



## Unveiling innovation in aroma attribute evaluation of Niulanshan Baijiu: An advanced exploration of two different processing methods via food sensory omics and penalty analysis

Zhen Wang<sup>a,b,1</sup>, Wenjun Hao<sup>c,1</sup>, Jinwang Wei<sup>c</sup>, Mingquan Huang<sup>a,\*</sup>, Xinan Zeng<sup>b,d</sup>, Ying Wang<sup>c,\*</sup>, Jihong Wu<sup>a</sup>, Boru Chen<sup>b,d</sup>

<sup>a</sup> Key Laboratory of Brewing Molecular Engineering of China Light Industry, Beijing Technology & Business University (BTBU), Beijing 100048, China

<sup>b</sup> School of Food Science and Engineering, South China University of Technology (SCUT), Guangzhou 510640, China

<sup>c</sup> Niulanshan Distillery, Beijing Shunxin Agriculture Co. Ltd, Beijing 101301, China

<sup>d</sup> Guangdong Key Laboratory of Food Intelligent Manufacturing, Foshan University, Foshan, Guangdong 528225, China

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### ABSTRACT

Niulanshan Baijiu (NLS), a notable variety of Baijiu known for its light flavor and extensive historical legacy, was subjected to a comparative analysis using two different processes: Hunzheng Xucha (HX) and Qingzheng Qingcha (QQ). The study combined sensory-oriented flavor analysis and penalty analysis to assess the differences between the two processes. Aroma compounds in NLS were extracted using liquid–liquid extraction and headspace solid phase microextraction. Gas chromatography–olfactometry–mass spectrometry was employed to identify 46 aroma-active compounds, including the first-time discovery of ethyl isohexanoate and 2,4-nonadienal in NLS. Quantification of 35 compounds with odor activity value (OAV)  $\geq 1$  was achieved using internal standard curve methods. Sensory assessments by a cohort of 111 participants highlighted the preference for HX-NLS in terms of flavor, while QQ-NLS exhibited a sour-Chen aroma that required improvement. The study further revealed the significant impact of acetic acid, butyric acid, hexanoic acid, octanoic acid, and 3-methylbutanal on the sour-Chen aroma in liquor.

### 1. Introduction

Baijiu, an eminent alcoholic beverage with a historical lineage spanning over two millennia, has earned the distinction of being hailed as China's national liquor, and is revered as one among the top four globally recognized distilled spirits, alongside brandy, vodka, and whisky (Liu & Sun, 2018). Notably, Baijiu exhibits a diverse array of flavor profiles, leading to its classification into four primary flavor-style categories and eight secondary flavor-style categories. The four primary flavor-style Baijiu categories comprise of light (mild), Nong (strong), Jiang (soy sauce), and Mi (rice) flavor Baijiu. Light flavor Baijiu, acknowledged for its unblemished and refined olfactory and gustatory characteristics, emanates an agreeable bouquet of floral and fruity notes, accentuated by a subtle hint of vinegary Chen-aroma and a gentle sweetness akin to yogurt (GB/T 15109-2021; GB/T 17204-2021). Among the esteemed selection of light flavor Baijiu, Niulanshan Baijiu (NLS), meticulously crafted in Beijing, has garnered considerable favor

among Chinese connoisseurs, owing to its timeless appeal.

NLS is usually brewed using the traditional Qingzheng Qingcha (QQ) process, which means that the grain and auxiliary materials are distilled separately, then the distilled grain and auxiliary materials are mixed together and fermented in Digang (a pottery jar buried underground, a kind of fermentation vessel) after the addition of Daqu. After the fermentation is completed, the first fermented Jiupei (Jiang et al., 2019) are distilled to gain the liquor, named crude Baijiu. When the distillation is over, the distilled Jiupei is added Jiuqu again for the second fermentation and distillation (Wang et al., 2022). With the continuous improvement of the process and the continuous expansion of product demand, NLS is also brewed using another traditional process of Hunzheng Xucha (HX) now. The main difference between the processes of HX and QQ is that HX is fermented in Jiaochi (a pit dug in the ground, a kind of fermentation container), and the added new grain is steamed together with the distilled Jiupei from the previous round production, and the lees are always partially recycled. The specific production

\* Corresponding authors.

<sup>1</sup> These authors are equal to this work.

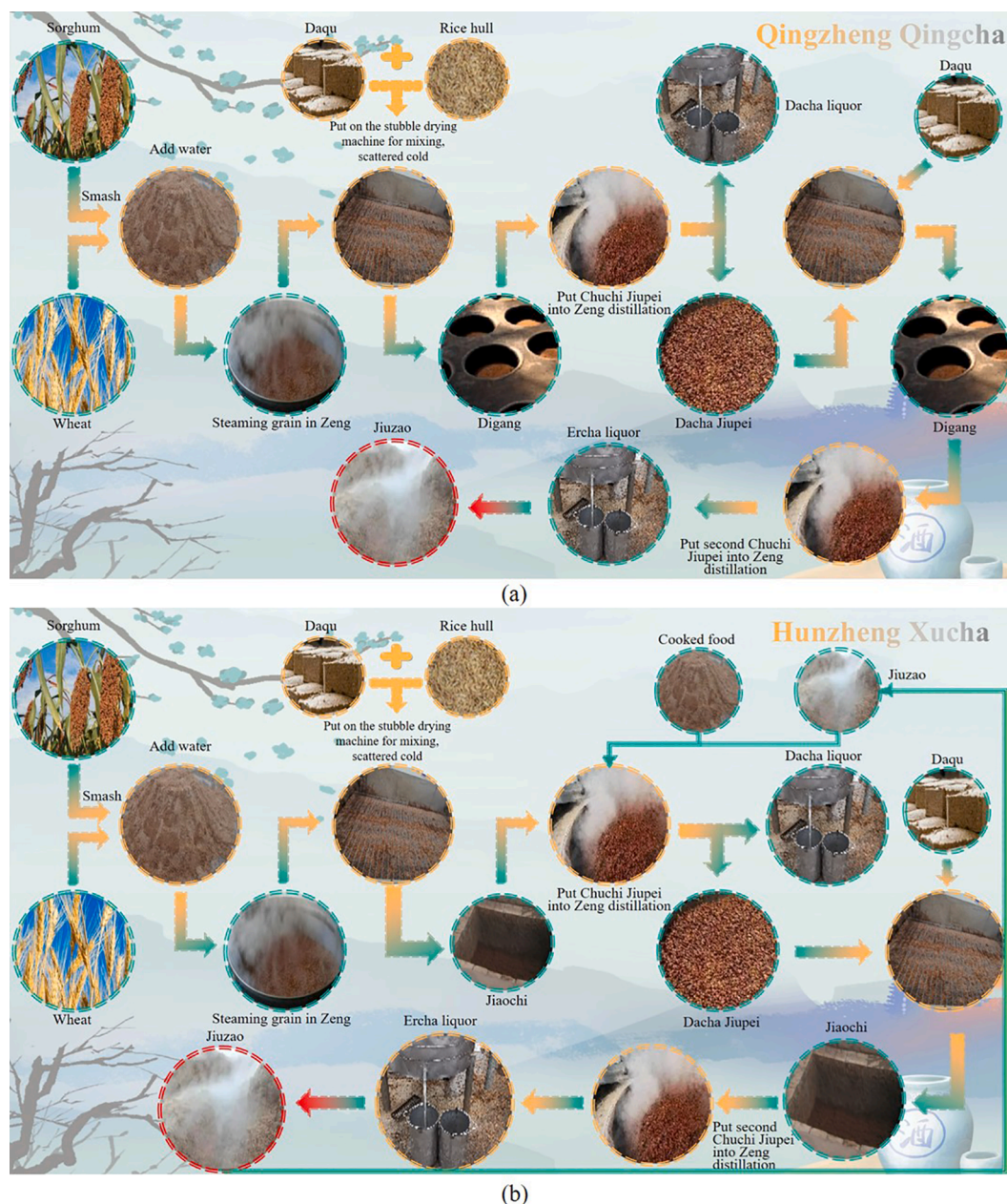


Fig. 1. Flow chart of Niulanshan Baijiu production process. (a) Qingzheng Qingcha of Niulanshan Baijiu brewing process; (b) Hunzheng Xucha of Niulanshan Baijiu brewing process.

process is shown in Fig. 1.

