and *α*-farnesene. Volatile aroma compounds with high relative content in fresh leaves were 3-hexene-1-alcohol, linalool, linalool oxide I, nerolidol, and phytol. In contrast, aroma compounds with high relative content in samples in the withering stage were 3-hexene-1-alcohol, linalool, phenylacetaldehyde, indole, *α*-farnesene, nerolidol, phytol, among others. In the rocking stage, aroma compounds with high relative content were 3-hexene-1-alcohol, 2-hexene-1-alcohol, phenylacetaldehyde, linalool, linalool oxide I, phenylethanol, indole, *α*-farnesene, nerolidol, and phytol, among others. The main aroma compounds along with their respective relative contents were comparable in the de-enzyming, rolling, and baking stages, and mainly included styrene, phenylacetaldehyde, linalool, phenylethanol, benzonitrile, epoxy linalool, indole, jasmine lactone, *α*-farnese, nerolidol, phytol, among others. The contents of various alcohols, ketones, esters, aldehydes, alkanes, hydrocarbons, acids, alkenes, and other aroma compounds in tea samples in the six production stages of Fenghuang Dancong tea are shown in Figure 1.

HCA analysis

HCA is used to classify and analyse objects according to a correlation between data points based on an unsupervised algorithm for pattern recognition (Chen *et al*., 2021c). In the present work, SPSS software was used to further determine the degree of aggregation and dispersion of samples in each production stage of Fenghuang Dancong tea. The clustering method was group connection, and the default distance measure was square Euclidean distance. GC-MS data of volatile aroma compounds in Fenghuang Dancong tea during production were systematically clustered and analysed, and a tree classification diagram was obtained (Figure 2). When the class spacing was 15, 24 samples were divided into four categories: the fresh leaves category clustered with four tea samples in the fresh leaf stage; the withering category clustered with four tea samples

Table 1. Relative content of key volatile aroma components during production of Fenghuang Dancong tea (%).

No.	RI	Compound	Identification	Fresh leaf	Sun-withering	Rocking	De-enzyming	Rolling	Baking
1	857	*(Z)*-3-Hexen-1-ol	MS, RI, S	20.03 ± 12.98	29.47 ± 11.99	2.54 ± 0.51	-	-	-
2	860	*(Z)*-2-Hexen-1-ol	MS, RI	-	-	14.85 ± 3.74	-	-	-
3	936	Styrene	MS, RI	-	-	-	3.66 ± 1.02	3.74 ± 0.69	3.79 ± 0.35
4	957	Benzaldehyde	MS, RI	-	-	0.37 ± 0.06	-	-	-
5	1011	*(E)*-3-Hexen-1-ol, acetate	MS, RI	2.08 ± 2.50	1.15 ± 0.99	-	-	-	-
6	1016	4-Hexen-1-ol, acetate	MS, RI	-	0.51 ± 0.67	-	-	-	-
7	1044	Benzyl alcohol	MS, RI	-	-	0.34 ± 0.11	0.24 ± 0.17	-	-
8	1052	Phenylacetaldehyde	MS, RI, S	1.44 ± 0.84	2.03 ± 0.31	5.03 ± 1.54	4.94 ± 0.66	3.48 ± 0.45	3.23 ± 0.35
9	1075	1-Octanol	MS, RI	-	0.58 ± 0.46	0.52 ± 0.06	-	-	-
10	1080	Linalool oxide I	MS, RI, S	2.12 ± 0.81	1.48 ± 0.33	2.28 ± 0.12	0.80 ± 0.17	0.53 ± 0.09	0.36 ± 0.24
11	1095	Linalool oxide II	MS, RI, S	1.44 ± 0.66	0.58 ± 0.10	1.21 ± 0.22	-	-	-
12	1104	Linalool	MS, RI, S	16.52 ± 5.71	11.34 ± 0.64	7.87 ± 0.60	4.98 ± 0.87	4.35 ± 0.70	2.58 ± 0.20
13	1111	Dehydrolinalool	MS, RI, S	-	-	-	0.43 ± 0.87	-	1.74 ± 0.22
14	1123	Phenylethyl Alcohol	MS, RI, S	-	0.39 ± 0.48	2.14 ± 0.88	1.60 ± 0.36	1.15 ± 0.19	1.02 ± 0.23
15	1147	Benzonitrile	MS, RI, S	-	1.32 ± 0.30	0.89 ± 0.61	7.15 ± 0.86	4.69 ± 0.68	3.09 ± 0.13
16	1177	Epoxylinalool	MS, RI	0.07 ± 0.13	0.31 ± 0.10	0.70 ± 0.25	1.77 ± 0.27	1.31 ± 0.10	1.31 ± 0.09
17	1200	α-Terpineol	MS, RI, S	1.66 ± 0.62	0.67 ± 0.21	0.53 ± 0.36	0.86 ± 0.26	0.72 ± 0.12	0.95 ± 0.02

