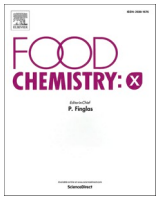
Food Chemistry: X 24 (2024) 101811

Contents lists available at ScienceDirect

Food Chemistry: X

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



UPLC-MS/MS and HS-SPME-GC–MS reveal the flavor profiles of two geographical indications woody vegetables: *Staphylea bumalda* and *Staphylea holocarpa*

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ARTICLE INFO

Keywords:
Staphylea bumalda
Staphylea holocarpa
Geographical indications
Nutrition
Flavor

ABSTRACT

Staphylea bumalda (SHC) and *Staphylea holocarpa* (PGG) were recognized as geographical indication agricultural products due to unique flavor. 1218 differential non-volatile compounds and 536 differential volatile compounds were detected and identified through UPLC-MS/MS and HS-SPME-GC–MS methods. In SHC samples, catechins, epicatechins, proanthocyanidins, quinic acid derivatives, and kaempferol glycoside derivatives were the main flavor compounds, with bitter and harsh taste. *L*-tartaric acid, citraconic acid and citric acid were contributed to increase acidity. 4-Hexen-1-ol acetate, butanoic acid butyl ester, 3-Hexen-1-ol acetate, (*E*)-, and 3-Hexen-1-ol acetate, (*Z*)- were identified as characteristic odor compounds with strong floral, fruity and sweet odor. In PGG samples, epicatechin gallate, quercetin glycoside derivatives, L-histidine, and L-tyrosine were the leading contributors to bitter and harsh taste. The spicy, herbal, and bad smell odor were mainly brought by 2-octanol, and 3-Octen-1-ol, (*Z*)-. Our results offered comprehensive insights into the flavor and quality characteristics differences between PGG and SHC.

1. Introduction

Staphylea bumalda (SHC) and *Staphylea holocarpa* (PGG), belonging to the genus Staphylea in the family Staphyleaceae, are deciduous shrubs or small trees (Yao et al., 2022). Its tender leaves and flower buds are high-quality edible woody vegetables with delicious taste and richer nutrients such as proteins, amino acids, dietary fibers, vitamins and minerals, which have higher health care and nutritional values (Sircelj et al., 2019).

The flavor (taste and odor) of SHC and PGG was derived from the non-volatile compounds and volatile compounds (Aisala et al., 2020; Zhang, Cui, et al., 2023). The taste, nutritional value and biological activity of SHC and PGG were mainly dominated by non-volatile compounds. The black tea taste compounds of leaves were attributed to amino acids, phenolic acids and flavonoids (Chen et al., 2024). The edible and medicinal values of SHC and PGG were attributed to the high content of crude fat, essential amino acids and vitamins in tender leaves, shoots and buds (Sircelj et al., 2019). 18 kinds of amino acids, containing 8 kinds of human essential amino acids, were detected in tender leaves and flower buds, which were comparable to those of *Moringa oleifera* leaves (19 kinds) (Agba et al., 2024). Aspartic acid and glutamic acid, 2 umami amino acids, contained higher proportion, which might be one of the reasons why flower buds and tender leaves were delicious as vegetables. Phenolic acids and flavonoids with higher content were isolated from the wild SHC flower buds and tender leaves (Novotny et al., 2003). The total phenolic contents of SHC leaves extracted through ethyl acetate was greatly higher than that extracted by chloroform (Lacikova et al., 2006). The quality of aroma characteristics was determined by volatile compounds (Feng et al., 2019). The aroma profiles were controlled by the key aroma substances, such as esters

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https://doi.org/10.1016/j.fochx.2024.101811
Received 20 July 2024; Received in revised form 19 August 2024; Accepted 1 September 2024
Available online 2 September 2024

(butanoic acid butyl ester, methyl salicylate, methyl jasmonate, etc.), ketones (1-octen-3-one, β-ionone, etc.), aldehydes (decanal, hexanal, nonanal, etc.), alcohols (linalool and its oxides, geraniol, and terpenoids (germacrene D, β-bourbonene, etc.) (Xie et al., 2023). Therefore, the characteristics of the key taste and odor compounds in SHC and PGG are particularly relevant. However, the current domestic and foreign researches on SHC and PGG mostly focuses on the medicinal value, practical value and promotion of its seeds, flowers and leaves. There is a lack of research on the characteristics of metabolites and the differences between nutrition and flavor of SHC and PGG.

In this study, SHC and PGG were selected as experimental materials to characterize flavor profile, and the taste and odor compounds were detected and identified through ultra performance tandem mass spectrometry (UPLC-MS/MS) and head-space solid-phase-micro-extraction gas-chromatography-mass-spectrometer (HS-SPME-GC–MS). Multi-statistical analysis was applied to reveal the differences between volatile compounds and non-volatile compounds of SHC and PGG, and the correlation between them was discussed. Our aim was to comprehensively analyze the volatile compounds and non-volatile compounds of SHC and PGG, and to investigate the differences in the quality of tender leaves and the key compounds of flavor quality formation. The results provided theoretical and practical basis for flavor research, variety identification, quality evaluation and production application of SHC and PGG.

2. Materials and methods

2.1. Leaves of PGG and SHC samples

'Zhenba SHC', a specialty of Zhenba County, Shaanxi Province, is a national geographical indication of agricultural products. In mid-April 2024, the tender leaves of SHC and PGG were collected in Houping Forest Farm (Zhenba County, Shaanxi province, China). The samples obtained in this experiment were all tender leaves at the top of the branches in the state of florescence, without pests and diseases (Fig. S1). 20 tender leaves were collected from 4 directions on 5 trees with consistent growth. The tender leaves were rapidly embedded in liquid nitrogen and then stored in refrigerator at −80 °C.

2.2. Volatile compounds determination

2.2.1. Sample preparation and treatment

The tender leaves were ground into powder in liquid nitrogen, and 0.50 g powder was put into the 20 mL head-space sample vial including NaCl saturated solution. The vials were sealed using crimp-top caps with TFE-silicone headspace septa (Agilent). At the time of SPME analysis, each vial was placed in 60 °C for 5 min, then a 120 μm DVB/CWR/PDMS fiber (Agilent) was exposed to the headspace of the sample for 15 min at 60 °C, and then desorbed at 250 °C for 5 min (Wen et al., 2023).

2.2.2. HS-SPME-GC–MS analysis

Agilent Model 8890 GC and a 7000E mass spectrometer (Agilent) equipped with a DB-5MS capillary column (30 m × 0.25 mm × 0.25 um) were utilized to detect the volatile compounds, with using high purity helium (≥99.999 %) as carrier gas of the linear flow rate 1.2 mL/min at 250 °C. The temperature programs were as follows: 40 °C at 3.50 min, increasing at 10.0 °C/min to 100 °C, at 7.0 °C/min to 180 °C, at 25.0 °C/min to 280 °C, hold for 5.0 min.

The mass spectrometry was recorded under the 70 eV electron impact (EI) ionisation mode, and the temperatures of quadrupole mass spectrometer, ion source and transfer line were 150 °C, 230 °C and 280 °C, respectively. Selection ion monitoring mode was applied to qualitatively and quantitatively analyze the MS data.

Based on the NIST 2011 mass spectrometry database, the chemical structure and name of the volatiles represented by each peak were confirmed according to the retention index, actual composition and characteristic ions. Peak area normalization method was applied to determine the relative content of each component after detection by GC–MS.

2.3. Non-volatile compounds determination

2.3.1. Samples extraction

Grinder (MM 400, Retsch) was utilized to ground tender leaves into powder. 0.05 g powder was transferred into 1.20 mL of −20 °C pre-cooled 70 % methanol extract. The extract was vortexed once 30.0 s every 30.0 min with 6 times, and then stored at 4 °C. After centrifugation (12,000 rpm, 3 min), the supernatant was filtered through a 0.22 μm microporous membrane, which was stored in the injection bottle for non-volatile compounds determination via UPLC-MS/MS system (Sun et al., 2023).

