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The influence of different geographical origins and grades on the flavor of Qingxiangxing Baijiu: An integrated analysis using descriptive sensory evaluation, GC×GC–MS, E-nose, and ICP-MS

Qiao Huang [a], Xianping Qiu [a,b], Tongwei Guan [a,*], Jianshen Yu [b], Yichen Mao [c], Yu Li [d], Yuan Rao [d], Hongguang Shang [e], Yanhui Zhao [f]

[a] College of Food and Biological Engineering, Xihua University, Food Microbiology Key Laboratory of Sichuan Province, Chengdu 610039, PR China
[b] Sichuan Quanxing Liquor Co., Ltd., Chengdu, 611637, PR China
[c] Xinjiang Kaiduhe Liquor Co., Ltd, Hejing 841302, China
[d] Sichuan Tujiu Liquor Co., Ltd, Nanchong 637919, PR China
[e] Chengdu Shuzhiyuan of Liquor Co., Ltd, Chengdu 610000, PR China
[f] Sichuan Shuncheng textile Co., Ltd, Nanchong 637005, PR China

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ABSTRACT

The origin and grade are the important factors that influence the flavor and price of Qingxiangxing Baijiu (QXB). As the grade increased, sensory differences and geographical differences become more pronounced. The aromas of floral, fruity, and sweet become more noticeable, while the acidity decreased. The different substances among three grades of QXB mainly focused on esters and alcohols. Correlation analysis indicated that the increase in the concentration of ester compounds was one of the reasons for the significant differences in floral, fruity, and sweet fragrances. Because of the higher concentration of acidic compounds in northern QXB, its acidic taste was more pronounced. Meanwhile, the esters and ketones in southern QXB accounted for a higher proportion, which made its floral and fruity fragrance more prominent. PLS-DA was more suitable for the classification of QXB based on grade and region. And the metal elements in QXB could be used for regional identification.

1. Introduction

Baijiu is China's national liquor and is beloved by people worldwide. It boasts a rich and captivating history closely intertwined with Chinese culture. The flavor analysis shows that ethanol and water account for approximately 98 % of the total mass of the Baijiu, while the remaining 2 % is attributed to flavor components (Zheng & Han, 2016). The flavor compounds mainly consist of alcohols, esters, aldehydes, ketones, acids, acetals, furans, terpenes, nitrogen-containing compounds, and sulfur-containing compounds. The changes in the composition and proportion of these compounds result in 12 different flavor characteristics (Niu et al., 2017). Among them, Qingxiangxing Baijiu (QXB) has the longest history and is the origin of all types of Baijiu.

The QXB originated in Shanxi Province, located in North China. Because of its colorless and transparent nature, pure fragrance, elegant and harmonious characteristics, it is also deeply loved by consumers in the southern region (Fan et al., 2024). And the south and north of the QXB appeared during the continuous development of the QXB. However, the fermentation process of Baijiu is very complex and sensitive to numerous external environmental factors, resulting in variations in sensory quality for the same flavor across different regions (Song et al., 2020). Previous studies on Nongxiangxing Baijiu from different regions have found that the concentrations of trimethylpyrazine and 2-acetyl-5-methylfuran in samples from the Yanghe region are significantly lower than those in the Sichuan region ($P < 0.01$), resulting in a more intense roasted aroma in the Sichuan samples (Song et al., 2020). The sensory

Abbreviations: QXB, Qingxiangxing Baijiu; GC–MS, gas chromatography–mass spectrometry; GC-O-MS, gas chromatography-olfactometry-mass spectrometry; FGB, first-grade Baijiu; PGB, premium-grade Baijiu; SGB, special-grade Baijiu; GC × GC–MS, comprehensive two-dimensional gas chromatography mass spectrometry; ICP-MS, inductively coupled plasma-Mass Spectrometry; ANOVA, the analysis of variance; PCA, principal compounds analysis; PLS-DA, partial least squares discriminant analysis; VIP, variable importance in projection; STAMP, statistical analysis of taxonomic and functional profiles; SQXB, southern Qingxiangxing Baijiu; NQXB, northern Qingxiangxing Baijiu; OAV, odor activity value.

* Corresponding author.
E-mail address: guantongweily@163.com (T. Guan).

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analysis shows that the aroma differences of Jiangxiangxing Baijiu from different regions are mainly reflected in the odors of “sauce”, “roast”, and “grass” ($P < 0.05$), and the odor intensity of “sauce” and “roast” in the Guizhou samples is the highest (Li et al., 2024). However, at present, the research of QXB mainly focuses on raw materials, fermentation, and cellaring. For example, gas chromatography–mass spectrometry (GC–MS), gas chromatography-olfactometry-mass spectrometry (GC-O-MS), aroma recombination, and aroma omission methods are used to analyze the aroma components of 5 different types of sorghum. It is found that acetic acid, ethyl acetate, ethyl lactate, phenylethanol, and ethyl caproate may be the key aroma compounds contributing to the differences in aroma characteristics among the 5 kinds of sorghum during brewing (Ma et al., 2024). The initial water content of fermented grains, fermentation temperature, and spatial location of fermented grains all affect the fermentation process and the quality of the base Baijiu (Huang et al., 2024; Tian et al., 2024; Wang, Sun, et al., 2023). Wang, Wu, et al. (2023) conducted a study on the QXB of different years and found that aged Xiaoqu Baijiu contains more active aroma components compared to fresh samples, with many components exhibiting higher flavor dilution values in aged samples. The geographical origin classification of alcoholic beverages has been the subject of many studies so far, with a primary focus on wine, rum, Nongxiangxing Baijiu, and Jiangxiangxing Baijiu (Jorge et al., 2008; Li et al., 2024; Song et al., 2020). However, there have been few studies on the classification of QXB based on its geographical origin.

In addition to geographical origin, the difference in grade is also an important factor that affects the flavor and price of Baijiu. China’s national standard classifies QXB into three grades: first-grade Baijiu (FGB), premium-grade Baijiu (PGB), and special-grade Baijiu (SGB) (GB/T 10781.2-2022). The prices of different grades of QXB range from dozens of yuan to thousands of yuan. The trend of interest has led some merchants to produce fake QXB using inferior products. However, there is limited research on the flavor of finished QXB, particularly regarding the composition and differences in flavor among different grades of finished QXB. Therefore, it is necessary to explore the flavor composition of each grade of QXB and identify the grade markers that can help distinguish between different grades.

Therefore, the main purposes of this study are: (1) to explore the sensory and volatile compound composition differences in different regions and grades of QXB; (2) to identify markers for distinguishing between regions and grades of QXB; (3) to determine whether the metal element composition in QXB has potential for grade and regional differentiation, as well as explore the influence of metals on flavor.

2. Materials and methods

2.1. Baijiu samples

In this study, 6 types of QXB were purchased from 6 manufacturers, with each type being obtained in three grades: first grade (L1-L6), premium grade (M1-M6), and special grade (T1-T6). A total of 18 samples, with each sample purchasing three bottles in parallel. The details of the samples are listed in Table S1. These manufacturers are representative in the region, with a stable production process and large sales volume. Store at 4 °C before further analysis.

2.2. Reagents and chemicals

All chemicals and internal standards (ISs) are of high purity grade (GC grade) and were obtained from Shanghai Titan Technology Co., LTD. (Shanghai, China). Mg, Al, Na, Li, V, Cr, Mn, Be, Sr, Co, In, Ni, Cu, Ga, Bi, As, Se, Ag, Cd, Ba, and Pb were purchased from Gangyannake Testing Technology Co., Ltd. (Beijing, China). The purity of all standards was≥98 %.

2.3. Sensory analysis

The previous references were used for sensory quantitative description analysis (Huang et al., 2022; Wang, Wu, et al., 2023). The aroma profiling was conducted by a panel of 6 assessors, comprising an equal number of male and female participants. After approximately one year of training, the assessors will have mastered the method for sensory evaluation of Baijiu. 7 odor descriptors associated with the sensory characteristics of QXB were chosen: floral, fruity, alcoholic, sweet, acidic, grassy and grain. The panelists then assessed the strength of each descriptor for each sample on a scale ranging from 0 (indicating very weak) to 9 (indicating very strong). The samples were subjected to three consecutive sniffs, with a 10-min time interval between each sniff. The results obtained from 6 evaluators were averaged and subsequently visualized as a radial plot.