The assessment of Baijiu's aroma holds significant importance in determining both product quality and consumer preference. Advancements in gas chromatography (GC) technology have enabled comprehensive investigations into the aroma components of Baijiu, furnishing a solid foundation for Baijiu blending techniques (Liu & Sun, 2018; Xu et al., 2021; Lv et al., 2022). Employing flavor sensory omics, our research team successfully identified 34 aroma compounds with odorant activity values (OAV) exceeding 1 in QQ-NLS (Wang et al., 2022). In a separate study by Wang et al. (2008), a total of 101 aroma compounds were identified in NLS through the application of solid-phase micro-extraction (SPME), liquid-liquid extraction (LLE), and gas chromatography-mass spectrometer (GC-MS) methodologies. However, to date, there remains a dearth of literature pertaining to the characterization of aroma compounds in HX-NLS, as well as the absence of consumer aroma evaluation comparisons encompassing two distinct NLS. These studies, encompassing NLS and other Baijiu, currently reside

within the realm of theoretical inquiry, necessitating an enhanced integration of theory and practical application to comprehend consumer preferences. Therefore, this study employs penalty analysis to evaluate the aroma attributes of two NLS products produced using distinct brewing processes, aiming to bridge the gap between theoretical research and practical implementation.

The penalty analysis is known as the “killer app for food sensory quality diagnosis” (Pagès et al., 2014). It is a consumer preference evaluation method that provides valuable insights into consumer preferences and decision-making processes. It aims to determine the relative importance of different product attributes or features and the corresponding penalties associated with their absence or inadequacy. By quantifying these penalties, researchers can gain a deeper understanding of consumer behavior and preferences (Narayanan et al., 2014). Narayanan et al. (2014) tested the appropriate concentrations of three processed stevia sweeteners/supplements in plain low-fat yogurt flavored with natural vanilla by penalty analysis. They found that it was

important to carefully select the type and concentration of Stevia, and optimize the culture and fermentation conditions of yogurt, prior to market. Using penalty analysis, Cong et al. (2017) successfully debugged yogurts that met the tastes of consumers. Morais et al. (2014) used penalty analysis to test the acceptability of added sucrose in chocolate dairy products and found that the ideal concentration of sucrose was 8%, which helped to improve product quality. In the case of Baijiu, the penalty analysis can make producers better understand consumers' preferences for Baijiu or its specific flavor attribute, which in turn helps Baijiu producers to make purposeful blending and accurately optimize the flavor intensity of products. The specific operation steps of penalty analysis are shown in Fig. S1. This method integrates the overall product preference scale test and a sensory attribute aptness scale test to carry out penalty analysis (Peryam & Pilgrim, 1957; Zhi et al., 2016). They are the hedonic scale and just-about-right (JAR) scales, respectively. The hedonic scale is a balanced bipolar scale centered around "neither like nor dislike", and mark evenly on both sides with words representing different degrees of liking (Juyun, 2011; Lim & Fujimaru, 2010; Villanueva & Da Silva, 2009). The most common hedonic scale is the nine-point hedonic scale ranging from 1 (dislike extremely) to 9 (like extremely) (Everitt, 2009). JAR is a direct way to obtain consumer feedback, seeking whether the flavor and sensory quality of products are just right (Popper, 2005; Rothman, 2007a; Rothman, 2007b). The 9-point hedonic scale and the 5-point JAR scale are shown in Table S1 (Lawless & Heymann, 2010; Rothman, 2007a; Rothman, 2007b). The penalty analysis of a large amount of data is used as a way to combine the aroma attributes of Baijiu with consumer testing. In recent years, most of the researches are focused on theoretical analysis of Baijiu flavor, but no research has been reported on the flavor of NLS with HX. No research has been done to combine penalty analysis with food sensory omics to regulate the flavor of Baijiu in a targeted manner to meet public demand, either.

This study presents a novel approach by employing food sensory omics to conduct a comparative and analytical assessment of the flavor constituents in NSL, which is brewed using the HX and QQ processes. Moreover, for the first time, a consumer-oriented analysis utilizing penalty analysis is utilized to identify Baijiu that exhibit higher popularity among the general public. Furthermore, the study successfully integrates food sensory omics with penalty analysis techniques to enhance the flavor characteristics of Baijiu. The specific procedural details are outlined as follows: 1) Aroma-active compounds were isolated and identified from NLS by LLE and headspace solid-phase microextraction (HS-SPME) combined with gas chromatography-olfactometry-mass spectrometer (GC-O-MS); 2) The important aroma active compounds were quantitated using internal standard curve methods combined with GC-MS and gas chromatography-flame ionization detector (GC-FID) and their OAV were calculated; 3) With penalty analysis, the specific differences between QQ-NLS and HX-NLS in preferences and aroma attributes were determined. This study aims to assess and compare the distinct consumer preferences and aroma attributes associated with QQ-NSL and HX-NSL, employing penalty analysis as a methodological approach. The findings of this research hold considerable significance in the realms of consumer preference analysis and quality enhancement of Baijiu.

## 2. Materials and methods

### 2.1. Liquors

Two high quality NLS with the same grade, including QQ-NLS (crude liquor, 60% ethanol by volume), HX-NLS (crude liquor, 60% ethanol by volume), produced in December 2013, were from Niulanshan Distillery, Shunyi District, Beijing, China. All liquors (500 mL for each) were stored at 4 °C until analysis.

### 2.2. Chemicals

All chemicals used for analysis were GC purities with at least 97%. The specific information was listed in Table S2. Linear retention indices (RIs) were determined using a C<sub>7</sub>-C<sub>27</sub> *n*-alkane paraffin mixture (Sigma-Aldrich, Shanghai, China). Dichloromethane (HPLC), ethanol (HPLC), sodium carbonate (AR), anhydrous sodium sulfate (AR), sodium carbonate (AR), and hydrochloric acid (AR) were purchased from Sino-pharm Chemical Reagent Co., Ltd. (Beijing, China).

### 2.3. Isolation of the Volatiles

#### 2.3.1. LLE

The extraction method of LLE was referenced from Wang et al. (2022), the samples of QQ-NLS and HX-NLS (25.0 mL) were diluted with saturated NaCl solution until the volume fraction of ethanol was 10%, and then the diluted solutions were extracted with methylene chloride for three times (50.0 mL/ time). The combined organic phases were further separated into acidic and neutral/basic components (AFs and NBFs) as follows: the organic phases were extracted with 0.5 mol/L Na<sub>2</sub>CO<sub>3</sub> solution for three times (50.0 mL/ time), and then washed with NaCl solution (about 70.0 mL) again to obtain NBFs. The combined Na<sub>2</sub>CO<sub>3</sub> and NaCl solutions were adjusted to pH 2.0 with hydrochloric acid (4.0 mol/L), and then extracted with dichloromethane for 3 times (70.0 mL/ time) to obtain AFs. Both NBFs and AFs were added with dried anhydrous sodium sulfate for 12 h. After filtration, the filtrates were concentrated to 0.5 mL by Vigreux column (50 cm × 1 cm i.d.) and nitrogen purge (99.999%). These concentrated fractions were stored at - 20 °C prior to GC-O-MS analysis.