2.3.2. UPLC-MS/MS analysis

4.0 μL extract above was injected into the Agilent SB-C18 column (1.8 μm, 2.1 mm × 100 mm, ExionLC™ AD) equipped with solvent A (ultrapure water with 0.10 % formic acid) and solvent B (Acetonitrile with 0.10 % formic acid), with the flow rate of 0.35 mL/min and the column oven at 40 °C. The 14.0 min linear gradient was programmed at 5.0 % B at 0 min; Within 9 min, a linear gradient to 5 % A, 95 % B was programmed, and a composition of 5 % A, 95 % B was kept for 1 min. Subsequently, a composition of 95 % A, 5.0 % B was adjusted within 1.1 min and kept for 2.9 min. The effluent was alternatively connected to an ESI-triple quadrupole-linear ion trap (QTRAP)-MS.

The ESI source parameters of the (QTRAP)-MS were as follows: temperature of positive ion (5500 V) and negative ion mode (−4500 V) was set at 550 °C, and ion sources of gas 1, gas 2, and curtain gas were 50, 60, and 25 psi, respectively; and the collision-activated dissociation was high. The collision gas (nitrogen) of QQQ scans was set to medium. DP and CE were optimized for individual MRM transitions. A specific set of MRM transitions were monitored for each period according to the metabolites eluted within this period.

2.4. Data analysis

Analyst 1.6.3 software was applied to process the UPLC-MS/MS data. The MultiaQuant™ software was used to integrate and correct the chromatographic peaks. MassHunter software was utilized to process the GC–MS/MS data for qualitative and quantitative analysis, and to integrate and correct the chromatographic peaks. The peak area of each chromatographic peak represented the relative content of the corresponding metabolite. The peak areas of non-volatile compounds and volatile compounds identified were introduced into Metware Cloud platform for principal component analysis (PCA) and orthogonal partial least squares discriminant analysis (OPLS-DA) to select the differential compounds. The metabolites with VIP $\geq$ 1, Fold Change $\geq$2 or $\leq$ 0.5, and p-value<0.05 were identified as differential compounds. Pie chart plot, heatmap plot, plot color mapped scatter, and bubble plot were completed through Origin Pro 2024b software.

3. Results

3.1. Characteristics analysis of non-volatile compounds of SHC and PGG samples

Nutrients are the key factors to determine the quality of SHC and PGG samples, and flavor is a comprehensive expression of the taste characteristics of nutrients under specific conditions. UPLC-MS/MS was utilized to investigate the flavor profiles in PGG and SHC samples, and the total ion current curves of the positive ion mode (P+) (Fig. S2A) and negative ion mode (N-) (Fig. S2B) were highly coincident. The obvious separation trends of SHC and PGG samples were observed in PCA plot (Fig. 1A), indicating that the metabolites accumulation in SHC and PGG

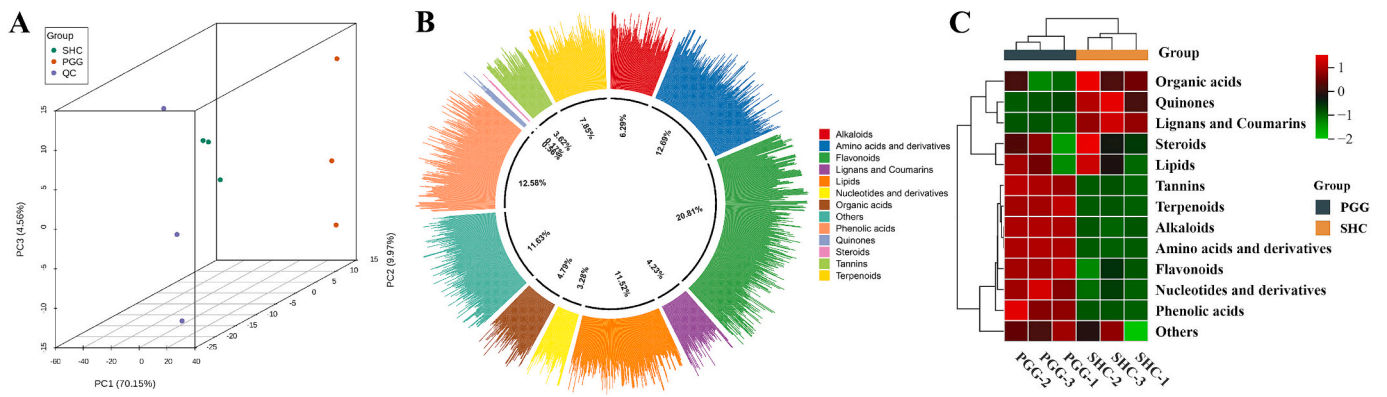

Fig. 1. Non-volatile compounds in SHC and PGG. A: 3D-PCA score plot. B: Pie chart of the 13 categories of 1797 non-volatile compounds. C: Heat map of the non-volatile compounds content in different categories.

samples was significantly different. 1797 non-volatile compounds in 13 categories were identified (Fig. 1B and Table S1), including 374 flavonoids, 226 phenolic acids, 228 amino acids and their derivatives, 207 lipids, 141 terpenoids, 113 alkaloids, 86 organic acids, 76 lignans and coumarins, 65 tannins, 59 nucleotides and their derivatives, 10 quinones, 3 steroids and other compounds (saccharides, vitamins and aldehyde compounds). The relative content of 13 categories was drawn to more intuitively understand the variations (Fig. 1C). The results elucidated that the relative contents of flavonoids, phenolic acids, amino acids, derivatives, terpenoids, alkaloids, tannins, and nucleotides and derivatives in PGG samples were higher, while the relative contents of lignans and coumarins, organic acids, quinones and other substances in SHC samples were higher. The differences in the types and contents of non-volatile compounds between PGG and SHC samples formed different taste characteristics.

3.1.1. *Identification of differential non-volatile compounds of SHC and PGG samples*

In our study, OPLS-DA was applied to investigate the differences between PGG and SHC samples. PGG and SHC samples were obviously divided into 2 groups in the OPLS-DA score plot, illustrating that there existed significant differences between PGG and SHC samples (Fig. 2A). The R^2X (0.821) and R^2Y (1) values of OPLS-DA model were greater than 0.5, and the Q^2 values (0.996) was greater than 0.90, indicating that the model was reliable, and which could be applied to further determination of differential non-volatile compounds (Fig. 2B). Permutation test (n = 200) was carried out to verify the OPLS-DA model to avoid model with overfitting (Fig. 2C). 1218 differential non-volatile compounds were achieved between PGG and SHC samples, among which 739 compounds were up-regulated, and 479 compounds were down-regulated (Fig. 2D). Among the differential non-volatile compounds, flavonoids (322) were the most abundant, followed by phenolic acids (231) and amino acids and derivatives (114), accounting for 26.44 %, 18.96 %, and 9.35 %,