2.4. Comprehensive two-dimensional gas chromatography mass spectrometry (GC × GC–MS) analysis

A 5 mL sample of Baijiu was placed in a 20-mL headspace bottle. A 1.5 g of sodium chloride solid was added to the headspace vial to promote saturation of the sample solution. Then, it was mixed with 20 μL of an internal standard solution (2-octanol, 0.822 mg/L). GC × GC–MS analysis was performed using an Shimadzu QP2020NX (Shimadzu Enterprise Management Co., Ltd.; Shanghai; China). The one-dimensional column was polar DB-WAX (30 m × 0.25 mm × 0.25 μm), and the two-dimensional column was medium polar DB-17 ms (1.2 m × 0.18 mm × 0.18 μm). Sample pretreatment: The sample was analyzed using headspace solid-phase microextraction technology. The sample was preheated and balanced for 15 min at 60 °C, and then a three-phase S2 solid-phase microextraction probe is inserted (DVB/C-WR/PDMS, 1 cm, Guangzhou ZhiDa Laboratory Technology Co., Ltd., InLab-4FS0-007). Headspace extraction was carried out at 60 °C for 40 min, and then inserted into the gas chromatograph inlet (inlet temperature 250 °C) for 3 min thermal analysis. The column carrier gas was high-quality helium (≥99.999 %) flowing at a rate of 1 mL/min. The temperature program is set as follows: The initial temperature is 40 °C and it is maintained for 5 min. Then, it is heated at a rate of 5 °C per minute to reach 230 °C and maintained for 9 min. The evaporation chamber temperature remains at 250 °C, and no fractionation is performed. The mass spectra were obtained in electron ionization mode using an ionization energy of 70 eV and a scan range from m/z 50 to 550 amu. The temperature of the ion source was set at 230 °C. The solution of C_6–C_{30} n-alkanes was analyzed using the same chromatographic conditions to calculate linear retention index (LRIs) based on retention times. The identification and quantification procedures for volatile compounds can be referenced from the previously reported method (Wang, Wu, et al., 2023).

2.5. Inductively coupled plasma-mass spectrometry (ICP-MS) analysis

The samples were tested according to the method described by Wei et al. (2023). Concentrate the 25 mL sample to 1 mL using a 65 °C water bath, and then transfer it to a 25 mL volumetric flask. The process should be repeated with the 1 % nitric acid solution for approximately three to five cycles, with the washing liquid being transferred to the volumetric flask at each stage. Finally, the solution should be diluted to 25 mL using a 1 % nitric acid solution, and a blank test should be performed. The inductively coupled plasma mass spectrometer (PerkinElmer ICPMS NexION 350×, PerkinElmer, America) was employed for the detection of metal ions. The RF power was 1.55 kW; the sampling depth was 8 mm; the plasma gas flow rate was 15 L/min; the auxiliary, carrier, and compensating/dilution gases were all set at 1 L/min each; the temperature of the atomization chamber was maintained at 2 °C; and the peristaltic pump speed was set to 0.1 rps (revolutions per second).

2.6. Electronic nose analysis

The PEN3 portable electronic nose (Win Muster Airsense Analytics Inc., Schwerin, Germany), equipped with a set of ten metal oxide gas sensors, was employed in this study. The detailed descriptions of the features for each sensor can be found in Table S2. Prior to electronic nose analysis, a 15 mL Baijiu sample was placed in a 60 mL injection sample bottle specifically designed for electronic nose analysis and allowed to equilibrate at room temperature for 15 min, ensuring complete accumulation of the volatile components present in the sample. The procedure for analysis was as follows (Xu, Guo, et al., 2022): the injection needle and air tonic needle were inserted at the same time. After sampling for 60 s, the injection needle was first removed, followed by the air tonic needle. Then, the system was cleaned (cleaning time lasted for 300 s) and standardized before each sample was determined three times in parallel.

2.7. Statistical analysis

The analysis of variance (ANOVA) was used to test the significance of assay ($P < 0.05$) with software SPSS 25.0. Origin 2019 and GraphPad Prism (v8.0.1) software were utilized for data processing. Principal compounds analysis (PCA), partial least squares discriminant analysis (PLS-DA), variable importance in projection (VIP), redundancy analysis (RDA), and statistical analysis of taxonomic and functional profiles (STAMP analysis) were performed with the online tools (https://www.omicstudio.cn/index, https://www.omicshare.com/tools/Home, and https://www.chiplot.online).

3. Results and discussion

3.1. Sensory analysis

Sensory evaluation was performed on 18 samples in this study (Fig. 1A). Sensory analysis indicated a clear difference between the three grades of QXB. There were significant differences between FGB and PGB in floral, fruity, and sweet ($P < 0.05$). There were significant differences between PGB and SGB in floral, fruity, sweet, and grain ($P < 0.05$). FGB and SGB exhibited extremely significant differences in floral, fruit, sweet, acidic, grain, grassy, and alcoholic ($P < 0.05$). As the grade increased, the sensory differences become more pronounced, with the aromas of fruit, floral, and sweet becoming more noticeable, and acidity decreasing. Previous studies have found that as the grade increases, the fruit and sweet fragrances of Zhimaxiangxing Baijiu become more pronounced, which is consistent with the findings of this study (Qin et al., 2023). Additionally, sensory data could differentiate three grades of QXB (Fig. 1B), which also explains why sensory evaluation is one of the commonly used methods to distinguish the quality of Baijiu. For the same grade of QXB, as the grade increased, the sensory differences between southern QXB (SQXB) and northern QXB (NQXB) increased. The sensory differences between SQXB and NQXB mainly manifested in floral, alcoholic, and acidic. The acidic and alcoholic of NQXB were higher than those of SQXB, while the floral of SQXB was higher than that of NQXB. Previous studies have found that the floral aroma of SQXB is generally more pronounced than that of some NQXB brands, and the acidic of QXB from Xinhuacun and Hongxing, originating from the north, is significantly higher than that of Jinmen from the south (Niu et al., 2017).

To investigate the reasons for the sensory differences between different grades and regions of QXB, the next step was to further study the volatile compounds in 18 samples using solid-phase microextraction combined with GC × GC–MS.

3.2. Analysis on the composition and difference of volatile compounds in different grades of QXB

Through Venn analysis, there were 60 common flavor substances among the three grades of QXB. As the grade increased, the number of unique flavor substances in QXB increased (Fig. 2A). After conducting statistical analysis on the total volatile flavor content, the concentration of volatile compounds significantly increased with higher grades (Fig. 2C). The total concentration of flavor substances in SGB (3487.42 mg/L) was significantly higher than those in FGB (1694.72 mg/L) and PGB (2301.17 mg/L) (Fig. 2C). As the grade increased, not only did the concentration of ester compounds increased (from 784.64 mg/L to 1832.37 mg/L), but also the proportion of ester compounds in the total volatile substances increased significantly (from 45.39 % to 51.55 %) (Fig. 2B and C). Esters in baijiu are usually from three sources. A slight amount of esters are from the raw materials, and another route for ester synthesis is spontaneous chemical esterification. The microbial synthesis of esters is the third route and is usually recognized as the main synthetic way for esters in baijiu (Xu, Zhao, et al., 2022). Esters primarily possess fruit, floral, and sweet scents, such as ethyl acetate (fruit scent), ethyl caprate (floral scent), and ethyl laurate (sweet scent) (Ru et al., 2024). They contribute to the fruity, sweet, and more complex aroma of Baijiu. And they serve as the main sources of quality indicators and aroma in Baijiu (Pang et al., 2020). According to national standards, ethyl acetate is considered a characteristic aroma substance and a grade indicator for QXB (GB/T 10781.2-2022). The level of ethyl acetate in FGB was 423.29 mg/L, while the level in SGB was significantly higher at 911.42 mg/L. At the same time, esters accounted for a large proportion of the compounds with an OAV (odor activity value) value greater than 1 (Table S4). The study found that an OAV value greater than 1 indicates that the concentration of this component is above its olfactory threshold