No.	RI	Compound	Identification	Fresh leaf	Sun-withering	Rocking	De-enzyming	Rolling	Baking
18	1204	Methyl salicylate	MS, RI	-	0.38 ± 0.14	0.20 ± 0.15	-	-	-
19	1260	Geraniol	MS, RI, S	1.12 ± 0.46	0.79 ± 0.24	1.33 ± 0.13	0.72 ± 0.34	0.68 ± 0.13	0.85 ± 0.11
20	1265	*β*-Pinene	MS, RI	0.62 ± 0.19	0.22 ± 0.16	0.29 ± 0.03	0.11 ± 0.12	0.11 ± 0.13	0.05 ± 0.10
21	1306	Indole	MS, RI, S	0.40 ± 0.52	5.48 ± 0.90	1.90 ± 0.70	13.40 ± 1.80	12.25 ± 1.13	10.06 ± 0.95
22	1383	*(Z)*-Hexanoic acid, 3-hexenyl ester	MS, RI	1.61 ± 0.72	0.75 ± 0.40	0.33 ± 0.17	0.11 ± 0.08	0.02 ± 0.03	0.04 ± 0.09
23	1390	*(E)*-Hexanoic acid, 2-hexenyl ester	MS, RI	0.30 ± 0.09	0.55 ± 0.26	0.90 ± 0.61	0.06 ± 0.04	-	0.02 ± 0.04
24	1409	*cis*-Jasmone	MS, RI	0.07 ± 0.13	0.04 ± 0.07	0.15 ± 0.10	0.32 ± 0.05	0.21 ± 0.14	0.28 ± 0.04
25	1457	Geranyl acetone	MS, RI	-	-	-	0.08 ± 0.07	0.15 ± 0.05	0.08 ± 0.05
26	1496	*β*-Ionone	MS, RI, S	0.22 ± 0.18	-	0.07 ± 0.13	0.24 ± 0.17	0.40 ± 0.15	0.44 ± 0.08
27	1504	Jasminlactone	MS, RI	-	0.35 ± 0.28	-	1.43 ± 0.50	1.09 ± 0.20	1.40 ± 0.38
28	1514	*α*-Farnesene	MS, RI, S	0.98 ± 0.93	6.26 ± 2.27	12.37 ± 2.06	13.73 ± 1.83	14.99 ± 1.47	14.29 ± 1.30
29	1536	*δ*-Cadinene	MS, RI	0.16 ± 0.13	-	0.16 ± 0.11	0.45 ± 0.11	0.18 ± 0.13	0.72 ± 0.01
30	1570	Nerolidol	MS, RI, S	3.97 ± 2.93	4.16 ± 0.92	6.46 ± 0.85	15.36 ± 1.37	18.56 ± 2.07	18.44 ± 0.81
31	1646	*β*-Citrulene	MS, RI	-	0.06 ± 0.07	0.07 ± 0.08	0.08 ± 0.09	-	-
32	1673	*α*-Cadinol	MS, RI	-	-	0.20 ± 0.05	0.40 ± 0.15	0.29 ± 0.09	0.25 ± 0.00
33	1929	Hexadecanoic acid, methyl ester	MS, RI	0.23 ± 0.17	0.03 ± 0.07	0.13 ± 0.10	0.20 ± 0.15	0.11 ± 0.13	0.07 ± 0.15
34	2120	Phytol	MS, RI, S	6.09 ± 1.91	7.24 ± 2.58	8.63 ± 2.42	4.67 ± 1.52	4.67 ± 0.92	9.79 ± 0.69

MS: mass spectrum in comparison of using NIST library; RI: retention index in agreement with literature value; and S: identified using reference standards.