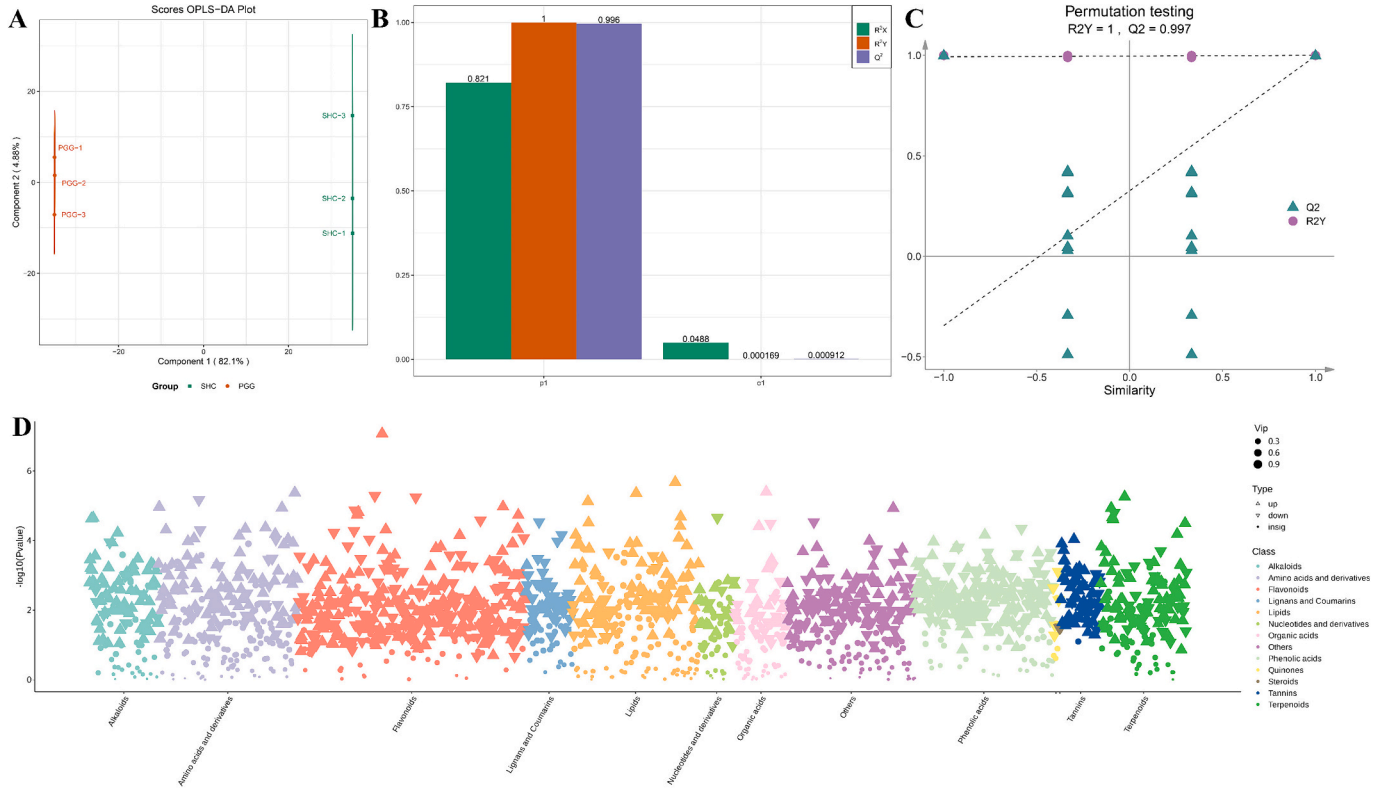

Fig. 2. A: Scores OPLS-DA plot. B: OPLS-DA model. C: OPLS-DA permutation. D: Categorical statistical scatter plot of differential non-volatile compounds.

respectively, indicating that the compounds in those 3 categories were the main compounds affecting the flavor of PGG and SHC samples.

3.1.2. Differential flavonoids in PGG and SHC samples

Flavonoids, as the widely distributed secondary metabolites in plants with the potential physiological functions such as anti-oxidation, anti-aging, anti-cancer and balancing blood sugar, are the important compounds influencing the flavor of PGG and SHC samples. Among the 322 differential flavonoids, 174 flavonoids were markedly up-regulated in PGG samples, and 148 flavonoids were evidently up-regulated in SHC samples, which were mainly enriched in flavonols and flavones (Table S2). The aglycones in the chemical structure of flavonol glycosides would affect the harsh taste (Scharbert & Hofmann, 2005). The differential flavonoids with higher content were mainly quercetins in PGG, whereas the differential flavonoids in SHC were kaempferols and luteolins (Fig. 3A). Quercetin-3-O-sambubioside, quercetin-3-O-(6"-O-arabinosyl) glucoside, quercetin-3-O-apiosyl(1 → 2)galactoside, and quercetin 3-O-glucoside 7-O-xyloside exhibited higher expression, with the fold change values of 6.90E+02, 6.65E+02, 5.90E+02, and 1.13E+03, respectively. Among the differential kaempferols and luteolins, kaempferol-4'-O-glucoside and luteolin-7-O-glucoside displayed highest content in SHC samples, followed by kaempferol-3-O-glucorhamnoside, luteolin-7-O-neohesperidoside, kaempferol-3-O-robinobioside, and luteolin-7-O-rutinoside. Catechins are a class of compounds with dual taste attributes (bitterness and harsh taste).

In PGG samples, gallocatechin, gallocatechin gallate, catechin-(4alpha → 8)-gallocatechin, and epigallocatechin-3-O-gallate were 2.93E+02-, 9.45E+00-, 3.85E+00-, and 9.62E+00- fold of those in SHC samples, whereas epicatechin, catechin, epicatechin-4'-O-β-D-glucopyranoside, epicatechin 3-glucoside, catechin 4'-O-beta-D-glucopyranoside, and epicatechin-3'-O-β-D-glucopyranoside were higher in SHC, which were contributed to bitterness and harsh taste in PGG and SHC samples (Table S2).

3.1.3. Differential phenolic acids in PGG and SHC samples

Phenolic acids are another kind of compounds exhibiting bitterness and harsh taste, mainly including benzoic acid and hydroxycinnamoyl compounds and their derivatives (Zhang et al., 2020). 231 differential phenolic acids were obtained, among which 131 compounds were up-regulated in PGG samples, and 80 compounds were up-regulated in SHC (Table S3). In SHC samples, 1-O-p-Coumaroylquinic acid, 3-O-p-Coumaroylquinic acid, 3-(Hydroxycinnamoyl)-quinic acid, 5-O-p-Coumaroylquinic acid, 4-O-Coumaroylquinic acid, chlorogenic acid (3-O-Caffeoylquinic acid), neochlorogenic acid (5-O-Caffeoylquinic acid) and cryptochlorogenic acid (4-O-Caffeoylquinic acid) were significantly higher than those in PGG samples (Fig. 3B). In PGG samples, the mainly differential phenolic acids were feruloylmalic acid, caffeic acid, isoferulic acid, gallic acid and gallic glycosides (6-O-Galloyl-β-D-glucose, 5-O-Galloyl-D-hamamelose, 3-O-Galloyl-D-glucose, 2,3-Di-O-Galloyl-β-D-Glucose, Methyl gallate, 1,6-Di-O-Galloyl-D-Glucose, and Digalloylglucose).

Hence, gallic acid, chlorogenic acid, caffeoylquinic acid and p-coumaroylquinic acids were the key differential phenolic acids that had great contribution to harsh taste, especially 4-O-p-coumaroylquinic