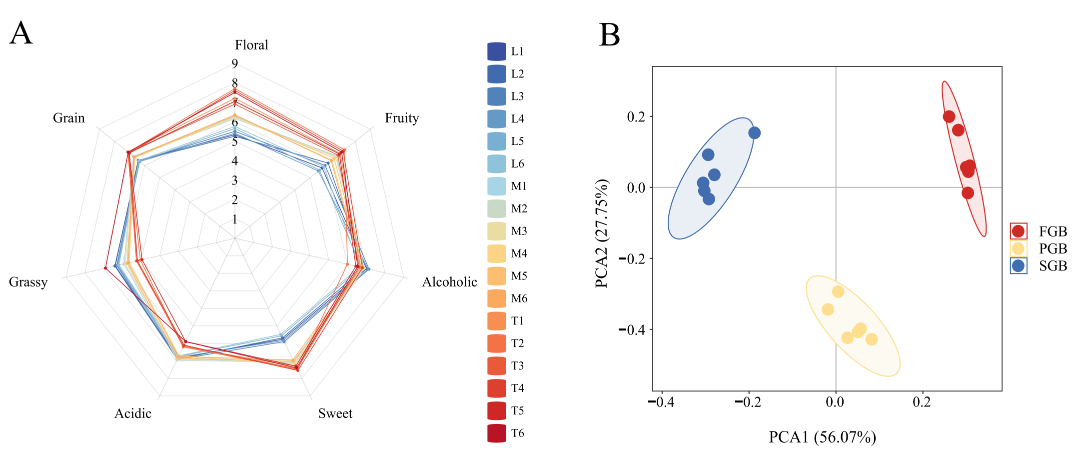

Fig. 1. Sensory radar map of all Qingxiangxing Baijiu samples (A).The principal component analysis (PCA) of sensory analysis (B).

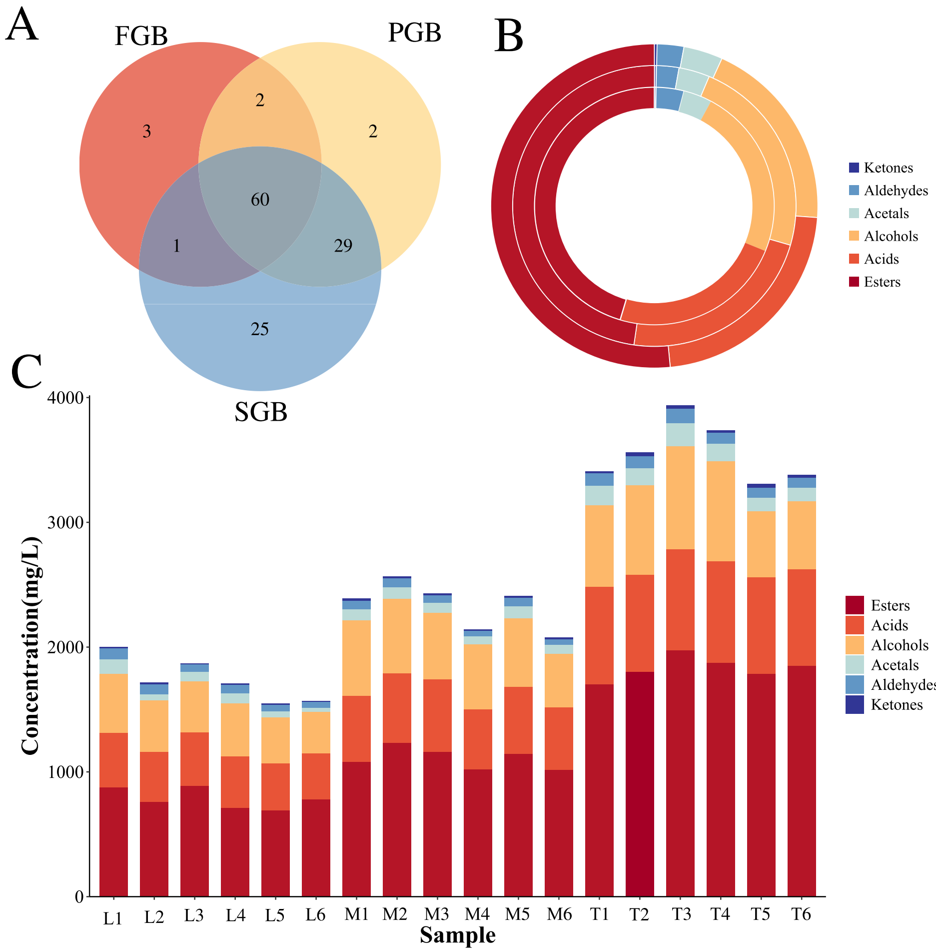

Fig. 2. Flavor substances in venn analysis (A), percentage of composition (B) and cumulative total content (C) of three grades Qingxiangxiang Baijiu.

and can significantly impact the overall odor. These esters, which produce floral, fruity, and sweet aromas, make a significant contribution to the flavor of QXB. Therefore, the increase in the content and proportion of ester compounds may be one of the reasons why the floral (ethyl caprate), fruity (ethyl acetate), and sweet (ethyl laurate) sensory experiences in QXB are enhanced. The increase in the content and proportion of ester compounds may be one of the reasons why the floral, fruity, and sweet sensory experiences in Baijiu were enhanced (Wang et al., 2024). As the grade increased, the content of alcohol compounds rose (from 403.66 mg/L to 687.80 mg/L), but the proportion decreased (from 23.35 % to 19.35 %) (Fig. 2B and C). The representative alcohol is ethanol with concentrations from 30 to 72 % (*v*/v) in baijiu. Ethanol synthesis in baijiu fermentation is mainly performed under anaerobic conditions by yeasts such as *Saccharomyces cerevisiae*. *Saccharomyces cerevisiae* converts glucose into pyruvic acid through the glycolysis pathway. Pyruvic acid is decarboxylated to generate acetaldehyde and then reduced to produce ethanol (Xu, Zhao, et al., 2022). However, it is generally believed that ethanol is the compound responsible for the taste rather than the aroma of baijiu, while non-ethanol alcohols influence the aroma. When the proportion is appropriate and reaches a balance, baijiu will present a sweet, pure, and palatable taste (Chen et al., 2023). Research has found that polyols play an important role in contributing to the sweetness of Baijiu. The multiple hydroxyl groups of polyols can activate sweetness receptors and enhance the perception of sweetness (Sun et al., 2021). By integrating into its body, polyols enhance the sweetness and richness of Baijiu, thereby promoting flavor enhancement (Wang et al., 2020). In this study, the types and contents of polyols in SGB were higher than those in FGB and PGB, which may enhance the sensory experience of SGB.

The higher the grade of QXB, the greater its content of acidic compounds and aldehyde compounds (Fig. 2C). While acids are characterized by a ‘sour’, ‘rancid’, and ‘sweaty’ odor under a sniffing device, their ability to contribute to aroma is poor, thus playing a smaller role in shaping the aroma of Baijiu or other liquors (Wang, Li, et al., 2022; Zhang et al., 2019). Despite this, acids play an essential role as trace compounds in shaping the style and mouthfeel of Baijiu. For example, acetic acid imparts a delightful sour taste to alcoholic beverages. Furthermore, acetic acid acts as the precursor of ethyl acetate and is considered one of the primary substances that determine its aroma (Miao et al., 2022; Zhou et al., 2021). Acetal compounds are mostly fruity in aroma, play a crucial role as indicators for aging Baijiu. They are believed to be the main substances that enhance the softness and aroma of wine (Wang, Wu, et al., 2023). Especially 1,1-diethoxypropane, 1,1-diethoxybutane, and 1,1-diethoxy-3-methylbutane are known as significant contributors to the flavor of Baijiu. In China’s famous and high-quality Baijiu, the higher the quality and grade of the Baijiu, the greater its acetal content. This increase in acetal content enhances both fragrance and mellowness (Jiang et al., 2023; Wang et al., 2024).