#### 2.3.2. HS-SPME

The samples (QQ-NLS, HX-NLS, 10 mL) were diluted with saturated NaCl solution until the volume fraction of ethanol was 10%, and then 8.0 mL of diluted solutions was placed in 20 mL headspace bottles for the following analysis: The samples were preheated at 45 °C for 30 min, then the solid phase extraction fiber (DVB/CAR/PDMS, film thickness 75 μm) was inserted into the headspace bottles, and adsorbed at 45 °C for 40 min. After adsorption, the fiber was inserted into the inlet of GC-MS quickly and desorbed at 250 °C for 5 min. The flow rate of helium (99.999%) is 1.5 mL/min.

### 2.4. Identification of Aroma-Active compounds

The aroma-active compounds were analyzed using GC-MS (Agilent 7890B GC / 5977A MSD, Agilent) with an olfactory detection port (ODP3, Gerstel, Germany), coupled with a DB-WAX (60 m × 250 μm i.d., 0.25 μm film thickness, Thermo Fisher Scientific, USA) or HP-5MS capillary column (30 m × 250 μm i.d., 0.25 μm film thickness, Thermo Fisher Scientific, USA). The carrier gas was helium (99.999%) with a constant flow rate of 1.5 mL/min, and the inlet temperature was 250 °C. The heating programs of the two capillary columns referred from Wang et al. (2022) as follows: the oven temperature was initially kept at 40 °C, then ramped to 50 °C at 10 °C/min and held for 5 min, then increased to 80 °C at 3 °C/min and held for 5 min, finally raised to 230 °C at 5 °C/min and held for 15 min. The effluent of the capillary column was split at a ratio of 1:1 v/v between mass spectrometer (MS) and ODP3. Mass spectra were generated in EI mode at 70 eV, with a scan range of *m/z* 30–450. The ion source temperature was 230 °C, and the transmission line temperature was 250 °C. The solvent delay was 8–10 min. The analysis conditions for HS-SPME were the same as described above, and all the samples were injected in the splitless mode. The mixture of *n*-alkanes (C<sub>7</sub> – C<sub>27</sub>) was used to determine RIs of compounds on two chromatographic columns above. Qualitative analysis was performed using NIST library 2020 mass spectrometry and the self-built database of Baijiu flavor compounds (Beijing Technology and Business University). The RI and aroma characteristics of the scented compounds



**Table 1**  
Aroma-active compounds identified by GC-O-MS in HX-NLS and QQ-NLS <sup>a</sup>

No.	Compounds	Pretreatment <sup>a</sup>	Odor descriptor	Osme values		Fraction <sup>b</sup>		Identification <sup>c</sup>	RI <sup>c</sup> DB- WAX	HP- SMS
				HX- NLS	QQ- NLS	HX-NLS	QQ- NLS			
1	ethyl acetate	LLE, HS-SPME	fruit, sweet	4/5	4.1/4.5	NBF/ AF	NBF/ AF	RI,S,Aroma, MS	865	610
2	3-methylbutanal	LLE, HS-SPME	aldehydic, fatty	5/4	5/5	NBF/ AF	NBF/ AF	RI,S,Aroma, MS	906	649
3	ethyl isobutyrate	LLE, HS-SPME	sweet	3.5	3.1	NBF	NBF	RI,S,Aroma, MS	975	751
4	ethyl acrylate	LLE	plastic	3	–	NBF	–	RI,S,Aroma, MS	991	628
5	ethyl butyrate	LLE, HS-SPME	fruit	3	3	NBF	NBF	RI,S,Aroma, MS	1039	805
6	ethyl 2-methylbutyrate	LLE, HS-SPME	fruit	3	3	NBF	NBF	RI,S,Aroma, MS	1055	850
7	ethyl isovalerate	LLE, HS-SPME	fruit, sweet	3	2	NBF	NBF	RI,S,Aroma, MS	1070	850
8	1,1-diethoxy-3-methylbutane	LLE, HS-SPME	greasy	1	1	NBF	NBF	RI,S,Aroma, MS	1083	–
9	2-methyl-1-propanol	LLE, HS-SPME	bitter	2/1	1/2	NBF/ AF	NBF/ AF	RI,S,Aroma, MS	1091	630
10	ethyl valerate	LLE	apple	3.5	1	NBF	NBF	RI,S,Aroma, MS	1132	1080
11	1-butanol	LLE	banana	3	–	NBF	–	RI,S,Aroma, MS	1137	670
12	3-methyl-1-butanol	LLE	spicy	4/2	4.5/3.5	NBF/ AF	NBF/ AF	RI,S,Aroma, MS	1208	740
13	ethyl hexanoate	LLE, HS-SPME	sweet	3	3	NBF	NBF	RI,S,Aroma, MS	1236	999
14	ethyl lactate	LLE	fruit	1	1	NBF	NBF	RI,S,Aroma, MS	1342	830
15	2-hydroxy-3-methylbutanoic acid ethyl ester	LLE	fruit	2.5	3	NBF	NBF	RI,Aroma,MS	1423	–
16	ethyl caprylate	LLE, HS-SPME	fruit	3	4	NBF	NBF	RI,S,Aroma, MS	1442	1193
17	1-heptanol	LLE	musty	3	–	NBF	–	RI,S,Aroma, MS	1449	973
18	ethyl nonanoate	LLE	rose	3.5	3.5	NBF	NBF	RI,S,Aroma, MS	1535	1295
19	ethyl 2-hydroxy-4-methylvalerate	LLE, HS-SPME	fresh	3.5	3	NBF	NBF	RI,S,Aroma, MS	1542	1078
20	ethyl benzoate	LLE, HS-SPME	fruit	5	2	NBF	NBF	RI,S,Aroma, MS	1672	1170
21	phenethyl acetate	LLE, HS-SPME	rose	4	3.5	NBF	NBF	RI,S,Aroma, MS	1820	1250
22	$\beta$ -damascenone	LLE, HS-SPME	sweet	4.5/4	4.5	NBF/ AF	NBF	RI,S,Aroma, MS	1834	–
23	geosmin	LLE, HS-SPME	soil	5	5	NBF	NBF	RI,S,Aroma, MS	1855	1400
24	phenethyl alcohol	LLE	floral	4	3.1	NBF	NBF	RI,S,Aroma, MS	1918	1110
25	$\gamma$ -nonanolactone	LLE	coconut, cream	4	–	NBF	–	RI,S,Aroma, MS	2046	1360
26	1-propanol	LLE	alcohol	1	2	AF	AF	RI,S,Aroma, MS	1040	570
27	acetic acid	LLE	sour	4.5	4	AF	AF	RI,S,Aroma, MS	1410	610
28	butyric acid	LLE	sharp	3	–	AF	–	RI,S,Aroma, MS	1620	790
29	isovaleric acid	LLE	sour	5	5	AF	AF	RI,S,Aroma, MS	1685	890
30	hexanoic acid	LLE	sweat	3	–	AF	–	RI,S,Aroma, MS	1850	975
31	octanoic acid	LLE	waxy	3	–	AF	–	RI,S,Aroma, MS	2060	1190
32	2-methyl-1-butanol	LLE	ethereal	3	–	NBF	–	RI,S,Aroma, MS	1200	730
33	1-hexanol	LLE	fusel oil	3	–	NBF	–	RI,S,Aroma, MS	1350	860
34	phenylacetaldehyde	LLE	sweet	3	–	NBF	–	RI,S,Aroma, MS	1655	1450
35	1,1,3-triethoxypropane	LLE	stuffy	2	–	NBF	–	RI,S,Aroma, MS	1295	–
36	diethyl succinate	LLE	fruit	2	2	NBF	NBF	RI,S,Aroma, MS	1670	1190