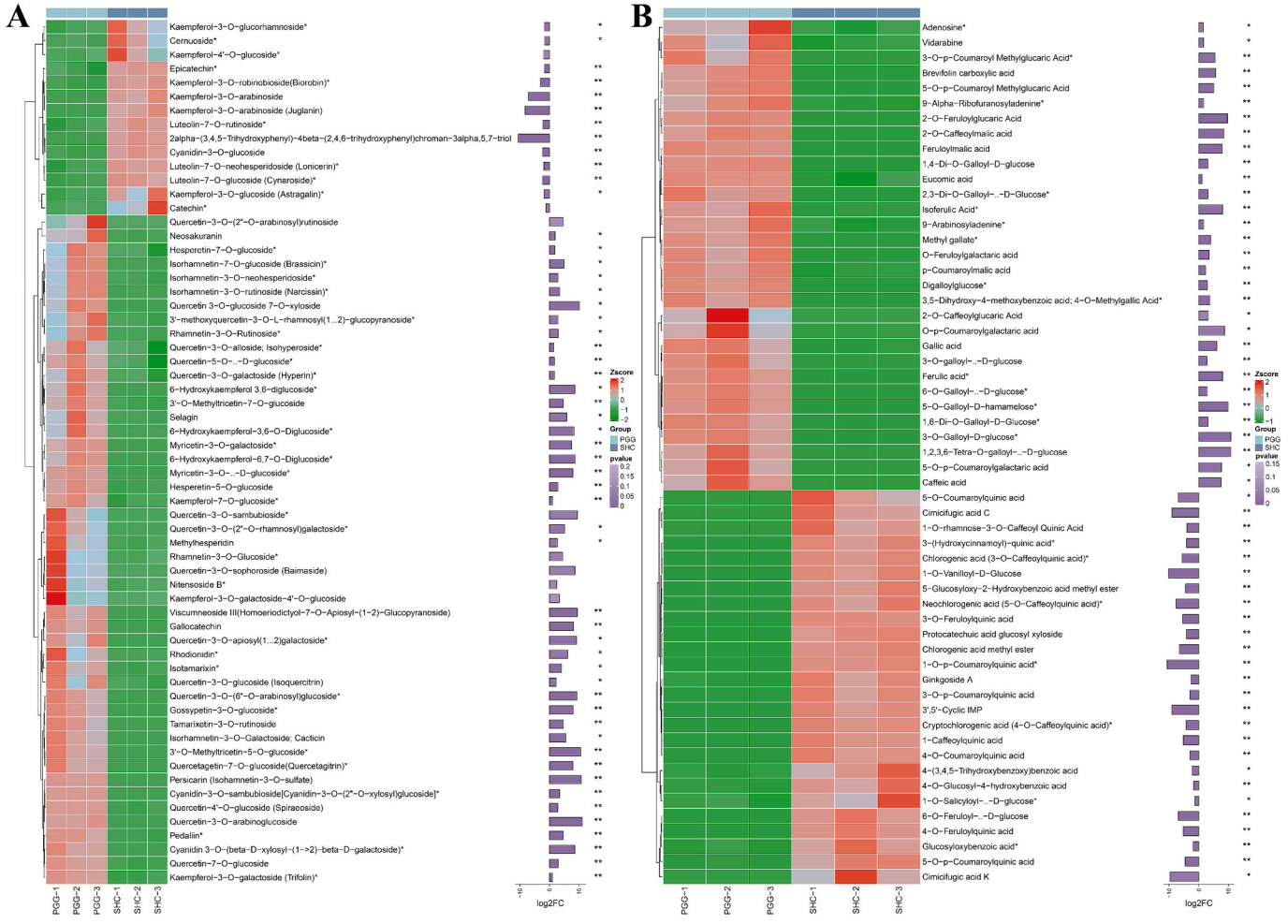

Fig. 3. A: Differential flavonoids with higher content in PGG and SHC. B: Differential phenolic acids with higher content in PGG and SHC.

acid.

3.1.4. Differential amino acids and derivatives in PGG and SHC samples

Amino acids are the basic unit of protein, and its type, content and composition determine the protein nutritional values (Kaneko et al., 2006). 228 amino acids and their derivatives were identified, containing 8 kinds of essential amino acids. And, 114 differential amino acids and their derivatives were achieved between PGG samples and SHC samples (Table S4). Cycloleucine, L-arginine, N(6),N(6)-Dimethyl-L-lysine, pyroglutamic acid, and DL-tryptophan exhibited higher content in PGG samples (Fig. S3A). Moreover, L-aspartic acid, and Ile-Asp with umami taste were also significantly enhanced at PGG samples, L-tyrosine and L-histidine with bitter taste were significantly accumulated in PGG than those in SHC samples. L-glutamic acid, 4-hydroxy-L-glutamic acid, and L-homonorleucine were 2.04E+00-, 1.35E+01-, and 3.11E+02- fold higher in SHC samples than that in PGG samples. In addition, the contents of His-Ala-Gln, Ser-His-Gln, and N-Acetyl-L-glutamine were higher in SHC samples, which were the characteristic umami taste compounds, which had been applied to regulate food flavor.

3.1.5. Differential alkaloids and organic acids in PGG and SHC samples

Alkaloids are the main single bitter compounds in PGG and SHC leaves, and 77 differential alkaloids were obtained (Table S5). The dominant differential alkaloids in the fresh leaves of PGG samples were L-Pipecolic Acid, 3-Indoleacrylic acid, Pyran[3,4-*b*]indole-2-ketone, naphthisoxazol A, Pipecolic acid, 3-amino-2-naphthoic acid, 3-hydroxy-1-methylpyrrolidin-2-one, L-Proline, and trigonelline, and those compounds were present with a certain bitter and harsh taste (Fig. S3B). The alkaloids compounds that caused bitter differences in SHC samples were 10-Hydroxymethyllycaconitine, N-[4-Hydroxycinnamoyl]-L-glutamate, neolinustatin, oxypinnatanine, spermidine, 3-quinolinecarboxylic acid.

6-aminocaproic acid, methyl brevifolincarboxylate, 1-methylpiperidine-2-carboxylic acid, 5′-glucosyloxyjasmanic acid, tuberonic acid glucoside, and *L*-tartaric acid displayed higher content in PGG samples (Fig. S3C). Citric acid, citraconic acid, citric acid diglucoside, 2-methyl-3-oxoadipic acid, and DMAPP exhibited higher content in SHC samples, which were contribute to sour taste.

3.1.6. Differential tannins and terpenoids in PGG and SHC samples

64 differential tannins (16 proanthocyanidins and 48 tannins) were identified between PGG and SHC samples (Table S5), among which proanthocyanidins were dimer or polymer compounds with catechin or epicatechin as a precursor, connected by the C4-C8 or C4-C6 bond of flavan-3-ol, and also bitter and harsh taste compounds (Ou & Gu, 2014). Among the differential proanthocyanidins, procyanidin B7, procyanidin B6, procyanidin B3, procyanidin B2, procyanidin B1, procyanidin B4, procyanidin B5, procyanidin C2, procyanidin B8, and procyanidin C1 displayed higher expression in SHC samples, whereas gallocatechin-(4alpha- > 8)-epigallocatechin, theasinensin E, theasinensin C, and epigallocatechin-(4beta- > 8)-catechin exhibited higher expression in PGG samples, which indicated that the bitter and harsh taste of SHC samples were greater than PGG samples. Moreover, most differential tannins were significantly enhanced at PGGs, including 3'-*O*-methyl-ellagic acid-4-O-beta-D-xylopyranoside, 3-*O*-methylellagic acid 4-sulfate, 4,6-(*S*)-Hexahydroxydiphenoyl-β-D-glucose, 3-*O*-methylellagic acid 4′-sulfate, strictinin, corilagin, isocorilagin, sanguiin H4, and isostrictinin.

98 differential terpenoids were identified, which were mainly composed of triterpenes, monoterpenoids, and sesquiterpenoids (Table S6), and 64 kinds of terpenoids were significantly greater in PGG samples than that in SHC samples. Among them, the differential terpenoids in PGG samples were mainly triterpenes and sesquiterpenoids, including cordianal C, maslinic acid, corosolic acid, 2-hydroxyoleanolic acid, alphitolic acid, 3-epiursolic acid, betulinic acid, and ursolic acid. 34 differential terpenoids in SHC samples were significantly greater than those in PGG samples. The content of (8*R*)-9-hydroxy-8-(hydroxymethyl)-6-methoxy-8-methylpyrano[2,3-*f*]chromen-2-one, pomolic acid, rubianol-f, and santalol A in SHC samples were 2.40-, 2.60-, 2.53-, and 43.42-fold higher than those in PGG samples, respectively (Fig. S3D).

3.2. Characteristics analysis of volatile compounds of SHC and PGG samples

3.2.1. Volatile compounds analysis in PGG and SHC samples

To investigate the aroma characteristics of PGG and SHC samples, the volatile compounds were identified and analyzed by HS-SPME-GC–MS, and the total ion current diagram was described in Fig. S4. As described in Fig. 4A, 1186 volatile compounds were detected, including 105 alcohols, 78 aldehydes, 137 ketones, 205 esters, 43 aromatics, 58 hydrocarbons, 256 terpenoids, 113 heterocyclic compounds, 40 acids, 29 ethers, 13 halogenated hydrocarbons, 4 sulfur compounds, 17 nitrogen compounds, 49 phenols and 39 amines (Table S7). The content of terpenoids in the volatile components of the two varieties was the highest, followed by esters and ketones, accounting for 21.59 %, 17.28 % and 11.55 % of the volatile components, respectively. The results of volatile compounds content in different categories illustrated that esters, heterocyclic compounds, ketones, hydrocarbons, alcohol, amine in SHC samples were higher than those in PGG samples, whereas aromatics, sulfur compounds, halogenated hydrocarbons, and terpenoids were abundant in PGG samples (Fig. 4B).

3.2.2. Identification of differential volatile compounds of SHC and PGG samples

In the PCA and OPLS-DA plots, it was clearly observed that PGG and SHC samples were obviously separated into 2 groups. Based on the VIP values and fold change, 536 differential volatile compounds were determined between PGG and SHC, accounting for 45.19 % of the total number of volatile compounds (Table S8). Among the differential volatile compounds, the number of terpenoids was the largest, with 119 kinds, accounting for 22.20 %, followed by esters (90 kinds), ketone (61 kinds), heterocyclic compounds (53 kinds), alcohols (47 kinds), aldehydes (31 kinds). The compounds of those 6 categories, a total of 401 kinds, accounted for 74.81 % of the total differential compounds (Fig. 4C). 315 up-regulated compounds in SHC samples were much greater than 221 down-regulated compounds (Fig. 4D).