As the grade increased, the proportion of aldehyde compounds decreased from 3.78 % to 2.64 % (Fig. 2B). Acetaldehyde is an odorant compound that has a ‘spicy’ and ‘grassy’ aroma in Baijiu. There are mainly two ways for acetaldehyde to form in baijiu: the first is its production during alcohol fermentation. During Baijiu fermentation, yeast converts glucose into pyruvate and releases carbon dioxide, ultimately generating ethanol. The second way is ethanol oxidation. When ethanol is oxidized, acetaldehyde can also be produced. This is one of the main sources of acetaldehyde in finished baijiu (Jiang et al., 2023). Compared to FGB, the content of acetaldehyde in SGB was lower (78.81 mg/L - 65.05 mg/L). However, as the grade increased, the variety of aldehyde compounds improved. For example, SGB contained phenylacetaldehyde, undecanal, benzaldehyde, and butanal, which were not present in FGB (Table S3). Among them, phenylacetaldehyde and undecanal are floral and fruity fragrances frequently used in the production of floral scents (Wang et al., 2024).

No phenolic compounds were detected in FGB, but there were more sulfur compounds present (Table S3). Volatile sulfur compounds are a class of aromatic compounds with low odor thresholds and strong odorous characteristics due to the presence of sulfur in their molecular structures. However, sulfur compounds often have the odors of rotten eggs and spoiled cabbage, thereby also playing a significant role in influencing the flavor and off-flavors of numerous foods (Wang, Wu, et al., 2023). Methanethiol, a common sulfur compound in Baijiu, smells like rotten cabbage and burnt rubber (Song et al., 2019). The level of methanethiol in FGB was significantly higher than that in SGB (Table S3). Volatile phenols are important flavor compounds in Baijiu, providing unique smoky, caramelized, and earthy flavors. They are more abundant in the Jiangxiangxing Baijiu, followed by the Nongxiangxing Baijiu (Huang et al., 2018; Mao et al., 2019). Phenolic active compounds are widely recognized for their excellent antioxidant properties, which can help prevent certain conditions such as coronary heart disease, diabetes, high cholesterol levels, and gout (Mao et al., 2019). The amount and type of phenols detected in SGB were significantly higher than those found in FGB and PGB (Table S3).

We selected volatile compounds with $P < 0.05$, FC ≥ 2, and FC ≤ 0.5 as the differential aroma substances for generating the volcano plots. According to the volcano plot, 13, 16, and 9 metabolites were found to be upregulated ($P < 0.05$, FC ≥ 2) in the comparisons of FGB vs PGB, PGB vs SGB, and SGB vs FGB respectively. Additionally, 6, 6, and 38 metabolites were identified as downregulated ($P < 0.05$, FC ≤ 0.5) in the same comparisons (Fig. 3A-C). The comparison revealed that there were 19 distinct substances between FGB and PGB. Compared with FGB, the content of furfuryl alcohol (bitter) in PGB decreased significantly, while the content of certain aromatic hydrocarbons such as phenylethanol and phenylacetaldehyde increased (Fig. 3A). Meanwhile, the OAV values of phenylethanal and phenylethanol in PGB were significantly higher than those in FGB, and both were greater than 1 (Table S4). Aromatic hydrocarbons often have distinctive odors, such as "rose", "honey", "sweet", or "fruity". Phenylethanol is identified as having a "rose" and "honey" aroma. Phenylacetaldehyde has been described as smelling like "honey", "floral", "sweet" (Huang et al., 2018). Although the amount of aromatic hydrocarbons is small, they exert a significant impact on the

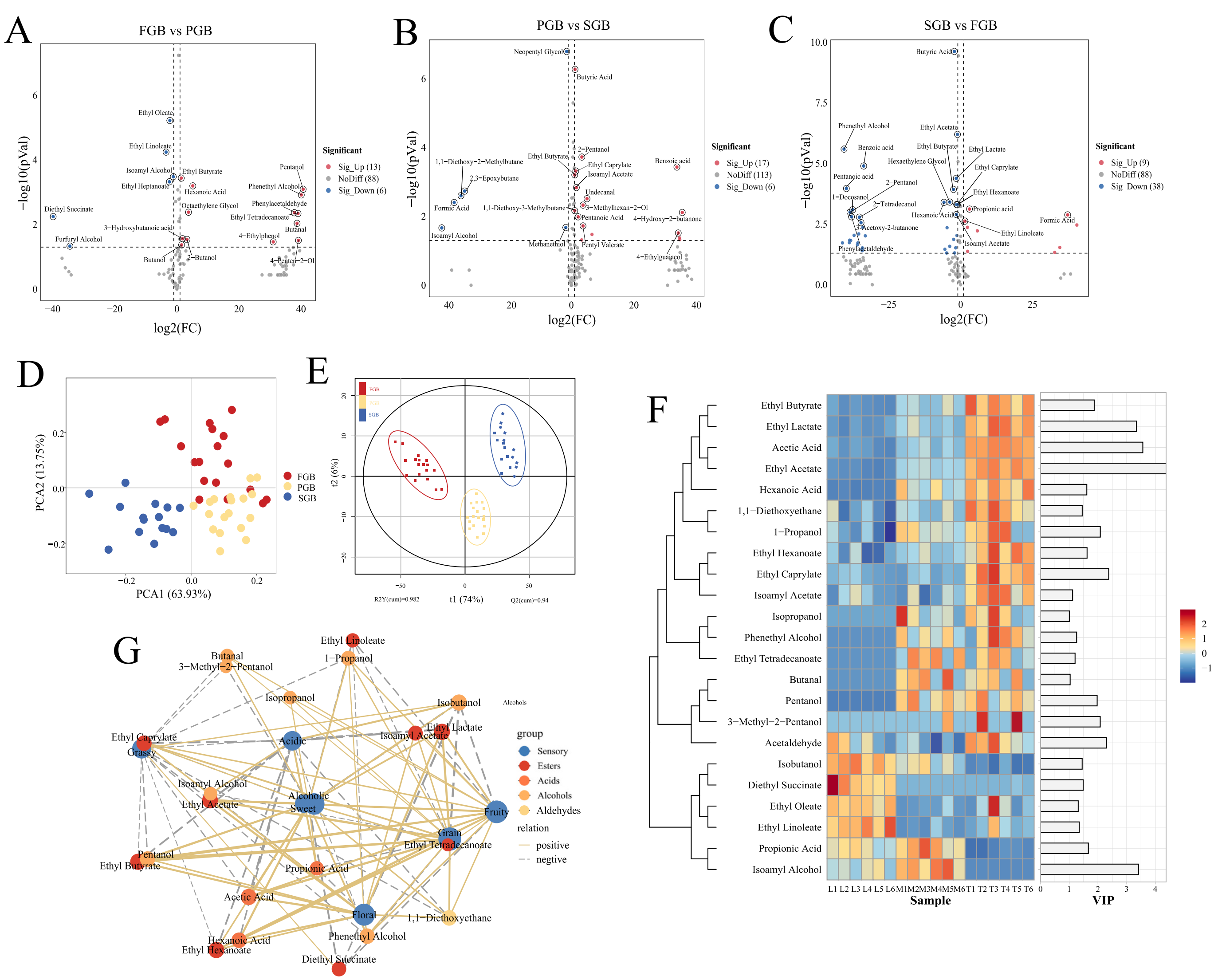

Fig. 3. Volcano plot of the differential metabolites of FGB vs PGB (A), PGB vs SGB (B), and SGB vs FGB (C), PCA analysis (D) and partial least squares discriminant analysis (PLS-DA) (E) of all samples, clustering heat map of flavor substances based on grade markers (VIP > 1) (F). Correlation network analysis of volatile flavor compounds (VIP > 1) and sensory (G).

overall flavor of Baijiu. This is due to their unique characteristics, such as distinctive aroma profiles, prolonged persistence, strong odor intensity, and low detection thresholds (Wang et al., 2024). The differences in the concentrations of these compounds may be one of the reasons why FGB and PGB had significant differences in floral, fruity, and sweet fragrances ($P < 0.05$). There were 21 differential substances between PGB and SGB. Among them, the contents of formic acid, isopentanol, and methanethiol were significantly lower in SGB (Fig. 3B). Formic acid is irritating, and methanethiol has a foul odor resembling rotten cabbage. There was a significant difference between SGB and FGB, with a total of 47 differential substances. The concentration of most flavor substances in the FGB group was lower than that in the SGB group, such as ethyl acetate, ethyl lactate, phenylethyl alcohol, and phenylethanal (Fig. 3C). In general, the differences between the three grades of QXB mainly lie in their ester and alcohol content.