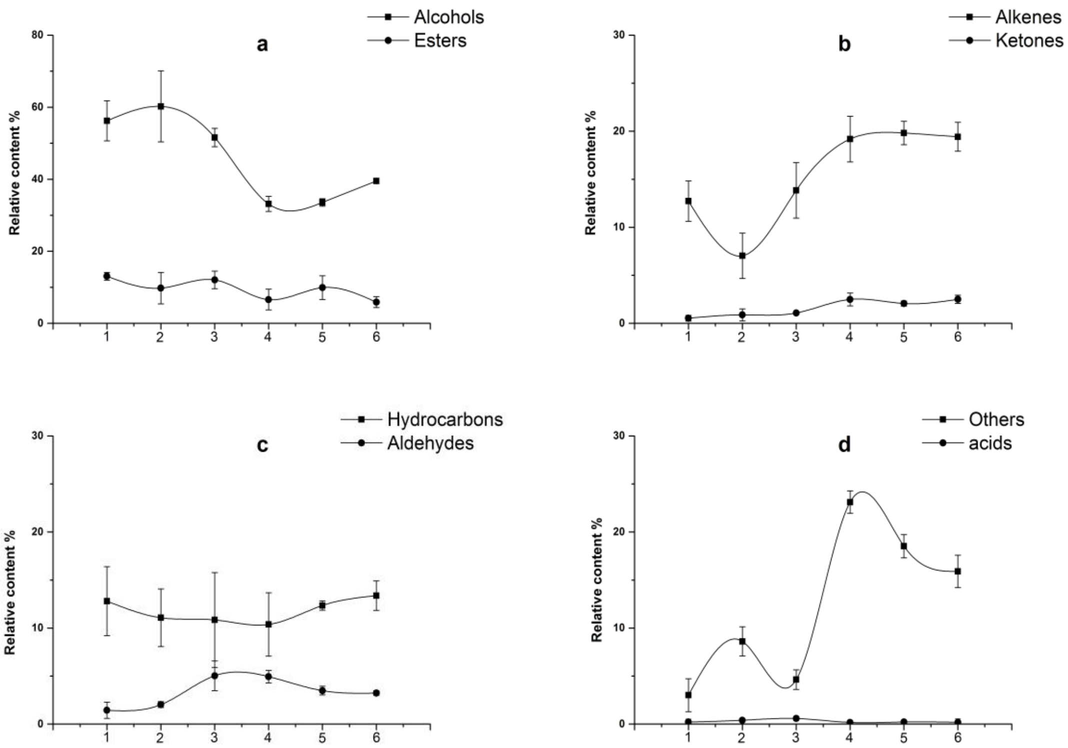

Figure 1. Dynamic changes of main aroma types of six stages during production of Fenghuang Dancong tea. 1, 2, 3, 4, 5, and 6 in abscissa axis referred to fresh, withering, rocking, de-enzyming, rolling, and baking leaves, respectively.