The changes of volatile compounds of those 6 categories were visualized by heat map and it could be found that most differential volatile compounds were significantly up-regulated in SHC samples, demonstrating that the aroma of SHC samples was more abundant than that of PGG samples (Fig. 5). Among the differential terpenoids, there were 7 substances with higher content in SHC samples presenting floral and woody odor, including trans-.beta.-ionone, 3-buten-2-one, 4-(2,6,6-trimethyl-1-cyclohexen-1-yl)-, 2,6-Octadien-1-ol, 3,7-dimethyl-, acetate, (*Z*)-, geranyl acetate, 1-penten-3-one, 1-(2,6,6-trimethyl-2-cyclohexen-1-yl)-, ionone, .alpha.-irone. 2,3-Dehydro-1,8-cineole, (+)-epi-Bicyclosesquiphellandrene, gamma.-muurolene, germacrene D, alpha.-muurolene, beta.-guaiene, bicyclosesquiphellandrene, and caryophyllene were mainly increased in PGG samples compared to those of SHC samples, presenting with herbal, woody, and spicy odor (Fig. 5A). Specifically, it was found that the esters compounds with fruity odor were slightly higher in SHC samples compared to PGG samples (Fig. 5B). 2-Furanmethanol acetate, 4-hexen-1-ol acetate, butanoic acid butyl ester, 3-hexen-1-ol acetate, (*E*)-, 3-hexen-1-ol, acetate, (Z)-, and butanoic acid phenylmethyl ester in SHC samples were significantly up-regulated by 3.58E+03-, 8.66E+00-, 7.09E+00-, 7.09E+00- and 1.22E+01- fold, respectively. Among the differential heterocyclic compounds, pyrazine, trimethyl-, 1,2,3,6-tetrahydropyridine, pyridine, 2,3,4,5-tetrahydro-, thiazole, 4-propyl-, 1,3-Dioxolane, 2-ethyl-4-methyl-, and 5,6,7,8-tetrahydroquinoxaline in SHC samples were significantly higher than those of PGG samples, but those 6 compounds did not have aroma properties (Fig. 5C). Furan, 2-hexyl-, pyrazine,

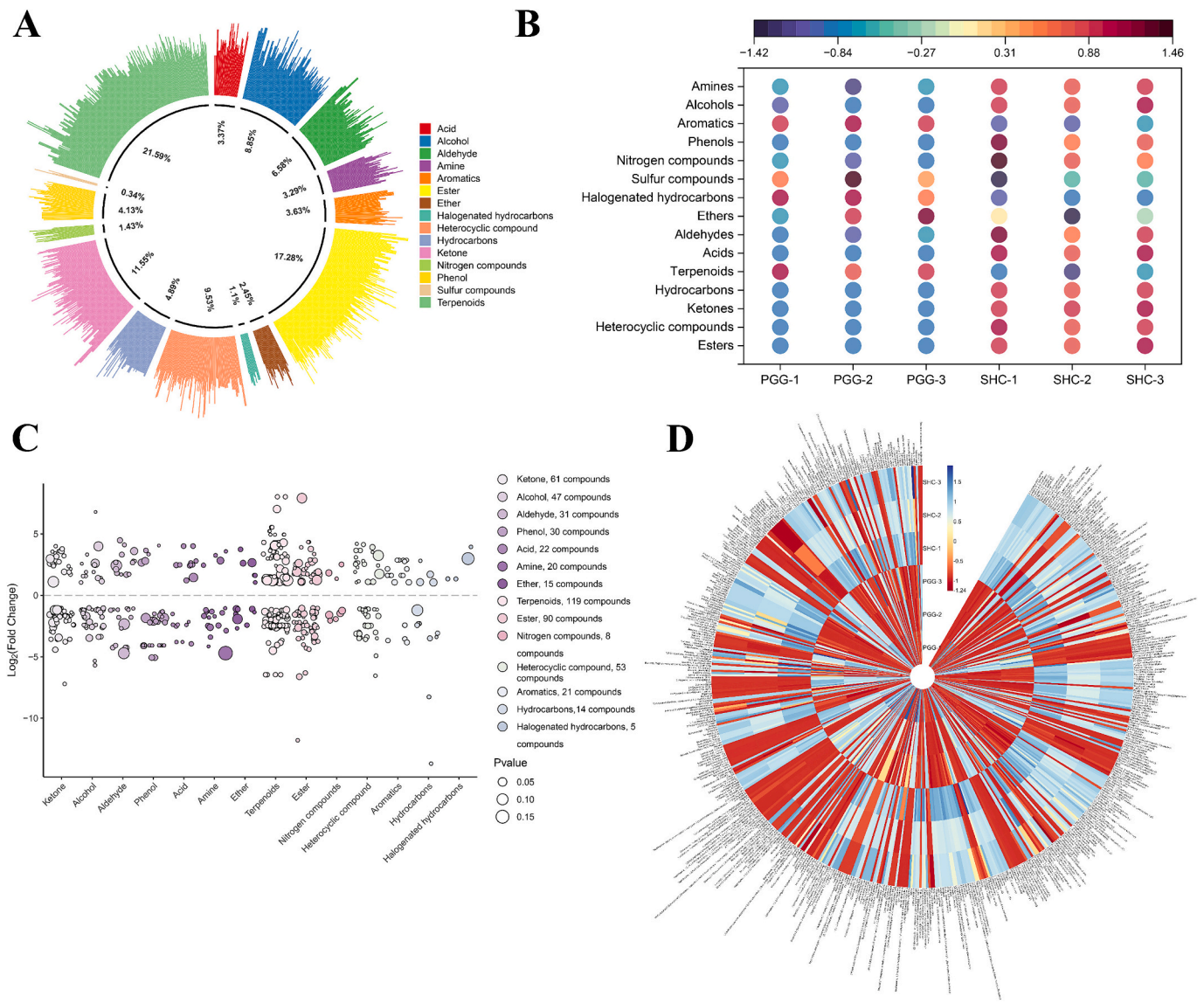

Fig. 4. A: Pie chart of the 1186 volatile compounds in different categories. B: Plot color mapped scatter of volatile compounds content in different categories. C: Bubble plot of differential volatile compounds in different categories. D: Heat map of 536 differential volatile compounds content.

ethoxy-, Pyrazine, 2-methoxy-3-(1-methylpropyl)-, pyrazine, 2-methoxy-3-(2-methylpropyl)-, and pyrazine, 2-methoxy-3-(1-methylethyl)- were obviously more abundant in PGG samples than those in SHC samples, which were contributed to green bell pepper, pea, and galbanum odor. Differential ketones contributing to sweet and fruity odor were significantly higher in SHC samples than in PGG samples, mainly including ethanone, 1-(2-methylphenyl)-, 2-buten-1-one, 1-(2,6,6-trimethyl-1,3-cyclohexadien-1-yl)-, 2-propanone, 1-(4-methoxyphenyl)-, 2-butanone, 4-(2,6,6-trimethyl-2-cyclohexen-1-yl)-, 3(2H)-furanone, dihydro-2-methyl-, and 4-acetylanisole (Fig. 5D). 4-methyl-5-nonanone, 2′-ethoxyacetophenone, 1,3-cyclohexanedione, 5,5-dimethyl-, 3-buten-2-one, 4-phenyl-, elsholtzia ketone, and acetophenone, 4′-amino- with none aroma characteristics were obviously higher PGG samples than those in SHC samples. The obvious increase of hexanal, butanal, 3,3-dimethyl-2-oxo-, hemihydrate, benzaldehyde, 4-ethyl-, benzaldehyde, 3-ethyl-, benzaldehyde, 2-ethyl-, 5-hexenal, 3-hexenal, (*Z*)-, and 3-hexenal in SHC samples, led to a stronger sweet, green, and apple odor compared to PGG samples (Fig. 5E). Nonanal greatly contributed to citrus and orange peel odor in PGG samples, with 3.25E+00- fold than that in SHC samples. 2,4-Decadienal, (E, *E*)- and 2,4-Decadienal, (E, Z)- were higher in PGG samples compared to SHC samples, contributing to waxy odor. 2,4-Heptadien-1-ol, (E, E)-, 2-furanmethanethiol, 5-methyl-, 3-hexanol, 2-hexen-1-ol, (E)-, 2-hexen-1-ol, (Z)- and 1-butanol, 2-ethyl- were remarkably higher in SHC samples, contributing to sweet, fresh, and fruity odor (Fig. 5F). There were 6 differential alcohols presenting spicy odor and combinations of herbal, woody, and mouldy odor, and the main contributing compounds included 2-Octanol, (*S*)- with spicy, oily, and fatty odor, 2-Octanol with spicy, woody, and herbal odor, 1,5,7-Octatrien-3-ol, 3,7-dimethyl- with mouldy odor. Hence, the comprehensive expression of these differential volatile compounds was contributed to the aroma difference between PGG and SHC samples.