3.3. PLS-DA was used to differentiate the grades of QXB and identify potential grade marker compounds

The PCA analysis were generated based on the quantitative untargeted omics data, as depicted in Fig. 3D. All samples of SGB were distinct from the other two groups, although there was some overlap between the FGB and PGB (Fig. 3D). The PCA analysis based on all volatile compounds cannot significantly separate the samples of different grades QXB. To achieve more accurate classification results, we applied a binary classification algorithm with PLS-DA to solve our multi-class problem by implementing the one-versus-the-rest framework. PLS-DA is a commonly used data analysis method that focuses on the differences between samples from different categories. Both PCA and PLS-DA can reduce the dimensionality of complex data, thereby selecting the major components with higher contributions. However, when analyzing complex omics data, PCA is not effective in distinguishing inter-group differences among complex samples in an unsupervised manner. Therefore, compared to PCA, the supervised PLS-DA model can better distinguish inter-group differences among complex samples (Li et al., 2024; Song et al., 2020). Prior to the work of Song et al. (2020), this strategy resulted in improved classification outcomes. PLS-DA analysis was performed on all volatile compound data, and a significant separation of samples at different grades was observed (Fig. 3E). The fitness of the classification model based on all untargeted flavor substances was 98.2 % ($R2Y(cum) = 0.982$). From the perspective of prediction ability, the model constructed from all untargeted flavor substances reached as high as 94.0 % ($Q2(cum) = 0.940$). The results indicated that PLS-DA was more effective in classifying different grades of QXB.

Furthermore, the VIP values were calculated based on the PLS-DA model using QXB samples of different grades (Fig. 3F). VIP analysis is often used to screen for potential differential compounds, and it has been confirmed that volatile compounds with VIP > 1 have a high discrimination potential (Wang, Zhang, et al., 2022). The VIP scores for the 23 volatile compounds were > 1 (Fig. 3F). These volatile compounds were considered potential markers that could cause differences in grade among QXB samples. Among them, ethyl butanoate, ethyl hexanoate, and hexanoic acid not only serve as marker compounds in different grades of QXB but also exhibit significant concentration differences among various grades of Zhimaxiangxing Baijiu (Qin et al., 2023). Previous studies have found that acetic acid, ethyl acetate, and ethyl tetradecanoate can serve as differential markers for different grades of Jiangxiangxing Baijiu (Huang et al., 2023). In this study, ethyl acetate had the highest VIP value (VIP $= 4.37$) mainly because it is an important indicator for evaluating the quality of QXB according to national standards (GB/T 10781.2–2022), and its content varies significantly among different grades of QXB ($P < 0.05$). The 23 compounds could be roughly divided into two groups, according to the clustering results (Fig. 3F). The first group consisted of isobutanol, propionic acid, isoamyl alcohol, etc. that exhibited a high concentration in FGB. Isoamyl alcohol and isobutanol serve as the primary constituents of fusel oil in QXB (Huang et al., 2024). The intoxicating and anesthetic effects of higher alcohols on the human body are stronger than those of ethanol itself, and their metabolism is slow, resulting in a long residence time in the human body. It is a type of substance that contributes to the bitterness and impurity of QXB, thereby affecting post-drinking comfort (Wang et al., 2024). This may be the reason why people feel more uncomfortable when drinking cheaper drinks. The second group consists of the remaining 17 compounds, which mainly comprised ester compounds and had a higher concentration in SGB. Odor network analysis of volatiles (VIP > 1) in QXB revealed significant positive correlations between most esters, such as ethyl hexanoate, ethyl acetate, ethyl lactate, etc., and sweet, fruity, and floral (Fig. 3G). These results indicated that the significant difference in the concentration of these compounds was one of the reasons for the significant difference in the sweet, fruity, and floral characteristics among the three grades of QXB.

3.4. Different sources of QXB distinction

By conducting PCA analysis on the same grade of QXB, it was found that samples from different regions were clearly dispersed (Fig. 4A–C). As the grade of QXB increased, the dispersion of the samples become more noticeable. The higher the grade of QXB, the more beneficial it was for source identification. The PCA analysis in this study also revealed that the samples were divided into two groups on the y-axis, with one group originating from southern China and the other from northern China. The dispersion distance between samples from the southern and northern SGB was more pronounced (Fig. 4C). It indicated that the higher the grade of Baijiu, the greater the influence of regional factors, and the differences in the composition of volatile aroma substances become more apparent.

Due to variations in raw materials, production processes, and production environments, there are flavor discrepancies among different regions of Baijiu (Li et al., 2024). Further investigation into the differences in flavor compounds between southern and northern QXB through non-targeted omics data was needed. At the same level, there was no significant difference in the total concentration of flavor substances between samples from southern China and northern China. However, overall, the total flavor concentration of NQXB was slightly higher than that of SQXB (Fig. 4D and E). Compared to the north, the proportion and diversity of ester compounds in southern Baijiu were higher. For acidic compounds, the total acid content of NQXB was higher than that of southern Baijiu (Fig. 4D). Through correlation analysis, it was found that acidic substances such as acetic ccid, propionic acid, and hexanoic acid were significantly positively correlated with the perception of acidity (Fig. 5D). Meanwhile, the OAV analysis also showed that the OAV values of acetic acid, butyric acid, and pentanoic acid in NQXB were higher than those in SQXB. This may be one of the reasons why NQXB had a stronger acidic than SQXB. Previous studies have found that the acidity of QXB from Xinghuacun and Hongxing, originating from the north, is significantly higher than that of Jinmen wine from the south (Lai et al., 2024). Interestingly, some studies have found that the total acid content of Nongxiangxing Baijiu is lower in the north than in the south (Song et al., 2020; Wang et al., 2024). The levels and proportions of aldehydes and acetals were higher in the northern samples compared to the southern samples, with significantly higher levels of acetaldehyde observed in NQXB than in south ($P < 0.05$) (Fig. 4D and E). In the same grade of Baijiu, the total content of alcohol compounds was higher in the north than in the south, including 1-propanol, 3-methyl-1-butanol, and pentanol (Fig. 4D). Previous studies have found that the concentrations of these compounds in northern Nongxiangxing Baijiu are higher than those in the south, making them key components of northern Nongxiangxing Baijiu (Song et al., 2020). In FGB, the proportion of alcoholic compounds was higher in southern Baijiu, while in SGB and PGB, the proportion of alcoholic compounds was higher in northern Baijiu (Fig. 4E). The distribution of ketones was the opposite of that for alcohols (Fig. 4E). Ketones in both regions were primarily composed of 3-