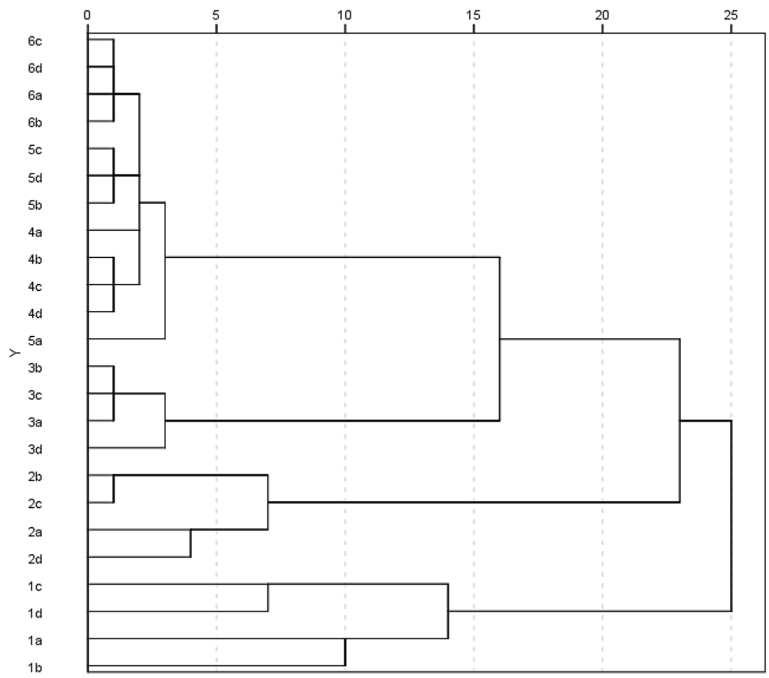

Figure 2. Hierarchical cluster analysis of aroma components during production of Fenghuang Dancong tea. 1a, 1b, 1c, and 1d represent four samples of fresh leaf stage; 2a, 2b, 2c, and 2d represent four samples of withering stage; 3a, 3b, 3c, and 3d represent four samples of rocking stage; 4a, 4b, 4c, and 4d represent four samples of de-enzyming stage; 5a, 5b, 5c, and 5d represent four samples of rolling stage; and 6a, 6b, 6c, and 6d represent four samples of baking stage.

in the withering stage; the rocking category clustered with four tea samples in the rocking stage; and the fourth category clustered with 12 tea samples in de-enzyming, rolling, and baking stages. Thus, during production of Fenghuang Dancong tea, the types and contents of aroma compounds were relatively similar in the de-enzyming, rolling, and baking stages, while great differences in terms of types and relative contents of aroma compounds in fresh leaves, withering, and rocking stages were observed.

PCA analysis

In recent years, with the advent of multivariate statistical analysis, a growing number of methods have been applied to the chemical identification of different varieties of the same compound, aroma fingerprinting, and the analysis of characteristic aroma compounds. PCA can greatly reduce data complexity and enable data visualisation (Wu *et al*., 2016; Chen *et al*., 2021d). PCA of GC-MS results indicated that the variance explained by principal component 1 was 0.243, while that by principal component 2 was 0.187; the sum of the four principal component interpretation rates was 0.592.

A seven-fold cross-validation was used in SIMCA-14 software. The results revealed that 24 samples of Fenghuang Dancong tea during production showed clear regional distribution, being thus divided into four categories: (i) the four samples in the fresh leaf stage; (ii) the four samples in the withering stage; (iii) the four samples in the rocking stage; and (iv) the twelve samples in the rolling, de-enzyming, and baking stages (Figure 3). Thus, significant differences were found in aroma compounds in Fenghuang Dancong tea in the fresh leaf, withering, and rocking stages, and the subsequent three stages, which include de-enzyming, rolling, and baking. The aroma compounds in the fresh leaf, withering, and rocking stages also significantly differed, whereas aroma compounds in the de-enzyming, rolling, and baking stages were comparable. Furthermore, these findings were highly similar to those obtained with HCA.

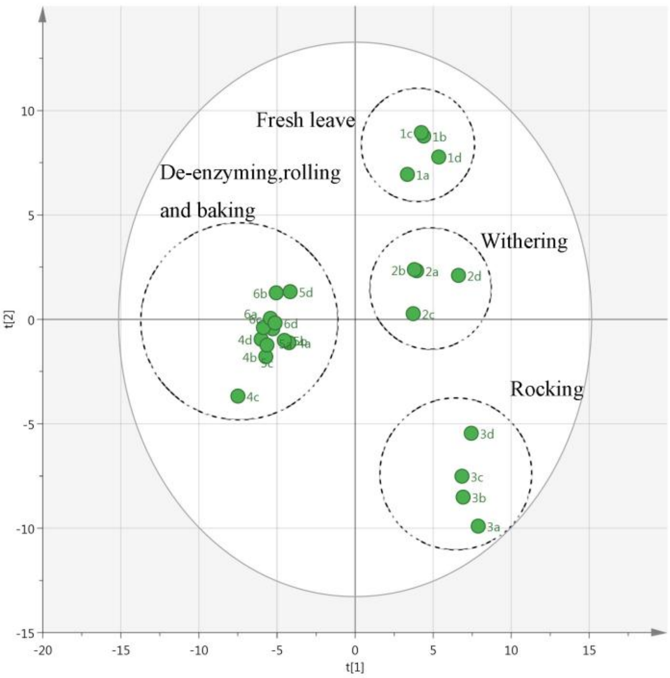

Figure 3. Principal component analysis of aroma components during production of Fenghuang Dancong tea. 1a, 1b, 1c, and 1d represent four samples of fresh leaf stage; 2a, 2b, 2c, and 2d represent four samples of withering stage; 3a, 3b, 3c, and 3d represent four samples of rocking stage; 4a, 4b, 4c, and 4d represent four samples of de-enzyming stage; 5a, 5b, 5c, and 5d represent four samples of rolling stage; and 6a, 6b, 6c, and 6d represent four samples of baking stage.