(continued on next page)

Table 1 (continued)

No.	Compounds	Pretreatment <sup>a</sup>	Odor descriptor	Osme values		Fraction <sup>b</sup>		Identification <sup>c</sup>	RI <sup>c</sup>	
				HX-NLS	QQ-NLS	HX-NLS	QQ-NLS		DB-WAX	HP-5MS
37	ethyl isohexanoate	LLE, HS-SPME	fruit	–	2	–	NBF	RI,S,Aroma, MS	1195	960
38	acetoin	LLE	sweet	–	1	–	NBF	RI,S,Aroma, MS	1288	720
39	trimethyl-pyrazine	LLE	nutty	–	3	–	NBF	RI,S,Aroma, MS	1411	1000
40	furfural	LLE	woody	–	2/1	–	NBF/AF	RI,S,Aroma, MS	1470	841
41	2,4-nonadienal	LLE	cucumber	–	4	–	NBF	RI,S,Aroma, MS	1690	1200
42	ethyl caprate	LLE	fruit	–	3.5	–	NBF	RI,S,Aroma, MS	1640	1390
43	ethyl phenylacetate	LLE	sweet	–	2	–	NBF	RI,S,Aroma, MS	1787	1240
44	ethyl 3-phenylpropionate	LLE	rose	–	3.5	–	NBF	RI,S,Aroma, MS	1901	1330
45	ethyl myristate	LLE	sweet	–	3	–	NBF	RI,S,Aroma, MS	2057	1791
46	tetramethylpyrazine	LLE	nutty	–	2	–	NBF	RI,S,Aroma, MS	1461	1081
	unknown	LLE	paint	3	–	–	NBF	–	–	–
	unknown	LLE	sharp	–	4	–	NBF	–	–	–

<sup>a</sup> Methods of sample pretreatment; LLE, liquid–liquid extraction; HS-SPME, headspace solid-phase microextraction; HX-NLS (Hunzheng Xucha Niulanshan Baijiu); QQ-NLS (Qingzheng Qingcha Niulanshan Baijiu).

<sup>b</sup> Components from which the aroma compounds are derived detected by GC–O–MS. AF, acidic fraction; NBF, neutral and basic fraction.

<sup>c</sup> MS, aroma compounds were identified by MS; RI, linear retention indices, the retention indices of the compounds on DB-WAX and DB-5MS columns were calculated based on the peak time of *n*-alkanes (C<sub>7</sub>–C<sub>27</sub>) on the instrument; S, compounds were identified by pure standards; aroma, the odors were compared with the standard compounds by GC–O–MS.

were compared with the standard compounds for identification. The Osme value was the average aroma intensity of each compound smelled by five sensory evaluators, and recorded as 1 (weak), 2–3 (moderate), and 4–5 (strong).

## 2.5. Quantitation analysis of Aroma-Active compounds

### 2.5.1. DI combined with GC–MS/GC–FID

The high content compounds 1, 2, 5, 9–14, 16, 19, 24, 26, 27, 32, 33, 38 and 40 in Table 2 were quantified by GC–FID according to the previous quantitative methods (Wang et al., 2022). To reduce the impact of moisture, chromatographic ethanol was used to blend the volume fraction of ethanol in the sample to 80%. IS1 and IS2 were spiked to the treated samples, and their final concentrations were 30.78 mg/L and 20.22 mg/L, respectively. The mixed samples were analyzed using GC–FID equipped with a DB–FFAP column (60 m × 250 μm i.d., 0.25 μm film thickness; J&W Scientific, USA). The samples (1.0 μL) were injected into the GC inlet with a split mode (20:1). The temperature procedure was as follows: the initial temperature was 40°C, then rose to 50°C at 10 °C/min and kept for 2 min, then rose to 80°C at 3 °C/min and kept for 2 min, and finally rose to 245°C at 5 °C/min and kept for 3 min. The inlet temperature was 250 °C, the carrier gas was nitrogen (99.999%), and the constant flow rate was 1.5 mL/min. The standard compounds were first dissolved in 80% (v/v) ethanol aqueous solution prepared by ethanol (HPLC) and ultra-pure water to obtain the mixed standard reserve solution. Then, the mixed solution was diluted to a series of concentration gradients (3000, 1500, 750, 375, 187.5, 93.75, 46.88, 23.44, 11.72 and 5.86 mg/L). After mixing the standard solutions (1.0 mL) with the internal standard solution (IS1, IS2), these solutions were analyzed using GC–FID. All experiments were repeated three times.

These compounds 8, 18, 21, 25, 34, 42, 43, 45 in Table 2 were quantified by direct injection (DI) with GC–MS. Similarly, in order to reduce the influence of the matrix, the ethanol content in the sample was raised to 80% (v/v) with ethanol (HPLC). Then, the internal standards (IS1, IS2, IS3) were added to them, and all their final concentrations were 100 mg/L. Subsequently, the mixed samples were analyzed using

GC–MS with selective ion monitoring (SIM) mode, and the split ratio of the injector was 20:1. The column was a DB–FFAP capillary column (60 m × 250 μm i.d., 0.25 μm film thickness; J&W Scientific, USA). The heating procedure was the same as the part of **Identification of Aroma-active compounds**. Standard solutions with a series of concentration gradients (2000, 1000, 500, 250, 125, 62.5, 31.25, 15.63, 7.81, and 3.91 mg/L) also were prepared using 80% (v/v) alcohol aqueous solution. The internal standards were also added and their final concentrations were the same as the above samples. Finally, these standard solutions were analyzed using GC–MS as above. Each experiment was repeated three times.

### 2.5.2. LLE combined with GC–MS

According to the preliminary experimental results, these compounds numbered 3, 4, 6, 7, 15, 17, 20, 22, 23, 28–31, 35–37, 39, 41, 44, 46 in Table 2 were quantitated using LLE with GC–MS (Shi et al., 2021). After the ethanol content in the samples were raised to 80% (v/v) and three internal standards (IS1, IS2, IS3) were added into the diluted samples, the mixed sample solutions were pretreated as **Isolation of the Volatiles (LLE)**. However, the extracts needed not be divided into AFs and NBFs. The final concentrations of three internal standards were 10.0 mg/L. A series of gradient standard solutions of these compounds (50, 25, 12.5, 6.25, 3.125, 1.56, 0.78, 0.39, 0.195 and 0.0975 mg/L), with 10.0 mg/L of internal standards, were also prepared using 80% (v/v) alcohol aqueous solution. After the mixed standard solutions were treated as the samples, all the concentrated extracts were analyzed by GC–MS with SIM as above. Finally, these compounds were quantified using the internal standard curve method. Their relevant parameters of ion fragments and standard curves are shown in Table S3.