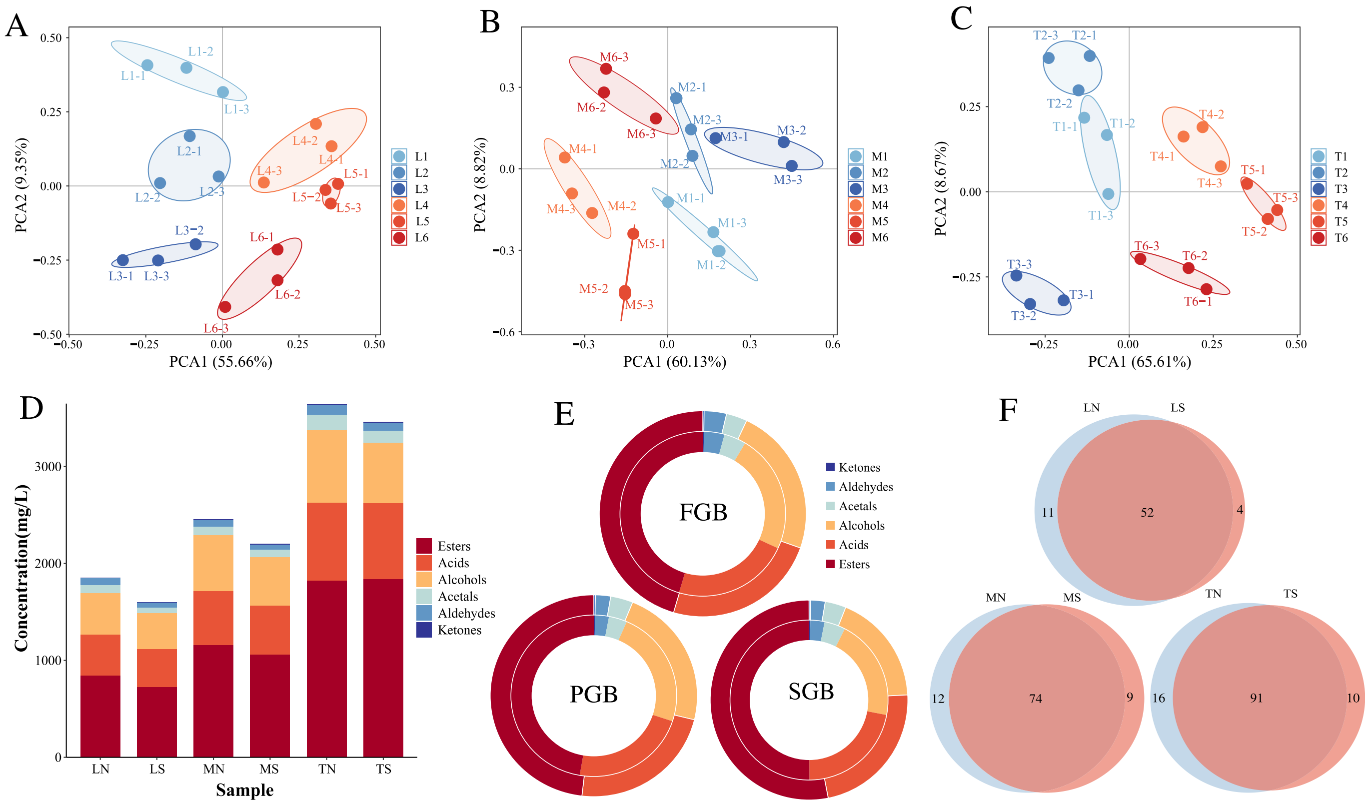

Fig. 4. PCA analysis of FGB (A), PGB (B), and SGB (C); percentage of composition (D), cumulative total content (E) and venn analysis (F) of flavor substances in south and north Qingxiangxiang Baijiu.

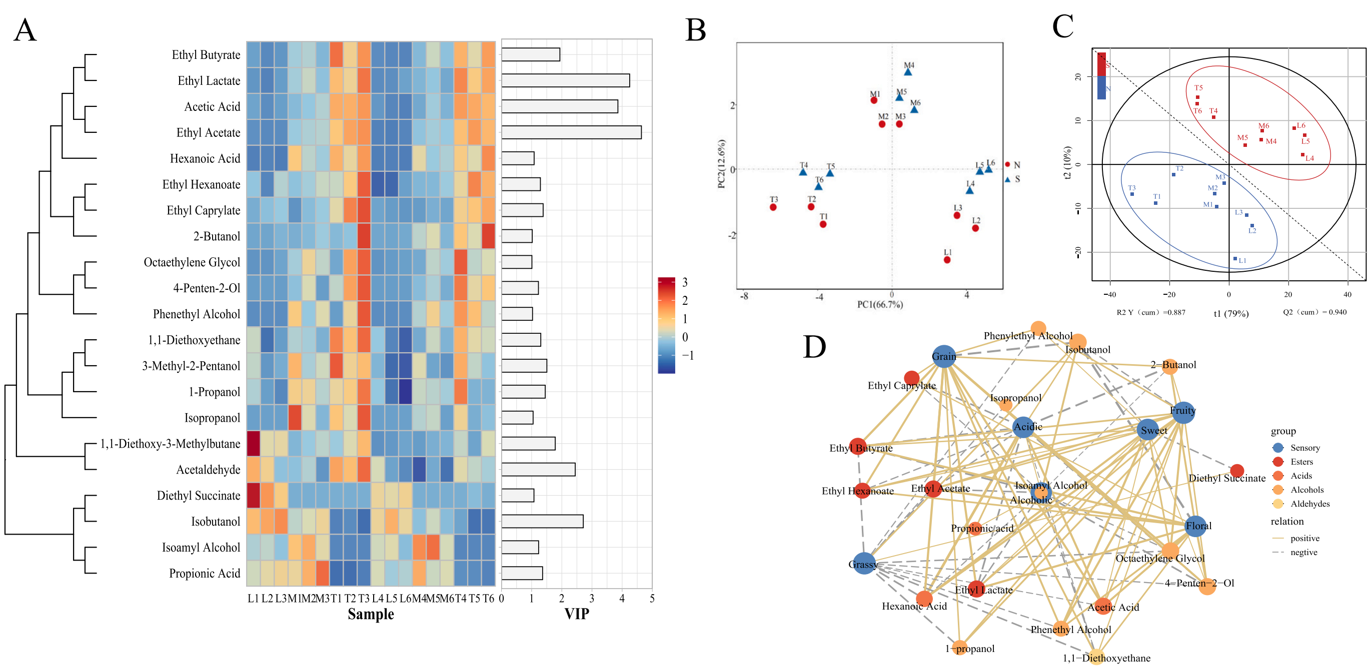

Fig. 5. The clustering heat map of flavor substances based on regional markers (VIP > 1) (A), PCA analysis (B) and PLS-DA analysis (C) were performed based on variable importance in projection (VIP) >1 flavor substances. Correlation network analysis of volatile flavor compounds (VIP > 1) and sensory (D).

hydroxy-2-butanone and 2,3-butanedione, which have low thresholds and often emit scents described as “floral”, “fruity”, and “herbal” (Wang et al., 2024). The concentration of 3-hydroxy-2-butanone is found to be much higher in SQXB than in the north, which confirms our results (Niu et al., 2017). The Venn diagram showed that the diversity of flavor substances was higher in the north than in the south (Fig. 4F). With the increase in grade, the number of different volatile flavor substances in SQXB and NQXB increased from 15 to 26. This also explained why there was an increasing degree of dispersion between SQXB and NQXB with the increase in grade in PCA analysis (Fig. 4A–C).

3.5. Identification of potential regional markers in northern and southern samples

Considering all volatile compounds to distinguish regions of QXB was neither a simple nor an appropriate method (Fig. 3D). Therefore, it was necessary to identify important volatile compounds associated with the QXB region through multivariate analysis. A total of 21 compounds with VIP > 1 were detected, which could potentially serve as regional markers (Fig. 5A). Among them, ethyl hexanoate, 1-propanol, hexanoic acid, acetaldehyde, and 1,1-diethoxyethane were considered as the potential markers causing regional difference among SFB samples (Li et al., 2024). The VIP of ethyl acetate was the highest (VIP = 4.64). It could be observed from the heat map that the content of ethyl acetate was generally higher in the NQXB. Previous studies have found that glutinous sorghum from northeast China and Shanxi Province produces higher levels of ethyl acetate compared to glutinous sorghum from Hunan, Sichuan, or Guizhou Province (Zhang et al., 2023). This could be one of the reasons for the high concentration of ethyl acetate in the northern QXB. In addition, studies have found that the QXB produced by Sichuan sorghum (Lunuo No.8 and Qingkeyang) is pure, sweet, and clean. However, the high content of ethyl lactate in the Baijiu produced by sorghum brewing in northern China leads to a high ratio of ethyl lactate to ethyl hexanoate, resulting in a slightly unpleasant taste (Lai et al., 2024 ; Ma et al., 2024). In this study, the VIP value of ethyl lactate, as a regional marker distinguishing the north from the south, was second only to that of ethyl acetate. This indicates that raw materials have a significant influence on the flavor of SQXB and NQXB. Furthermore, for the same grade of sample, the majority of marker compounds were found to be more concentrated in NQXB, including isobutanol, propionic acid, and 1,1-diethoxy-3-methylbutane (Fig. 5A).

The 18 samples were differentiated between the northern and southern regions based on these 21 markers. The PCA analysis revealed that samples of the same grade tend to cluster together, while there is no significant dispersion observed between samples from the north and south regions. However, a slight trend towards discreteness can be observed within samples of the same grade (Fig. 5B). To obtain more accurate classification results, PLS-DA was employed for further analysis. The samples from the north and south were distributed on both sides of the diagonal in a PLS-DA analysis, with the samples from the north clustered in the lower left and those from the south clustered in the upper right (Fig. 5C). The fitness of the classification model was 88.7 % (R2Y(cum) = 0.887). From the perspective of prediction ability, the model achieved a high accuracy rate of 84.1 % (Q2(cum) = 0.841). This proves that PLS-DA was more effective in regional differentiation of QXB.