Changes in main aroma compounds during production of Fenghuang Dancong tea

Linalool and geraniol in tea plant cells are produced by D-glucose *via* the plastidic methylerythritol phosphate (MEP) pathway to geranyl pyrophosphate (GPP), and then produced by GPP under the action of terpene synthases (TPS) (Kaminaga *et al*., 2006). In previous studies, linalool and geraniol have been detected in Fenghuang Dancong tea (Li and Wang, 2021). Linalool and geraniol are important aroma compounds of Fenghuang Dancong tea. Linalool confers a floral and citrus aroma, and its odour threshold is very low, *i.e.*, 0.6 μg/L, making it easily captured by human olfactory sensors (Schuh and Schieberle, 2006). In the present work, the relative content of linalool was highest in fresh leaves of Fenghuang Dancong tea, and then decreased during the withering and rocking stages, with the content ratio as low as 2.58% in the baking stage (Figure 4). These results were similar to those reported elsewhere (Ma *et al.*, 2018), which showed that during production of the oolong tea "Huangdan" variety, the content of linalool also decreased with tea processing, reaching the lowest content in the baking stage. This may be related to the conversion of linalool into dehydrolinalool and other derivatives of linalool during production of Fenghuang Dancong tea. Dehydrolinalool confers a fragrance of flowers and plants, with a pungent and lavender-like fragrance, mainly derived from linalool glycosidic precursors (Yang *et al.*, 2013). During production of Fenghuang Dancong tea, dehydrolinalool is produced in the de-enzyming and baking stages. Nerolidol is produced by D-glucose *via* the cytosolic mevalonic acid (MVA) pathway, which occurs in two steps: initially, C5-based isopentenyl pyrophosphate (IPP) is produced by D-glucose, and then converted by IPP through a series of reactions to finally yield nerolidol (Ma *et al.*, 2014). Nerolidol confers a sweet neroli aroma, which was found in high relative content in Fenghuang Dancong tea, making it an important source of floral and fruity aroma in Fenghuang Dancong tea. In the present work, the relative content of nerolidol increased gradually during production of Fenghuang Dancong tea, from 3.97% in fresh leaves to 18.44% in the baking stage (Figure 4), which confirmed that nerolidol was produced throughout production of Fenghuang Dancong tea. This was corroborated by Ma *et al.* (2018); nerolidol in fresh oolong tea was found in the form of glycosides. During tea production, especially in the rocking stage, fresh tea leaves are damaged during smashing, which activates the endogenous glycosidase in cells, thus acting on glycosides, and releasing a large amount of nerolidol from the cells. Therefore, the content of nerolidol increased significantly during production of Fenghuang Dancong tea observed in the present work.