## 2.6. Penalty analysis

### 2.6.1. Aroma evaluation

Based on the training method of Wang et al. (2021). Thirty panelists (15 males and 15 females, aged 22–30 years) with experience in olfactory experiments and quantitative descriptive analysis were recruited

**Table 2**  
Quantitation of aroma-active compounds in HX-NLS and QQ-NLS by GC-FID/GC-MS <sup>c</sup>.

No.	Compounds	Pretreatment <sup>d</sup>	Detector <sup>e</sup>	Concentration (mg/L)			Odor thresholds (μg/L)	OAV	HX-NLS	QQ-NLS
				HX-NLS	QQ-NLS	av.				
3	ethyl isobutyrate	LLE	MS	86.70 ± 8.26	23.18 ± 2.66	54.94	57.47 <sup>a</sup>	1509	403	
22	β-damascenone	LLE	MS	0.055 ± 0.001	0.12 ± 0.01	0.0875	0.1 <sup>a</sup>	549	1236	
2	3-methylbutanal	DI	FID	8.70 ± 0.009	8.13 ± 0.76	8.415	16.51 <sup>a</sup>	527	492	
16	ethyl caprylate	DI	FID	5.48 ± 2.23	5.54 ± 0.04	5.51	12.87 <sup>a</sup>	426	430	
13	ethyl hexanoate	DI	FID	18.37 ± 3.12	4.07 ± 0.04	11.22	55.33 <sup>a</sup>	332	74	
10	ethyl valerate	DI	FID	2.00 ± 2.54	4.24 ± 0.17	3.12	26.78 <sup>a</sup>	172	158	
5	ethyl butyrate	DI	FID	10.69 ± 2.75	2.21 ± 0.36	6.45	81.5 <sup>a</sup>	131	27	
23	geosmin	LLE	MS	0.012 ± 0.001	0.013 ± 0.001	0.0125	0.11 <sup>a</sup>	114	122	
7	ethyl isovalerate	LLE	MS	0.59 ± 54.10	0.61 ± 0.07	0.6	6.89 <sup>a</sup>	86	89	
1	ethyl acetate	DI	FID	1940.83 ± 1.12	2370.75 ± 2.12	2155.79	32551.6 <sup>a</sup>	60	73	
4	ethyl acrylate	LLE	MS	0.96 ± 0.00	–	–	18 <sup>a</sup>	53	–	
6	ethyl 2-methylbutyrate	LLE	MS	0.39 ± 21.65	1.12 ± 0.01	0.755	18 <sup>a</sup>	22	62	
19	ethyl 2-hydroxy-4-methylvalerate	DI	FID	18.18 ± 6.45	10.73 ± 9.03	14.455	1268 <sup>a</sup>	14	8	
14	ethyl lactate	DI	FID	1199.25 ± 0.12	498.28 ± 1.12	848.765	128000 <sup>a</sup>	9	4	
9	2-methyl-1-propanol	DI	FID	360.78 ± 10.03	252.90 ± 1.00	306.84	40000 <sup>a</sup>	9	6	
11	1-butanol	DI	FID	24.52 ± 3.54	–	–	2733.35 <sup>a</sup>	9	–	
26	1-propanol	DI	FID	329.78 ± 3.34	246.52 ± 1.34	288.15	53952.63 <sup>a</sup>	6	5	
28	butyric acid	LLE	MS	4.60 ± 0.14	–	–	964.64 <sup>b</sup>	5	–	
34	phenylacetaldehyde	DI	MS	1.22 ± 0.04	–	–	262 <sup>a</sup>	5	–	
8	1,1-diethoxy-3-methylbutane	DI	MS	8.98 ± 2.37	27.80 ± 5.35	18.39	3000 <sup>a</sup>	3	9	
27	acetic acid	DI	FID	433.23 ± 0.00	405.60 ± 2.00	419.415	160000 <sup>a</sup>	3	3	
30	hexanoic acid	LLE	MS	6.18 ± 1.81	–	–	2517 <sup>b</sup>	2	–	
29	isovaleric acid	LLE	MS	1.89 ± 0.06	3.43 ± 0.05	2.66	1045.47 <sup>b</sup>	2	3	
25	γ-nonanolactone	DI	MS	0.12 ± 0.005	–	–	90.66 <sup>a</sup>	1	–	
33	1-hexanol	DI	FID	6.09 ± 4.00	–	–	5370 <sup>a</sup>	1	–	
31	octanoic acid	LLE	MS	2.73 ± 0.80	–	–	2701 <sup>b</sup>	1	–	
32	2-methyl-1-butanol	DI	FID	143.95 ± 0.00	–	–	179190 <sup>b</sup>	1	–	
21	phenethyl acetate	DI	MS	0.21 ± 0.003	0.21 ± 0.01	0.21	407 <sup>a</sup>	1	1	
12	3-methyl-1-butanol	DI	FID	54.84 ± 2.00	44.96 ± 1.20	49.9	179191 <sup>a</sup>	<1	<1	
20	ethyl benzoate	LLE	MS	0.38 ± 0.025	0.28 ± 0.01	0.33	1433.65 <sup>a</sup>	<1	<1	
24	phenethyl alcohol	DI	FID	5.50 ± 0.001	2.33 ± 1.04	3.915	28900 <sup>a</sup>	<1	<1	
35	1,1,3-triethoxypropane	LLE	MS	0.55 ± 0.02	–	–	3700 <sup>b</sup>	<1	–	
18	ethyl nonanoate	DI	MS	0.41 ± 0.003	0.27 ± 0.002	0.34	3150.61 <sup>a</sup>	<1	<1	
36	diethyl succinate	LLE	MS	23.26 ± 4.88	8.63 ± 0.35	15.945	353193.25 <sup>a</sup>	<1	<1	
17	1-heptanol	LLE	MS	0.086 ± 0.001	–	–	26600 <sup>a</sup>	<1	–	
15	2-hydroxy-3-methylbutanoic acid ethyl ester	LLE	MS	0.048 ± 0.001	0.85 ± 0.02	0.449	–	–	–	
37	ethyl isohexanoate	LLE	MS	–	0.39 ± 0.03	–	6 <sup>b</sup>	–	65	
38	acetoin	DI	FID	–	15.24 ± 2.00	–	259 <sup>a</sup>	–	59	
39	trimethyl-pyrazine	LLE	MS	–	0.19 ± 0.02	–	729.86 <sup>b</sup>	–	<1	
40	furfural	DI	FID	–	11.86 ± 3.30	–	122 <sup>a</sup>	–	97	
41	2,4-nonadienal	LLE	MS	–	0.67 ± 0.04	–	0.64 <sup>b</sup>	–	1054	
42	ethyl caprate	DI	MS	–	2.81 ± 0.04	–	1122.3 <sup>a</sup>	–	3	
43	ethyl phenylacetate	DI	MS	–	2.20 ± 0.04	–	407 <sup>a</sup>	–	5	
44	ethyl 3-phenylpropionate	LLE	MS	–	0.065 ± 0.007	–	125.21 <sup>a</sup>	–	1	
45	ethyl myristate	DI	MS	–	0.50 ± 0.03	–	46606 <sup>a</sup>	–	<1	
46	tetramethylpyrazine	LLE	MS	–	0.01 ± 0.002	–	80073.16 <sup>a</sup>	–	<1	

<sup>a</sup> Odor thresholds were taken from (Wang et al., 2022).

<sup>b</sup> Odor thresholds were taken from (Liu & Sun, 2018).

<sup>c</sup> HX-NLS (Hunzheng Xucha Niulanshan Baijiu); QQ-NLS (Qingzheng Qingcha Niulanshan Baijiu).

<sup>d</sup> Methods of sample pretreatment. LLE, liquid-liquid extraction; DI, direct injection.