3.2.3. *Identification of key aroma compounds in PGG and SHC samples*

The aroma characteristics are not affected by all volatile compounds, and only a few of them play a major role in sensory contribution. OAV values were commonly applied to evaluate the contributions of each volatile compound, among which the volatile compound with OAV > 1 was usually thought to be the important contributors to aroma. 117 volatile compounds (OAV > 1) were screened out in SHC samples and 113 in PGG samples, among which 98 volatile compounds were in common (Table S9).

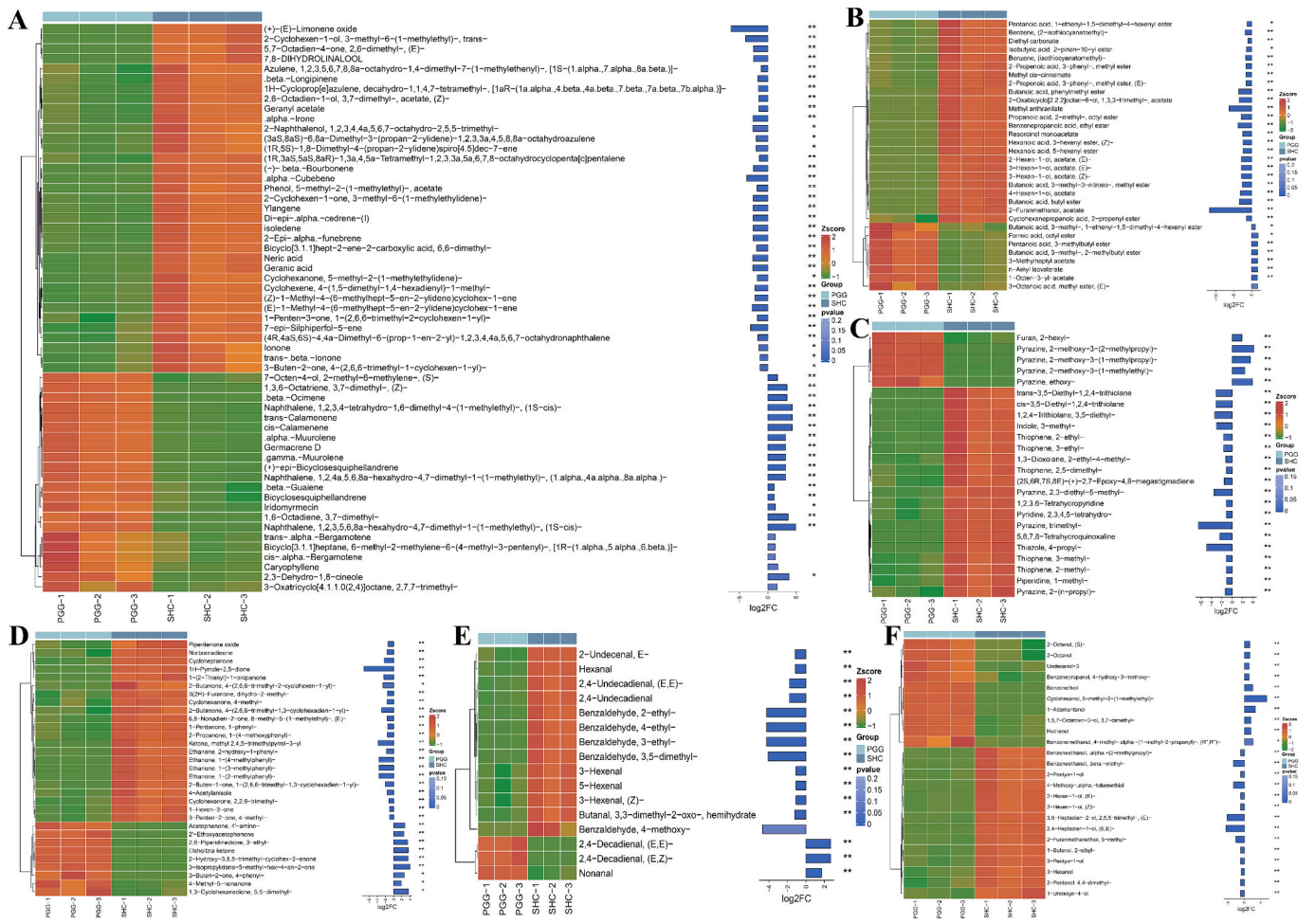

Fig. 5. A: Differential terpenoids with higher content in PGG and SHC. B: Differential esters with higher content in PGG and SHC. C: Differential heterocyclic compounds with higher content in PGG and SHC. D: Differential ketones with higher content in PGG and SHC. E: Differential aldehydes with higher content in PGG and SHC. F: Differential alcohols with higher content in PGG and SHC.

Pyrazine, trimethyl- (OAVs, 1.64E+01), 4-hexen-1-ol acetate (OAVs, 2.11E+01), butanoic acid butyl ester (OAVs, 4.60E+01), 3-Hexen-1-ol acetate, (*E*)- (OAVs, 1.44E+00), 3-Hexen-1-ol acetate, (*Z*)- (OAVs, 4.05E+01), 2-furanmethanethiol, 5-methyl-(OAVs, 1.44E+04), butanoic acid, phenylmethyl ester (OAVs, 1.56E+00), trans-.beta.-ionone (OAVs, 2.30E+03), 3-buten-2-one, 4-(2,6,6-trimethyl-1-cyclohexen-1-yl)- (OAVs, 6.57E+04), methyl anthranilate (OAVs, 3.05E+01), and benzaldehyde, 4-methoxy- (OAVs, 1.51E+01) were more abundant in SHC samples than that in PGG samples. 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-Hexen-1-ol, acetate, (E)-, and 3-Hexen-1-ol, acetate, (*Z*)- were critical contributors to aroma characteristics due to their lower odor thresholds and higher r-OAVs values, which led to the stronger sweet, floral, and fruity odor.

The distinct different volatile compounds with higher abundance in PGG samples were nonanal, pentanoic acid, 3-methylbutyl ester, germacrene D, 1,5,7-Octatrien-3-ol, 3,7-dimethyl-, 2-Octanol, pyrazine, 2-methoxy-3-(1-methylpropyl)-, and pyrazine, 2-methoxy-3-(1-methylethyl)-, among which the OAVs of them were greater than 1. Nonanal was a considerable contributor to the aldehyde, citrus, and orange peel odor of PGG, germacrene D had woody and spicy odor, and germacrene D, 2-Octanol was major contributor to spicy, herbal, and bad smell odor, and pyrazine, 2-methoxy-3-(1-methylpropyl)- and pyrazine, 2-methoxy-3-(1-methylethyl)- would likely be responsible for galbanum, bell pepper, and pepper odor. Meanwhile, the OAVs of pyrazine, 2-methoxy-3-(1-methylpropyl)-, 2-Octanol, and pyrazine, 2-methoxy-3-(1-methylethyl)- were 1.94E+02, 2.25E+04, and 1.75E+03, respectively. 3-Octen-1-ol, (*Z*)- was also present with herbal and spicy odor. Therefore, 2-Octanol and 3-Octen-1-ol, (Z)- were the characteristic volatile compounds in PGG samples.

Hence, SHC samples were present with floral, fruity and sweet odor characteristics, while PGG had spicy, herbal, and bad smell odor, which were in agreement with the sensory evaluation of aroma.

3.3. *Correlations between non-volatile compounds and volatile compounds in PGG and SHC samples*

The differential volatile compounds in PGG and SHC leaves were mainly consisted of esters, alcohols, aldehydes, ketones, lactones and terpenoids, which were mainly generated from fatty acid metabolic pathway, terpenoid metabolic pathway and amino acid metabolic pathway (Chen et al., 2020).