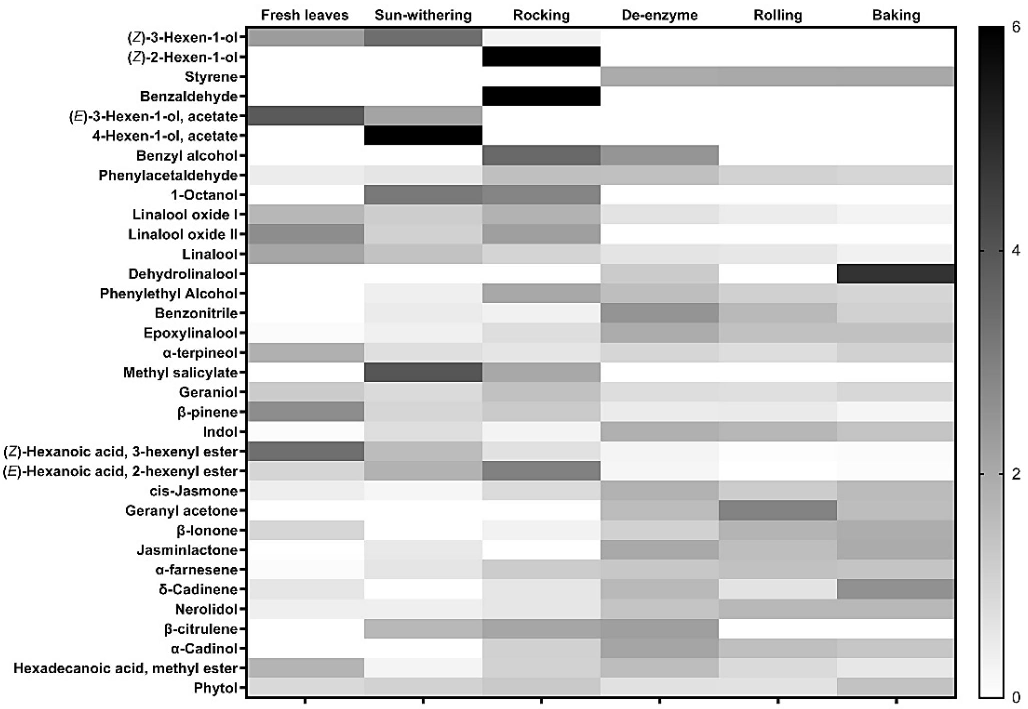

Figure 4. Heat map of relative contents of key volatile compounds during production of Fenghuang Dancong tea. Colour of boxes indicates value of $C/C_{average}$. C: average relative contents of volatile compound in every production stage; and $C_{average}$: average level of volatile compound in 24 samples.

Phytol confers a floral fragrance, and is an important constituent of the aroma of Fenghuang Dancong tea. During tea production, the relative content of phytol initially exhibited an increasing trend, then decreased, and then increased again. In fresh leaves, the relative content of phytol in Fenghuang Dancong tea was as low as 6.09%, and then increased during the withering and rocking stages, reaching 7.24 and 8.63%, respectively, before decreasing to 4.67% in the de-enzyming stage, and increasing to 9.79% in the baking stage (Figure 4).

Epoxylinalool has a strong, fresh, sweet, woody, and floral fragrance, conferring unique aroma to Fenghuang Dancong tea. In the fresh leaf stage, the relative content of epoxylinalool was very low (0.07%); as production progressed, the content of epoxylinalool gradually increased, peaking in the de-enzyming stage (1.77%) (Figure 4). Subsequently, epoxylinalool content decreased in the rolling and baking stages, although still higher than that in fresh leaves, which indicated that the processing of Fenghuang Dancong tea promoted epoxylinalool production.

During production of Fenghuang Dancong tea, the generation of floral and fruity aroma, such as nerolidol and phytol, confers a fragrant smell to the tea. Simultaneously, certain aroma compounds that confer a green grass flavour gradually dissipate during production, thus intensifying the floral and fruity aroma of Fenghuang Dancong tea. In particular, 3-hexene-1-ol confers a strong smell of freshly cut grass, and its content in fresh leaves and withering stages was very high (20.03 and 29.47%, respectively), while its relative content rapidly decreased to 2.54% in the rocking stage (Figure 4); 3-hexene-1-ol could not be detected in the de-enzyming, rolling, and baking stages, indicating that this aroma compound gradually dissipated throughout production stages, rendering the floral and fruity flavours more evident in Fenghuang Dancong tea, whereas the grass-like flavour was absent. A similar phenomenon occurred with 3-hexen-1-ol acetate, which also confers a strong smell of green grass. In the fresh leaf and withering stages of Fenghuang Dancong tea production, the relative content of 3-hexen-1-ol acetate was 2.08 and 1.15%, respectively, while in subsequent stages, 3-hexen-1-ol acetate was not detected, thus indicating that this aroma compound gradually dissipated throughout production of Fenghuang Dancong tea. Generally, most alcohol aroma compounds that confer floral and fruity aroma gradually increased throughout production of Fenghuang Dancong tea, thus endowing Fenghuang Dancong tea with strong floral and fruity aroma.

Phenylacetaldehyde is converted from phenylalanine (Zeng *et al*., 2017), and has a strong honey flavour, being considered an important aroma compound of Fenghuang Dancong tea. During production of Fenghuang Dancong tea, the content of phenylacetaldehyde initially increased and then decreased; in the fresh leaf stage, the relative content of phenylacetaldehyde was 1.44%, and then peaked in the rocking stage (5.03%), and then decreased, reaching 3.23% in the baking stage (Figure 4).