<sup>e</sup> MS, mass spectrometer; FID, flame ionization detector.

from the Key Laboratory of Brewing Molecular Engineering of China Light Industry. They were trained for 3 weeks (30 min/day) so that they could characterize and distinguish the aromas in the 54 Aroma Master Kit (Le Nez du Vin®, France) (Pu et al., 2020) and different flavor style Baijiu. Finally, 20 panelists were selected based on their sensitivity to smell and their ability to describe the scents accurately. Next, 20 panelists were trained for an additional 3 h to identify and define the descriptive terms for NLS using different flavor style Baijiu and related aroma compounds or materials reported in literatures (Dong et al., 2019; Gao et al., 2021; Huang et al., 2023). They were required to evaluate the aroma characteristics of NLS (HX, QQ) and write down 7–10 aroma descriptors, from which seven of the highest frequency terms were selected to evaluate the aroma characteristics of NLS. Seven aroma

characteristics and their corresponding reference standards (Wang et al., 2021; Wang et al., 2022) (shown in Table S4) were fruity (ethyl acetate), floral (phenethyl alcohol), alcoholic (ethanol), Chen-aroma (mature vinegar), pickles-aroma (pickled pickles in three months), Zao-aroma (NLS' Jiupai), sour (acetic acid). The samples (HX-NLS, QQ-NLS, 20.0 mL) were placed in a tasting glass of IOS standard international professional liquor with a capacity of 50 mL. All sensory evaluation experiments were conducted in a sensory evaluation room at a temperature of 21 ± 1 °C and humidity of 45–50%.

## 2.6.2. Classify consumers

Recruited volunteers at fast-moving consumer goods locations such as supermarkets, coffee shops, and fast-food restaurants. Asked them if

they were interested in participating in sensory evaluation experiments and paid each participant \$10 as compensation. The volunteer questionnaire was shown in Table S5. Based on the results of the questionnaire, volunteers who met the requirements were selected to participate in the sensory evaluation experiment. Finally, one hundred and eleven people (25–45 years old) with rich drinking experience were recruited. They were not just students from the brewing engineering department of Beijing Technology and Business University. First, HX-NLS and QQ-NLS were scored with the 9-point hedonic scales. At the same time, the consumers were asked to carry out sensory attribute JAR test on the seven aroma characteristics of Baijiu. According to the scores of different sensory attribute JAR test, the consumers were divided into three categories: the first group (exactly 1–2 points), who thought the aroma intensity was too less or too weak (TL); the second group (exactly 3 points), who thought the aroma was just right (JR); the third category (exactly 4–5 points), who thought the aroma was too more or too strong (TM). “P” represents the percentage of a certain category of consumers with a certain aroma attribute in the total number of consumers (Pagès et al., 2014).

### 2.6.3. Calculation of “Penalty coefficient”

For the above three groups with a certain aroma attribute, the arithmetic mean values of the overall preference degree of each group for different liquors were calculated, which were denoted as L(TL), L(JR) and L(TM) (Rothman, 2007a; Rothman, 2007b). The penalty coefficient of a certain aroma feature is the deviation between the mean of the overall preference degree of the consumers and the mean of the overall preference degree of JR, which is  $L(TL)-L(JR)$  or  $L(TM)-L(JR)$  (Zhi et al., 2016; Pagès et al., 2014; Popper, 2014).

### 2.6.4. Calculation of “Total mean Drops”

The penalty coefficient is multiplied by the proportion of people with the bias “P” to indicate the total scores that the overall consumer preference is reduced, because the sensory attribute is biased in one direction or the other (Cong et al., 2017; Lawless et al., 2010; Nie, Tong & Huang, 2019; Xiang, 2005).

## 2.7. Experiment on liquor blending

According to the results of penalty analysis, in order to better improve the quality of liquor, it is necessary to improve the aroma characteristics of liquor whose sensory evaluation is not appropriate. The compounds with OAVs greater than 1 in HX-NLS and QQ-NLS were compared to calculate the content differences among these compounds, followed by addition experiments. The sensory evaluation team used triangulation test to improve the aroma of the samples (Wang et al., 2022), including two original samples and one aroma improved sample, which were random numbered with three letters or numbers. When the aroma characteristics of the “improved sample” were not significantly different from those of the sample with better results in penalty analysis, a penalty analysis test was conducted on the aroma properties of the “improved sample” to verify whether the experimental results were satisfactory.

## 2.8. Statistical analysis

Sensory data were analyzed by ANOVA using SPSS26 (SPSS Inc., Chicago, IL) to compare significant differences. GC-MS data was analyzed using software Qualitative Analysis 10.0 (Agilent, America). Results of penalty analysis were statistically analyzed by Microsoft Excel 2019 (Microsoft, America). Images were drawn by Origin 2021 (Origin Lab, America).

## 3. Results and discussion

### 3.1. Identification of aroma active compounds

GC-O-MS combined with Osme was used to analyze the aroma components in HX-NLS and QQ-NLS. The experimental results were shown in Table 1. A total of 46 aroma-active compounds and two unknown compounds were found, including 22 esters, 8 alcohols, 5 acids, 4 aldehydes, 2 ketones, 2 pyrazines and 3 other compounds. Esters are the most common compounds in Baijiu (Liu & Sun, 2018). They mainly come from the esterification of acid and alcohol, which was reported as early as 1961 (Nordström, 1961). At present, there are three main formation pathways of esters in liquor: 1) Microbial metabolic formation, depending on the activity of *acyl-coA synthetase* and *alcohol acetyl transferase* (Hiroyuki, Daisuke, Takayuki & Naoshi, 2001; Saerens, Delvaux, Verstrepen, Van Dijck, Thevelein & Delvaux, 2008); 2) Being formed by esterification of acid and alcohol under the catalysis of lipase (Peryam et al., 1957); 3) Being produced by chemical reaction in liquor aging stage (Nordström, 1961). Most of alcohols in Baijiu were higher alcohols, which were important components of liquor flavor (Liu & Sun, 2018). They are mainly produced by *Saccharomyces cerevisiae* (Van Nedervelde, Drukker, Benvenisty, & Debourg, 2001). Baijiu also contains a large number of organic acids, and acetic acid was the most common acid in light flavor Baijiu (Wang, Ye, Zhu et al., 2022). These acids in Baijiu were the precursors of esters and are mainly produced by metabolic activities of microorganisms (Gao, 2017). Aldehydes and ketones generally had strong odors and contribute to the aroma release of Baijiu, and were generally derived from photooxidation degradation, thermal oxidation degradation, as well as husk, bran and other brewing materials (Liu, Huang, Zheng, Chen & Sun, 2010; Yang, 2009). Pyrazines are usually formed by Maillard reactions (Fan, Xu & Zhang, 2007).

A total of 36 aroma-active compounds were identified in HX-NLS, and 35 odorants were identified in QQ-NLS. Among them, ethyl acrylate, 1-butanol, 1-heptanol,  $\gamma$ -nonanolactone, butyric acid, hexanoic acid, octanoic acid, 2-methyl-1-butanol, 1-hexanol, phenylacetaldehyde and 1,1,3-triethoxypropane were only detected in HX in this study. Ethyl isohexanoate, acetoin, trimethyl-pyrazine, furfural, 2,4-nonadienal, ethyl caprate, ethyl phenylacetate, ethyl 3-phenylpropionate, ethyl myristate and tetramethylpyrazine were found only in QQ-NLS. Ethyl isohexanoate and 2,4-nonadienal were found in NLS for the first time, after investigating previously reported literature.