3.6. Electronic nose analysis

For different grades of QXB, there was a significant difference in the response values measured by the W1C and W2S sensors ($P < 0.05$) (Fig. 6A–C). W1C and W2S are respectively sensitive to aromatic hydrocarbons and alcoholic compounds (Table S2). As the grade increased, the W1C and W2S response values increased. It was evident that the W2S response value of the NQXB was higher than that of the SQXB in SGB ((Fig. 6C). Analysis of the volatile compounds in the initial portion also revealed that aromatic hydrocarbons and alcoholic compounds exhibited significant differences between the three grades of QXB (Fig. 6A–C), with the electronic nose data once again confirming this conclusion. In order to determine whether the electronic nose can recognize different grades of QXB, the data of different grades of electronic nose were analyzed based on PCA and PLS-DA (Fig. 6D and E). It was found that SGB could be clearly separated from FGB and PGB, while FGB and PGB had some overlap. Through Venn analysis and volcano plot analysis of the flavor substances in the three grade samples, it can be found that the

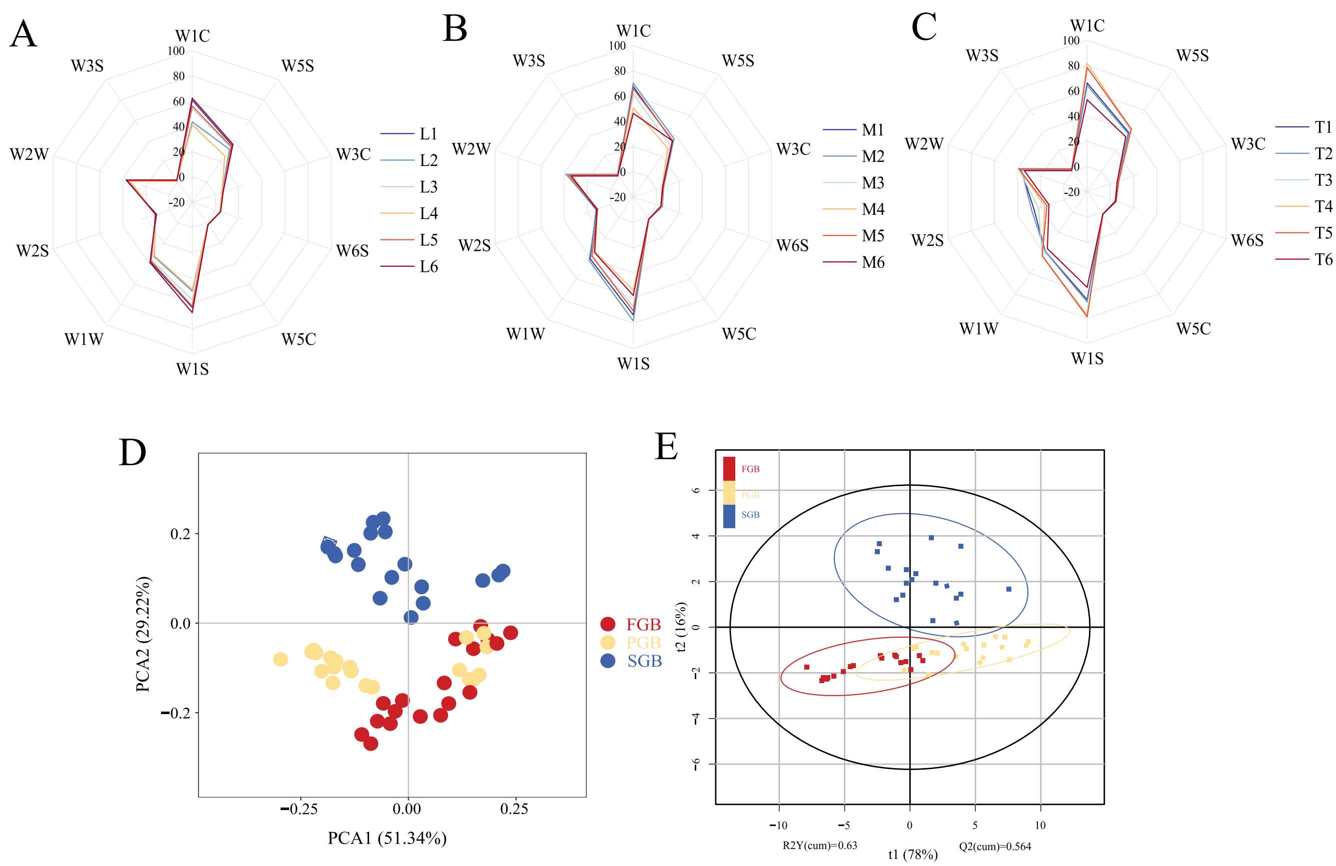

Fig. 6. Electronic nose radar map of FGB (A), PGB (B), and SGB (C). The PCA analysis (D) and PLS-DA analysis (E) of electronic nose.

differences between FGB and PGB in the three grades were relatively small, while the differences between SGB and FGB with PGB were greater (Fig. 2A and Fig. 3A–C). It also indicated that electronic nose data cannot differentiate between different grade QXB. For the same grade of QXB, the W1C and W1S sensors showed a significant response to the sample, while the response intensity of W2W, W5S, and W1W was similar. The response intensity of other sensors was lower (Fig. 6A–C). Through PCA analysis, it was found that the dispersive distance between SQXB and NQXB becomes more obvious as the grade increased (Fig. S1), which was consistent with the results obtained by analyzing volatile compounds (Fig. 4A–C). However, the distinction between SQXB and NQXB by electronic nose data was generally less effective than that by volatile compounds.

3.7. Application of metal elements in the regional differentiation of QXB

According to studies on metallic element contamination in wine, the sources of metal elements in Baijiu include raw materials, food additives, packaging materials, treatment processes for raw materials, winemaking equipment, aging/storage containers, winemaking water and post-winemaking treatment processes (Ibanez et al., 2008). Due to differences in raw materials, processes, and production environments of Baijiu in different regions, the composition of metal elements in Baijiu with the same flavor varies. The levels of Al, Fe, Mg, K, and Cu were higher in pottery jars compared to stainless steel tanks. The increased presence of these elements in pottery jars enhanced the sensory quality of Jiangxiangxing Baijiu (Huang et al., 2020; Wei et al., 2023). Furthermore, the metal content is associated with the production environment, making it a useful indicator for distinguishing alcoholic beverages from different regions. Previous studies have found that the content of certain metal elements remains stable throughout the entire production process of German Baijiu and Portuguese red wine, which can be used as geographical indications (Gómez et al., 2004; Ibanez et al., 2008). Therefore, exploring the composition of metal elements in Baijiu from different regions can not only provide another method for regional identification of QXB but also study the influence of metal elements on the flavor of Baijiu in different regions and further analyze the potential causes of flavor differences in QXB across different regions.

A total of 21 metals were detected in the samples by ICP-MS. According to the observation in Fig. 7A, significant differences were found in the composition of metal elements among different grades of QXB, as well as within the same grade of QXB from the south and the north. The