Indole is converted from tryptophan in tea leaves (Zeng *et al*., 2016). During production of Fenghuang Dancong tea, the relative content of indole was as low as 0.40% in fresh leaves, and continued to increase, peaking in the de-enzyming stage (13.40%), and then started to decrease, reaching 10.06% in the baking stage (Figure 4), which was similar to findings reported in a previous study (Zeng *et al*., 2016). These authors reported that indole content in oolong fresh tea was very low, and indole was mainly produced during tea production. In addition, production of indole in tea can be related to injury stress of tea leaves during processing. In this context, damage to tea leaves stimulates the expression of tryptophan synthesis β-subunits, which leads to the generation of a large amount of tryptophan β-subunits that are eventually converted into indole. At low contents of indole, a faint floral flavour prevails, whereas an increase in indole content is directly related to a large amount of tea aroma (Lin *et al*., 2013).

Benzonitrile has a distinctive almond odour, which is mainly generated by phenylalanine in tea plant cells *via* the phenylpropanoid pathway (Wang *et al*., 2020). Benzonitrile is also an important aroma component of Fenghuang Dancong tea. During production of Fenghuang Dancong tea, the relative content of benzonitrile initially increased, and then decreased. Interestingly, benzonitrile could not be detected in fresh tea leaves. In the withering and rocking stages, benzonitrile was gradually produced in tea, and its content peaked (7.15%) in the de-enzyming stage, and then decreased (3.09%) in the baking stage (Figure 4). These observations were similar to those reported elsewhere (Ma *et al*., 2018; Chen *et al*., 2020). A previous study reported that the content of benzonitrile in oolong "Huangdan" tea was low in fresh tea leaves and during the withering stage, but increased rapidly in the rocking and de-enzyming stages (Ma *et al*., 2018). In another study, the presence of benzonitrile was virtually undetectable in fresh leaves, and the content of benzonitrile gradually increased in withering and rocking stages (Chen *et al*., 2020). Therefore, benzonitrile is a special aroma compound produced during the processing of oolong tea, including Fenghuang Dancong tea.

Alkenes are important aroma compounds of Fenghuang Dancong tea, among which *α*-farnesene plays a major role which has the aroma of fresh petals and a faint apple scent. A large amount of *α*-farnesene is produced during production of Fenghuang Dancong tea. *α*-farnesene is generated in tea from pyruvic acid *via* the cytosolic MVA pathway (Yang *et al*., 2013). In the present work, the relative content of *α*-farnesene increased continuously during tea production (from 0.98% in fresh leaves to 14.29% in the baking stage), which was similar to the results of previous studies (Ma *et al*., 2018). In a previous study, tea cell vacuoles were disrupted during tea manufacturing due to collision, friction, and other factors in the rocking and rolling stages; as a result, polyphenols and the precursor of *α*-farnesene leaked from the vacuoles, which led to increased *α*-farnesene production under the action of cell-related enzymes (Suzuki *et al*., 2003; Wang *et al*., 2017). Thus, *α*-farnesene is released due to the fragmentation of tea cells in the rolling and other stages of tea production, resulting in higher relative content of *α*-farnesene in the baking stage. Styrene has a unique floral and sweet aroma, being rarely detected during production of other oolong teas. During production of Fenghuang Dancong tea, styrene was detected only in the de-enzyming, rolling, and baking stages, with contents reaching 3.66, 3.74, and 3.79%, respectively. This indicated that these processing stages stimulated production of styrene, and kept its content relatively stable after its generation, making it one of the characteristic aroma compounds of Fenghuang Dancong tea. In addition, δ-cadinene confers a sweet fruit flavour, and its relative content in fresh tea leaves was as low as 0.16%, increasing to 0.72% in the baking stage. Taken together, these findings indicated that production of Fenghuang Dancong tea promoted the formation of δ-cadinene.

Aroma formation in Fenghuang Dancong tea may originate mainly during production. In fresh tea leaves, the contents of 3-hexen-1-ol, 3-hexen-1-ol acetate, and (*z*)-3-hexenyl hexanoate, which have a green grass flavour were higher, and these compounds gradually dissipated during production until they became undetectable in baking, thus not contributing to the aroma of Fenghuang Dancong tea. Linalool, linalool oxide I, and linalool oxide II confer a strong sweet and floral fragrance; these compounds were found at high levels in fresh tea leaves, but their content gradually decreased during tea production, which may be related to their conversion to other derived compounds, such as oxidised linalool. Moreover, during tea production, the content of aroma compounds conferring a strong floral and sweet flavour is reduced, being transformed into compounds that confer a soft and mellow flavour, thus resulting in a more lasting and subtle fragrance in the baking stage. Finally, during production of Fenghuang Dancong tea, the relative contents of certain compounds with floral, fruity, and sweet fragrances, such as phenylethanol, nerolidol, β-ionone, phenylacetaldehyde, styrene, *α*-farnesene, indole, and benzonitrile, significantly increased. These aroma compounds confer to Fenghuang Dancong tea a unique floral and fruity flavour. Therefore, these aroma compounds can be used as markers of aroma quality during production of Fenghuang Dancong tea.