The Osme values of ethyl acetate, 3-methylbutanal, 3-methyl-1-butanol, ethyl benzoate, phenylethyl acetate,  $\beta$ -damascenone, geosmin, phenethyl alcohol,  $\gamma$ -nonanolactone, acetic acid, and isovaleric acid in HX-NLS were higher (Osme greater than 4). The Osme values of ethyl acetate, 3-methylbutanal, 3-methyl-1-butanol, ethyl caprylate,  $\beta$ -damascenone, geosmin, acetic acid, isovaleric acid, and 2,4-nonadienal in QQ-NLS were higher (Osme greater than 4). Among them, the Osme values of ethyl acetate, 3-methylbutanal, 3-methyl-1-butanol,  $\beta$ -damascenone, geosmin, acetic acid and isovaleric acid in two NLS with different processes were all greater than 4. In previous studies (Wang et al., 2022), our team found that ethyl acetate, 3-methylbutanal,  $\beta$ -damascenone and geosmin are the key aroma components in QQ-NLS. It could be found that they existed simultaneously in HX-NLS and QQ-NLS, and were the important aroma components in NLS. Ethyl acetate,  $\beta$ -damascenone and geosmin also had higher Osme values in Fen Jiu (3.83, 4.00, 2.67), Baofeng Jiu (4.33, 4.67, 4.17) and Qingke Jiu (2.33, 2.50, 2.17) (Gao, Fan & Xu, 2014). 3-Methyl-1-butanol had been identified as aroma-active compound in all kinds of light flavor Baijiu (Wang et al., 2022). In addition, the compounds with Osme scores of 3–4 in both NLS of HX and QQ included ethyl isobutyrate, ethyl butyrate, ethyl 2-methylbutyrate, ethyl hexanoate, ethyl nonanoate and ethyl 2-hydroxy-4-methylvalerate. They also have strong aroma and were all ester compounds, which may be important odorants in NLS.

**Table 3(a)**  
Penalty analysis results for HX-NLS <sup>a</sup>

Proportion of people who prefer this Baijiu		51.4%							
Sensory attributes		fruity	floral	Zao-aroma	Chen-aroma	Pickles aroma	alcoholic	acidic	
	TL <sup>b</sup> (1-2 points)	Total mean drops	-0.2857	-0.1518	-0.0577	-0.0439	-0.0845	-0.0197	0.0416
		Penalty coefficient	-0.6607	-0.2674	-0.7121	-0.1477	-0.4464	-0.3636	0.1538
JR <sup>b</sup> (3 points)		P (%) <sup>c</sup>	43.24%	56.76%	8.11%	29.73%	18.92%	5.41%	27.03%
		P (%) <sup>c</sup>	37.84%	35.14%	59.46%	43.24%	43.24%	29.73%	35.14%
		P (%) <sup>c</sup>	18.92%	8.11%	32.43%	27.03%	37.84%	64.86%	37.84%
TM <sup>b</sup> (4-5 points)		Penalty coefficient	-0.1429	0.5897	-0.0455	0.5250	0.4821	-0.5720	0.2253
		Total mean drops	-0.0270	0.0478	-0.0147	0.1419	0.1824	-0.3710	0.0852

<sup>a</sup> HX-NLS (Hunzheng Xucha Niulanshan Baijiu).<sup>b</sup> TL, people who thought the aroma intensity was too less or too weak (TL); JR, people who thought the aroma intensity was just right (JR); TM, people who thought the aroma was too more or too strong.<sup>c</sup> Values are reserved for two significant digits after the decimal point.**Table 3(b)**  
Penalty analysis results for QQ-NLS <sup>a</sup>

Proportion of people who prefer this Baijiu		43.2%							
Sensory attributes		fruity	floral	Zao-aroma	Chen-aroma	Pickles aroma	alcoholic	acidic	
	TL <sup>b</sup> (1-2 points)	Total mean drops	-0.3058	-0.1532	-0.3586	-0.4955	0.0118	-0.1757	-0.5081
		Penalty coefficient	-1.6165	-0.4048	-0.8292	-1.3095	0.0257	-1.3000	-1.5667
JR <sup>b</sup> (3 points)		P (%) <sup>c</sup>	18.92%	37.84%	43.24%	37.84%	45.95%	13.51%	32.43%
		P (%) <sup>c</sup>	51.35%	48.65%	40.54%	56.76%	43.24%	27.03%	54.05%
		P (%) <sup>c</sup>	29.73%	13.51%	16.22%	5.41%	10.81%	59.46%	13.51%
TM <sup>b</sup> (4-5 points)		Penalty coefficient	-1.2919	0.1667	-1.2667	-0.7381	1.1875	-0.3727	-1.4000
		Total mean drops	-0.3841	0.0225	-0.2054	-0.0399	0.1284	-0.2216	-0.1892

<sup>a</sup> QQ-NLS (Qingzheng Qingcha Niulanshan Baijiu).<sup>b</sup> TL, people who thought the aroma intensity was too less or too weak (TL); JR, people who thought the aroma intensity was just right (JR); TM, people who thought the aroma was too more or too strong.<sup>c</sup> Values are reserved for two significant digits after the decimal point.

### 3.2. Quantification of Aroma-Active compounds and OAV analysis

To further determine important odorants in NLS, the aroma-active compounds in NLS were quantified by internal standard curve methods, and their OAV were calculated. The results were shown in Table 2. A total of 35 compounds had OAV  $\geq 1$ . The content of ethyl acetate (av. 2155.79 mg/L) was the highest, followed by ethyl lactate (av. 848.77 mg/L), 2-methyl-1-propanol (av. 306.84 mg/L) and 1-propanol (av. 288.15 mg/L). Ethyl acetate and ethyl lactate usually have the higher contents and are the skeleton components in Baijiu (Liu et al., 2018). These compounds were present in both NLS of HX and QQ, and their OAVs were more than 1. This indicated that they might play an important role in the flavor of NLS. Ethyl acetate can speed up the body's metabolism and dilate blood vessels (Long, Tang, Wang, Shi & Wu, 2022; Wang, Zhou & Xia, 2005). Ethyl lactate may enhance the mellow sense of NLS and coordinate the aroma (Yang, 2012). 1-Propanol and 2-methyl-1-propanol are both high alcohols. The right amounts of high alcohols can make the liquor coordination, but once their concentrations are too high, they will make people headache, and difficult to sober up after drunk (Huang, Zhang, Tong & Zhang, 2022). Their concentrations should be controlled within an appropriate range. The average contents of ethyl isobutyrate, 3-methyl-1-butanol, 1,1-diethoxy-3-methylbutane, diethyl succinate, ethyl 2-hydroxy-4-methylvalerate and ethyl hexanoate in the two kinds of liquors were in the range of 10–100 mg/L. Ethyl isobutyrate exists widely in light flavor Baijiu of Daqu (Wang et al., 2022), and its content in HX-NLS was significantly higher than that in QQ-NLS, which may be related to their different processes. The OAV of ethyl isobutyrate (HX-NLS, 1509) was the highest, and its OAV in QQ-NLS (403) was also higher. Although the content of 3-methyl-1-butanol was high, its aroma intensity was not obvious, and OAV was also less than 1 due to its high threshold. The OAV of 1,1,3-triethoxypropane and diethyl succinate were also not high, and they were all less than 1. The OAV of ethyl hexanoate (HX-NLS, 332; QQ-NLS, 74; the latter was the same) were also very high. Ethyl hexanoate also had higher OAV in

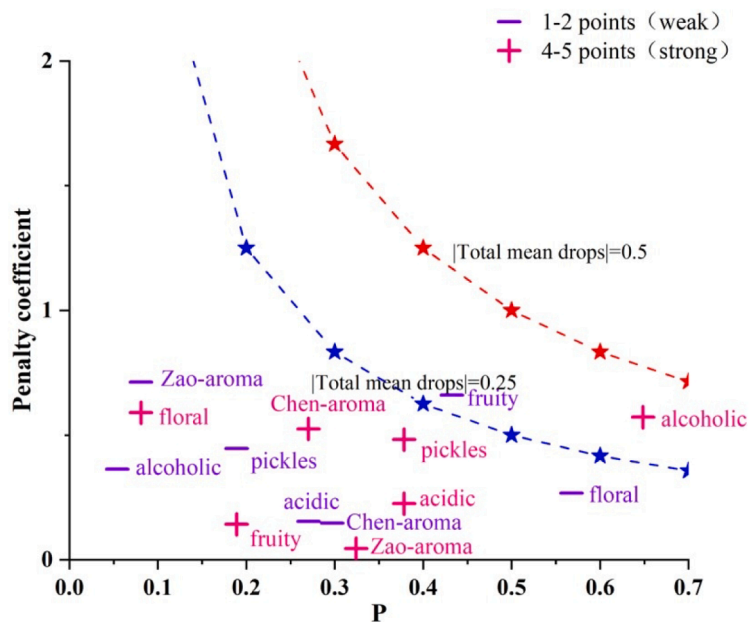
other light-flavor Baijiu, such as Caoyuanwang Baijiu (av.195) (Wang et al., 2021), Fen Jiu (56), Baofeng Jiu (97) and Qingke Jiu (139) (Gao et al., 2014). The OAV of  $\beta$ -damascenone (549; 1236), 3-methylbutanal (527; 492), ethyl caprylate (426; 430), ethyl valerate (172; 158) and geosmin (114; 122) were greater than 100. These compounds were also been found to be key aroma components in QQ-NLS (Wang et al., 2022), making important contributions to the aroma of QQ-NLS. Clearly, they were also likely to be key aromas in HX-NLS, providing a certain fruity sweetness and aldehyde odor to HX-NLS.