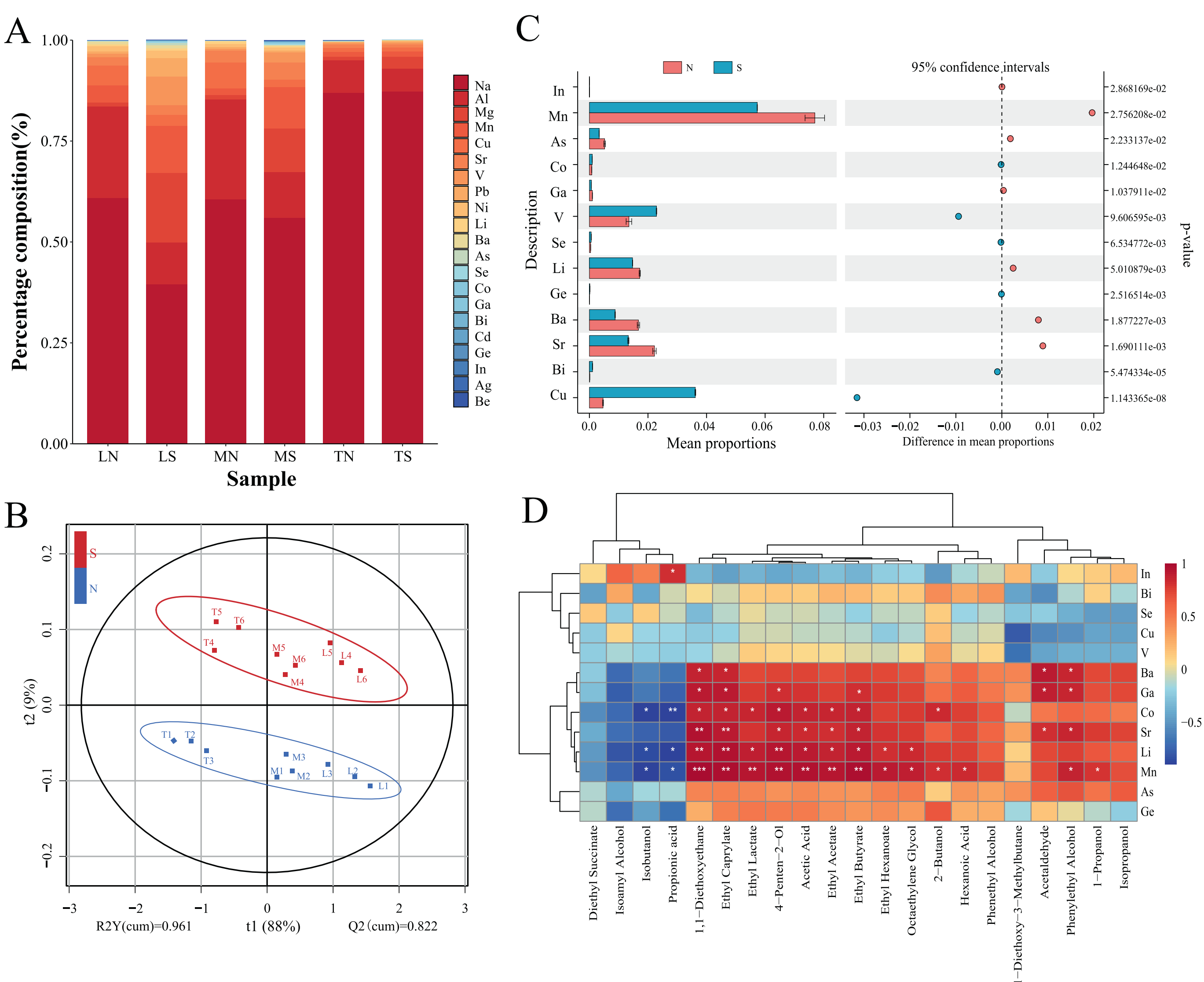

Fig. 7. The percentage composition (A) and PLS-DA analysis (B) of metal ions detected in south and north Qingxiangxiang Baijiu. The statistical analysis of taxonomic and functional profiles (STAMP) analysis between south and north Qingxiangxiang Baijiu (C). Spearman correlation analysis between 13 different metals and 21 regional markers (VIP > 1) (D).

proportion of Na in all samples was the highest, which is consistent with previous studies (Wei et al., 2023). The reason for this may be that the Na element is mainly present in the fermented mash in the form of ions, which can be allocated in a higher proportion to the liquid products produced by fermentation, such as water and ethanol, and can be transferred to a greater extent to the bottom pot water or the distillate (Ibanez et al., 2008; Wei et al., 2023). Previous studies have shown that adding Na to Baijiu can promote the evaporation of certain esters, such as ethyl pentanoate, ethyl lactate, and ethyl phenylhexanoate (Wei et al., 2023). Based on these 21 metals, 18 samples were differentiated into northern and southern regions. Through PLS-DA analysis, samples from the north and south were distributed on both sides of the x-axis, with samples from the north clustered below and samples from the south clustered above (Fig. 7B). The classification model fitness of model was 96.1 % (R2Y(cum) = 0.961). From the prediction ability point of view, the model was as high as 82.2 % (Q2(cum) = 0.822). It had been proven that the metal elements in Baijiu can be utilized for regional differentiation of QXB.

The metal elements with significant differences between north and south Baijiu were selected through stamp analysis. There were 13 types of metal elements, including Cu, Bi, Sr, Ba, and so on (Fig. 7C). Previous studies have found that the Sr element may originate from certain mineral water, and it has been reported that Sr isotopes are used to trace the origin of food (Yin et al., 2020). Therefore, it was speculated that the Sr element in Baijiu was probably introduced from the brewing water. Due to the difference in water used for production between the two regions, there was a significant disparity in Sr content ($P < 0.05$). The Cu and Ba in Baijiu may be introduced through the raw materials used in brewing, so the significant difference of Cu and Ba in QXB between the two regions may be caused by variations in brewing raw materials (Li et al., 2016). Furthermore, previous studies have revealed significant differences in Se and Cu content between Shandong Baijiu and Sichuan Baijiu, which can be used to distinguish the Baijiu from these two regions (Song et al., 2018). In this study, there were significant differences in Se and Cu between SQXB and NQXB, which is consistent with previous research findings. Ni, Al, Pb, and Sn may originate from the connecting tube, condensing tube, and receiving vessel that come into direct contact with the samples during distillation. There was no significant difference between SQXB and NQXB (Li et al., 2016). To further investigate the impact of metal elements on flavor, Spearman correlation analysis was conducted between 13 different metals and 21 potential regional markers with VIP > 1 in both southern and northern QXB (Fig. 7D). Except for isobutanol and propionic acid, Mn exhibited a significant positive correlation with the remaining acids and most alcohols ($P < 0.05$). Except for diethyl succinate, Co, Li, and Mn showed a significant positive correlation with esters ($P < 0.05$). Ba, Ga, Co, Sr, Li, and Mn exhibited a significant positive correlation with 1,1-Diethoxyethane ($P < 0.05$), but did not show any significant correlation with 1,1-Diethoxy-3-Methylbutane. Ba, Ga, and Sr showed a positive correlation with acetaldehyde ($P < 0.05$). These results indicate that the metal ions present in Baijiu can influence its aroma and taste during production and sales of QXB.

4. Conclusion

As the grade increased, the floral and sweet fragrances of QXB become more pronounced, while the acidic taste decreased. The main differences among the three grades of Baijiu were primarily concentrated in esters and alcohols. Through a comparison of QXB from the south and north, it was found that the floral fragrance of the SQXB was stronger, while the acidic taste of the NQXB was more pronounced. The difference in volatile flavor substances between SQXB and NQXB increased with the increase in grade, leading to a greater degree of dispersion between them. A total of 23 grade markers and 21 regional markers with VIP > 1 were identified, and ethyl acetate had the highest score among both types. PLS-DA was more suitable for the classification of grade and region in QXB. The electronic nose had poor effect on differentiating the grades and regions of QXB. A total of 21 metals were detected in the QXB by ICP-MS, among which 19 metals, including Cu, Bi, and Sr, showed significant differences ($P < 0.05$), and these metal elements could be used as indicators to distinguish QXB in various regions. In general, this study can help people better understand the flavors of different grades and regions of QXB, and provide a theoretical basis for regulating the aroma and flavor profiles of QXB.

Ethical Statement

The appropriate protocols were utilized to protect the rights and privacy of all participants during the execution of the research and were approved by the Ethics Committee of Xihua University. All participants provided verbal consent and had the option to withdraw from the study at any time.

CRediT authorship contribution statement

Qiao Huang: Writing – review & editing, Writing – original draft, Methodology, Data curation. **Xianping Qiu:** Validation, Investigation, Conceptualization. **Tongwei Guan:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization. **Jianshen Yu:** Visualization, Software, Methodology. **Yichen Mao:** Visualization, Software, Methodology. **Yu Li:** Visualization, Software, Methodology. **Yuan Rao:** Visualization, Resources, Investigation. **Hongguang Shang:** Visualization, Resources, Investigation. **Yanhui Zhao:** Visualization, Resources, Investigation.

Declaration of competing interest

The authors state that they do not have any known financial interests or personal relationships that could be perceived as influencing the work presented in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2024.102141.

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