Key stages during production of Fenghuang Dancong tea

Rocking is an important step during production of Fenghuang Dancong tea. Tea leaves collide, allowing for the edges of the leaves to rub against each other through manual or mechanical shaking. The cell membrane on the surface of certain tea leaves becomes damaged, and the enzymes in the cytoplasm are exposed to oxygen, thus accelerating enzymatic oxidation, resulting in production of large number of aroma compounds (Zhou *et al*., 2017; Zeng *et al*., 2018). During production of Fenghuang Dancong tea, the types of aroma compounds increased from 64 types in the fresh leaf stage to 90 types in the withering stage, peaking at 104 types in the rocking stage (Table 1). Specifically in the rocking stage, the types of aroma compounds increased, and the relative content of certain important aroma compounds changed. For instance, the content of phenylethanol, phenylacetaldehyde, and *α*-farnesene increased significantly in the rocking stage compared with their content in the fresh leaf and withering stages, whereas the content of 3-hexen-1-ol, *trans*-3-hexenyl acetate, and (*z*)-3-hexenyl hexanoate, which confer a green grass aroma, decreased significantly in the rocking stage, indicating that the rocking stage plays an important role in the formation of floral and fruity aroma in Fenghuang Dancong tea.

De-enzyming is another important stage during production of Fenghuang Dancong tea. In this stage, tea leaves are fried in an iron pot at 220°C for approximately 3 - 5 min, which allows for the evaporation of certain aroma compounds in tea, whereas other aroma compounds will undergo oxidation, deamination, and decarboxylation at high temperatures, thus decomposed or converted into other compounds (Sheibani *et al*., 2016b; Wu *et al*., 2020). Therefore, in the present work, the types of aroma compounds in Fenghuang Dancong tea decreased from 104 in the rocking stage to 77 types of aroma compounds in the de-enzyming stage (Table 1). In particular, the relative content of alcohols decreased from 51.59% during rocking to 33.15% in the de-enzyming stage (Figure 1), which may be due to the low boiling point of certain alcohols, and evaporation is related to the conversion of alcohols (Cui *et al*., 2016; Chen *et al*., 2019). In the de-enzyming stage, the relative content of alkenes and other compounds increased significantly compared to that in the rocking stage, with alkenes increasing from 13.85 to 19.18%, and other compounds increasing from 4.63 to 23.11% (Figure 1). Thus, the de-enzyming stage could be considered important for aroma formation in Fenghuang Dancong tea. In addition, in the de-enzyming stage, the relative content of certain important aroma compounds changed significantly. For instance, the relative contents of nerolidol, styrene, δ-carbene, geranyl acetone, β-ionone, benzonitrile, and indole increased significantly, which is likely to play an important role in aroma formation in Fenghuang Dancong tea. Collectively, the formation and transformation of volatile aroma compounds during production of Fenghuang Dancong tea require further elucidation. In future studies, the mechanism of formation of characteristic aroma compounds during production of Fenghuang Dancong tea will be further explored.

Conclusion

In the present work, HS-SPME combined with GC-MS was used to extract, analyse, and identify volatile aroma compounds in Fenghuang Dancong tea across six production stages. The relative content of alcohols and esters exhibited a downward trend, while that of aldehydes, ketones, alkenes, and other aroma substances exhibited an upward trend. The types of aroma compounds in Fenghuang Dancong tea peaked during the rocking stage, indicating its significant role in aroma development in Fenghuang Dancong tea. In the de-enzyming stage, the relative content of key aroma compounds, including nerolidol, styrene, δ-carbene, geranyl acetone, indole, and benzonitrile increased significantly, likely contributing to the aromatic profile of Fenghuang Dancong tea. This exploratory analytical study will be further validated through sensory and OAV techniques.

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