As mentioned above, C8 compounds, 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-Hexen-1-ol, acetate, (*E*)-, and 3-Hexen-1-ol, acetate, (*Z*)-, as the characteristic volatile substances, were found to be critical contributors to the sweet, floral, and fruity odor in SHC samples, which were usually produced by lipids catalyzed by lipoxygenase. 4-Hexenal, 3-Hexenal, (Z)- and 3-Hexenal, (E)- was produced by α-linolenic acid decomposed by hydroperoxidase lyase (HPL), and then reduced to 4-Hexen-1-ol, 3-Hexen-1-ol, (Z)- and 3-Hexen-1-ol, (E)-, respectively, with the participation of alcohol dehydrogenase. 4-Hexen-1-ol, acetate, 3-Hexen-1-ol, acetate, (E)-, and 3-Hexen-1-ol, acetate, (Z)- were produced by 4-Hexen-1-ol, 3-Hexen-1-ol, (Z)- and 3-Hexen-1-ol, (E)-,

respectively, under esterification (Fig. 6). Those results were consistent with the findings in differential lipids that the content of α-linolenic acid was significantly higher in SHC samples than that in PGG samples. In PGG samples, 2-octanol, and 3-Octen-1-ol, (Z)- were selected as the characteristic volatile compounds and main contributors to herbal and spicy odor, which were derived from α-linoleic acid by LOX and subsequent cleavage by HPL. Results of 2-octanol, and 3-Octen-1-ol content were concordance with that of PGG with higher α-linoleic acid content (Fig. 6). The results elucidated that α-linolenic acid and α-linoleic acid were the main source of the aroma difference between PGG and SHC samples.

In addition, L-histidine and L-tyrosine (bitter amino acids) were presented with higher accumulation in PGG samples, which might partially illustrate the result that PGG had a strong bitter taste. Except for the significant contribution to bitter taste, amino acids were also believed to be vital precursors for the synthesis of aroma components (Han et al., 2024). The carbonyl derivatives generated from the interaction between amino acids and carbohydrates were involved in the Strecker degradation reaction, which in turn produced aroma substances, such pyrazine, 2-methoxy-3-(1-methylpropyl)- and pyrazine, 2-methoxy-3-(1-methylethyl)- with higher content in PGG samples. Germacrene D, (+)-Dihydrocarvone and hotrienol were derived from the terpene skeleton, which were also contributed to the herbal, woody, spicy odor of PGG samples. Carotenoid degradation produced trans-β-ionone, ionone, and α-irone, which were higher in SHC samples with floral odor.

In general, the combination of fatty acid biosynthesis pathway derived from lipids, terpenoid biosynthesis pathway derived from carbohydrates, and amino acid biosynthesis pathway derived from proteins provided PGG and SHC samples with unique aroma characteristics.

4. Discussion

SHC and PGG, geographical indication agricultural products, are deeply loved by consumers with unique taste, rich nutrients, and health effects. The tender leaves and buds of SHC and PGG are the high-quality sources of protein, dietary fiber, flavonoids, amino acids, vitamins, polysaccharides and lipids, and have broad prospects for plant-original new food source development (Yao et al., 2022). Therefore, how to explain the regional characteristics of SHC and PGG from the perspective of flavor substances and aroma substances is particularly important.

Phenolic acids, alkaloids, flavonoids, amino acids, organic acids, and soluble sugars are the material basis for the richer taste (Fan et al., 2024). Flavonoids and phenolic acids with higher content in leaves display a leading role in the flavor formation, and organic acids, soluble sugars, and free amino acids with lower content could also improve the overall taste (Ye et al., 2022). The metabolic differences of amino acids and flavonoids in black tea from the two regions were significant, resulting in different tastes and quality (Lin et al., 2022). SCHARBERT et al. found that aglycones in the chemical structure of flavonol glycosides affected the astringency perception of flavonol glycosides (Scharbert & Hofmann, 2005). Catechins are a class of compounds with dual taste attributes (bitterness and astringency) (Zhang, Li, et al., 2023). It was found that epigallocatechin gallate mainly changed into astringency and bitterness in the study of visual simulation intensity scale evaluation of taste intensity (Zhang et al., 2020). Phenolic acids are another kind of harsh taste compounds, mainly including gallic acid, quinic acid, chlorogenic acid, caffeoylquinic acid and p-coumaroylquinic acid (Ye et al., 2022). Glutamic acid and aspartic acid, the 2 main umami free amino acids, have a synergistic effect on umami taste (Kaneko et al., 2006). The sour substances were mainly composed of organic acids, free amino acids and phenolic acids, such as malic acid, citric acid, succinic acid, and fumaric acid (Chen et al., 2022). Here, the utilization of UPLC-MS/MS was to construct and reveal the flavor profile of SHC and PGG. In SHC samples, epicatechin, catechin, 4-O-coumaroylquinic acid, chlorogenic acid, neochlorogenic acid and cryptochlorogenic acid exhibited a higher accumulation, which were contributed to the harsh and bitter taste. In PGG samples, quercetin-3-O-

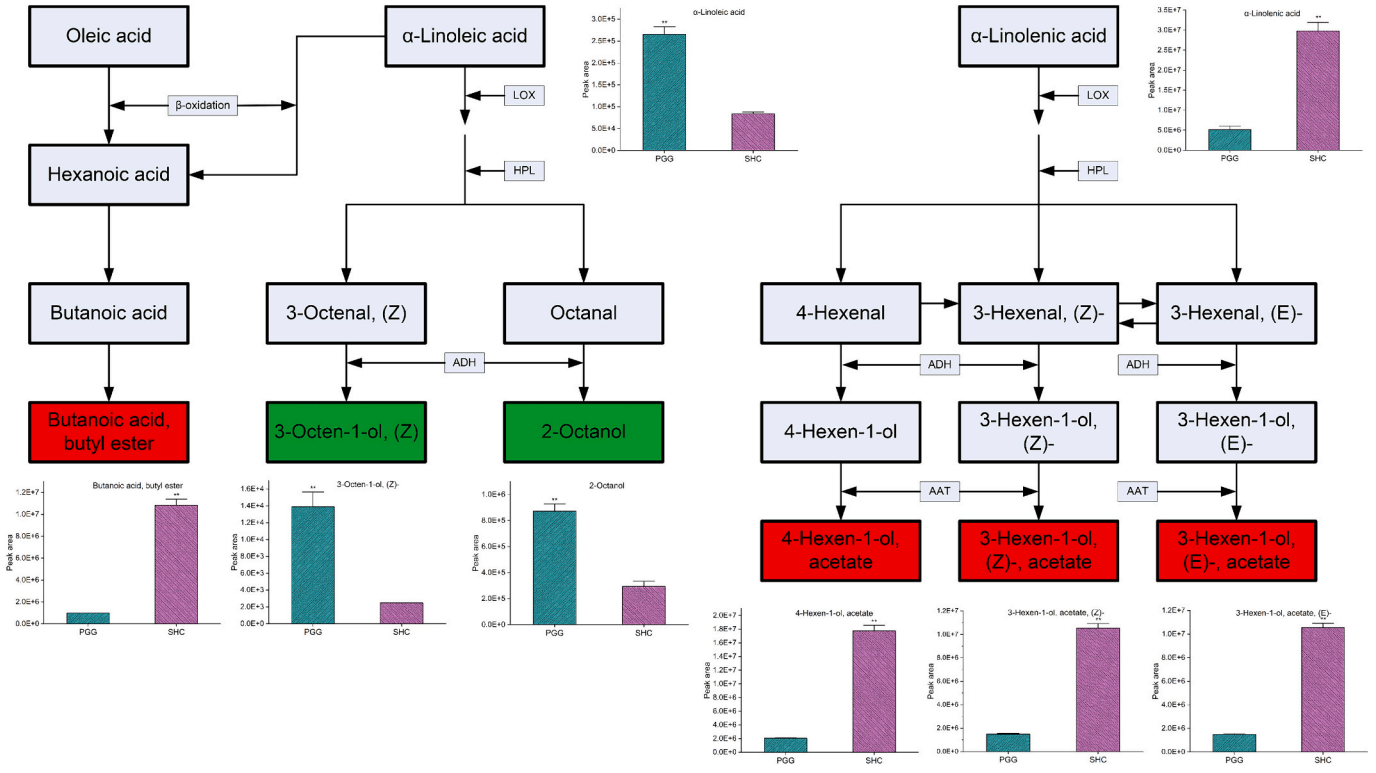

Fig. 6. The biosynthesis pathway of 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-hexen-1-ol, acetate, (*E*)-, and 3-hexen-1-ol, acetate, (*Z*)-, in SHC samples, and 2-octanol, and 3-Octen-1-ol, (Z)- in PGG samples. **, $p < 0.01$.

sambubioside, quercetin-3-O-glucoside, quercetin-7-O-glucoside, hesperetin-5-O-glucoside, gallocatechin, gallocatechin gallate, epigallocatechin, and epicatechin gallate displayed a higher expression, which were the main contributors to harsh and bitter taste. Moreover, L-tyrosine and L-histidine were significantly enhanced at PGG samples, resulting in increasing the bitter taste. Citric Acid and citraconic acid were up-regulated in SHC samples, increasing the acidic flavor. L-aspartic acid and Ile-Asp were found at higher accumulation levels in PGG samples, and L-glutamic acid, His-Ala-Gln, Ser-His-Gln, and N-Acetyl-L-glutamine were significantly enhanced in SHC samples, which were the characteristic umami taste compounds in PGG and SHC samples. Our results were in concordance with the results that epicatechin, L-glutamic acid, L-aspartic acid, L-lysine, L-tyrosine, L-histidine chlorogenic acid, and flavonol glycosides made great contributions to the different taste formation of black tea originated from two regions (Lin et al., 2022). Hence, the quality and flavor of SHC and PGG was induced by the significant difference in flavonoids, amino acids and derivatives, and phenolic acids.

The aroma of leaves is greatly controlled by the type and content of aroma compounds present (Liu et al., 2023; Zeng et al., 2023). The variety is the fundamental reason for determining the difference of aroma substances and aroma characteristics (Yue et al., 2023). Sequential linalool, decanal, β-ionone, dimethyl sulfide, geraniol, heptanal, nonanal, (*Z*)-3-hexenyl hexanoate, and (*E*)-2-nonenal were identified as the key aroma components in the 6 high-quality tea leaves strains through HS-SPME-GC–MS (Ma et al., 2019). (E)-β-ionone, nonanal, linalool, geraniol, β-myrcene, heptanal, and hexanal were selected as the dominant components leading to aroma difference among the 9 black tea samples by using HS-SPME-GC–MS through OPLS-DA analysis (Yang et al., 2022). Unique aroma characteristics in different leaves, such as fruity, sweet, floral odor and so on, were induced by the difference in the composition, content and characteristic aroma of its aroma substances (Wu et al., 2024). The five Congou black tea varieties mainly presented floral, fruity, and sweet aromas, with the six of main aroma components being decanal, linalool, benzyl alcohol, geraniol, hexanal, and isovaleraldehyde (Xu et al., 2023). 1-octen-3-ol, 3-octanol, 1-octen-3-one, and 3-octanone with low odor threshold were considered as the key contributors of mushroom aroma in *Morchella importuna* (Zhang, Cui, et al., 2023). In our study, 1186 volatile compounds were detected, among which 536 differential volatile compounds were identified. Esters, heterocyclic compounds, ketones, hydrocarbons, alcohol, amine in SHC samples were higher than those in PGG samples, whereas aromatics, sulfur compounds, halogenated hydrocarbons, and terpenoids were abundant in PGG samples. 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-Hexen-1-ol, acetate, (E) -, and 3-Hexen-1-ol, acetate, (*Z*)- were identified as the characteristic aroma compounds in SHC samples, which made a significant contribution to sweet, floral, fruity odor with low odor threshold. 2-octanol and 3-Octen-1-ol, (Z)- were the dominant aroma compounds in PGG samples, causing the herbal, spicy and bad smell odor.

Numerous volatiles compounds were generated through fatty acid metabolic pathways, terpenoid metabolic pathways, and amino acid metabolic pathways under the catalytic action of some enzymes (Tu et al., 2023; Zhang et al., 2021). Fatty acids are the main aroma precursors for the formation of various aroma components, and the fruity, sweet, floral odor were generated from alcohols, aldehydes, ketones and esters, which were mainly derived from through the lipoxygenase pathway by the gradual peroxidation, cracking and reduction of precursor fatty acids (Domínguez et al., 2019; Tasaki et al., 2019). The accumulation level of α-Linolenic acid was remarkably differently between SHC and PGG samples, which might be the key to reveal the aroma differences. Moreover, α-linolenic acid had a higher content in SHC samples than PGG samples, which was favorable to form 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-Hexen-1-ol, acetate, (E) -, and 3-Hexen-1-ol, acetate, (Z)-. The higher α-linoleic acid content in PGG samples was beneficial to generating 2-octanol, and 3-Octen-1-ol. Pyrazine, 2-methoxy-3-(1-methylpropyl)- and pyrazine, 2-methoxy-3-(1-methylethyl)- with higher content in PGG samples were generated from carbonyl derivatives participated in the Strecker degradation reaction, which were produced by amino acids interacting with carbohydrates. Germacrene D, (+)-Dihydrocarvone and hotrienol were derived from terpenoid metabolic pathways (Li et al., 2020), and trans-β-ionone, ionone, and α-irone were generated from the carotenoid degradation (Yang et al., 2013). Thus, the unique aroma characteristics of PGG and SHC samples were caused by interaction of lipids, amino acids, and terpenoids.

5. Conclusion

In our study, UPLC-MS/MS and HS-SPME-GC–MS methods were utilized to reveal the flavor profiles of PGG and SHC samples. 1797 non-volatile compounds and 1186 volatile compounds were detected, among which 1218 non-volatile compounds and 536 volatile compounds were identified. Epicatechin, catechin, 4-O-coumaroylquinic acid, chlorogenic acid, neochlorogenic acid and cryptochlorogenic acids were significantly up-regulated in SHC samples, which were the main contributors to harsh and bitter taste. Quercetin-3-O-sambubioside, quercetin-3-O-glucoside, quercetin-7-O-glucoside, hesperetin-5-O-glucoside, gallocatechin, gallocatechin gallate, epigallocatechin, epicatechin gallate were significantly up-regulated in PGG, and were contributed to bitter and harsh taste of PGG. Citric Acid and citraconic acid increased the acidic flavor of SHC. L-aspartic acid and Ile-Asp were significantly enhanced at PGG samples with umami taste, and his-Ala-Gln, Ser-His-Gln, and N-Acetyl-L-glutamine were the characteristic umami taste compounds in SHC samples. L-tyrosine and L-histidine with higher content in PGG were increased the bitter taste. α-Linolenic acid was significantly enriched in SHC samples, and it produced C8 compounds, such as 4-Hexen-1-ol, acetate, butanoic acid, butyl ester, 3-Hexen-1-ol, acetate, (E) -, and 3-Hexen-1-ol, acetate, (Z) -, which were particularly contributed to sweet, floral, fruity odor with low odor threshold. 2-octanol, and 3-Octen-1-ol, (*Z*)- were derived from α-linoleic acid, presenting with herbal, spicy and bad smell odor in PGG samples. Comparative analysis elucidated that flavonoids and phenolic acids, amino acids, alcohol and esters compounds had a great effect on taste and odor of PGG and SHC samples. Our study elucidated the characteristic flavor compounds and internal causes of the difference in taste and odor quality, and provided new evidence for the difference in flavor between the metabolic profiles between PGG and SHC.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2024.101811.

Fundings

This study was supported by the "Talent Initiation Project of Shaanxi University of Technology" (SLGRCQD-2307) and the "State Key Laboratory (Cultivation) of Qinba Mountains Biological Resources and Ecological Environment, 'co-construction of city and school' scientific research project industrialization project" (SXC-2309).

CRediT authorship contribution statement

Tao Zheng: Writing – review & editing, Funding acquisition, Conceptualization. **Zhuang Deng:** Visualization, Investigation. **Min Tian:** Visualization, Investigation. **Qi Tang:** Investigation, Data curation. **Zhubing Hu:** Software, Methodology. **Guodong Wang:** Resources, Methodology. **Haitao Zeng:** Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

All authors declared that they had no known competitive financial interests or personal relationships in this manuscript.

Data availability

Data will be made available on request.

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