### 3.3. Penalty analysis of sensory evaluation data

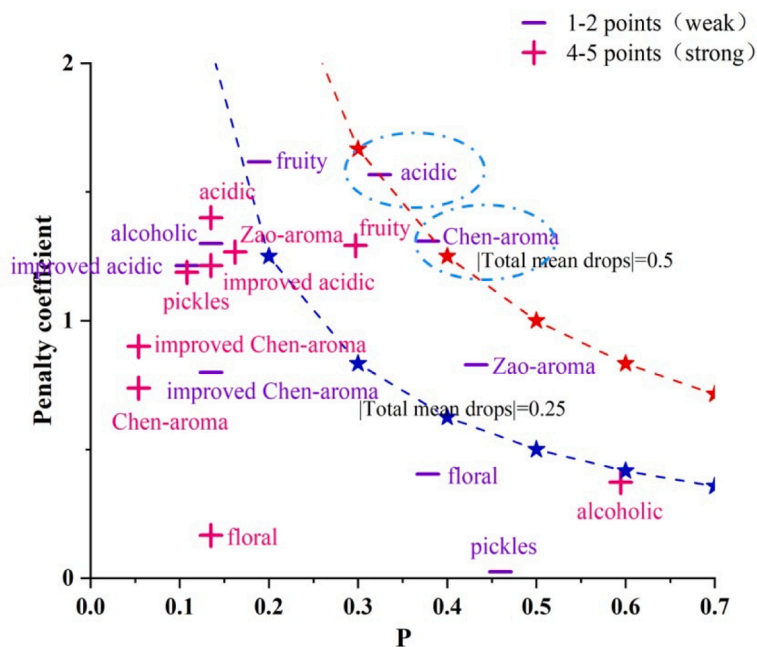
The overall aroma characteristics and each aroma attributes of the HX-NLS and QQ-NLS were tested by penalty analysis, and the results of the 9-point hedonic scales and the five-point JAR scale test were tallied. The final results are shown in Tables 3(a) and (b). Table 3(a) and Table 3 (b) respectively show the statistical results of penalty analysis from 111 people with rich drinking experience. According to the overall preference scale test of HX-NLS and QQ-NLS (Tables 3(a) and (b)), 51.4% of them preferred HX-NLS, 43.2% liked QQ-NLS, and about 5% didn't like or dislike these two liquors. P (%) in Tables 3(a) and (b) represents the proportion of a certain group of people. It is generally believed that if the number of people in the first (TL) and third (TM) groups exceeds 30%, the aroma attribute may have potential problems, that is to say, at least 30% of people think the aroma attribute is not appropriate. For example, as showed in Tables 3(a) and (b) (HX-NLS), the proportion of fruity (43.24%) and floral (56.76%) in the first group (TL), and the proportion of alcoholic (64.86%) in the third group (TM) was much more than 30%. The proportion of floral, Zao, Chen, pickles, and acidic aroma in QQ-NLS were more than 30% in the first group (TL). It showed that, compared with HX-NLS, the consumers thought that these aroma intensities of QQ-NLS were slightly weaker.

The penalty coefficient indicated the degree of deviation, usually with 0.5 as the dividing line. The larger the absolute value of the aroma





(a)



(b)

**Fig. 2.** Penalty analysis diagram of Niulanshan Baijiu produced by two different processes. (a) Hunzheng Xucha process; (b) Qingzheng Qingcha process Note: The penalty coefficient of a certain aroma feature is the deviation between the mean of the overall preference degree of the consumers of each group and the mean of the overall preference degree of Just right (JR); “P” represents the percentage of a certain category of consumers with a certain aroma attribute in the total number of consumers.

penalty coefficient was, the greater the influence of the aroma attribute on the overall product preference was (Cadot et al., 2010). According to this condition, these aroma attributes, including fruity, Zao, floral, Chen, alcoholic aroma in HX-NLS, fruity, Zao, Chen, pickles, alcoholic, and acidic aroma in QQ-NLS, needed to be improved as much as possible. However, only based on this condition, it was easy to enter the wrong area, and could not make a reasonable judgment on the overall aroma of

the two liquors. For example, the floral aroma in HX-NLS group 1 and the pickles aroma in QQ-NLS group 1 both had higher P (%), but the penalty coefficients were very low. This was because it did not affect their preference for these two liquors at all, even though many people thought the floral and pickles odor were not strong enough. This showed that this index was not the most core index of the products, whether to improve it or not would not have great impact on the evaluation results.

**Table 4**  
Penalty analysis of QQ-NLS after aroma improvement<sup>a</sup>

Sensory attributes		Chen-aroma	acidic
TL <sup>b</sup> (1-2 points)	Total mean drops	-0.1081	-0.1313
	Penalty coefficient	-0.8000	-1.2143
	P (%) <sup>c</sup>	13.51%	10.81%
JR <sup>b</sup> (3 points)	P (%) <sup>c</sup>	81.08%	75.68%
	P (%) <sup>c</sup>	5.41%	13.51%
TM <sup>b</sup> (4-5 points)	Penalty coefficient	-0.9000	-1.2143
	Total mean drops	-0.0486	-0.1641

<sup>a</sup> QQ-NLS (Qingzheng Qingcha Niulanshan Baijiu).

<sup>b</sup> TL, people who thought the aroma intensity was too less or too weak (TL); JR, people who thought the aroma intensity was just right (JR); TM, people who thought the aroma was too more or too strong.

<sup>c</sup> Values are reserved for two significant digits after the decimal point.

Therefore, it was necessary to combine P (%) and penalty coefficient, called “total mean drops” (multiply two values). Generally speaking, the larger is the|Total mean drops|of aroma attributes, the more improvement is needed. If|Total mean drops|are  $\geq 0.5$ , they needed to be improved first (Nordström, 1961). In order to see more clearly which aroma attribute needed to be improved in priority, penalty analysis charts were drawn and shown in Fig. 2 (a) and 2 (b), corresponding to Tables 3(a) and (b) respectively. The absolute value of penalty coefficient was taken as the ordinate, and “P” as the abscissa. Two curves were drawn based on|Total mean drops|= 0.5 and|Total mean drops|= 0.25, and the attributes that have priority for improvement were near the upper right of the pictures. As seen intuitively from the Fig. 2, HX-NLS had few aroma attributes that needed to be improved, and most people were satisfied with the flavor of HX-NLS. However, some aroma properties in QQ-NLS urgently needed to be improved, especially the acidic and Chen-aroma needed to be improved first.

### 3.4. Aroma improvement experiment results analysis

According to the test results of preference, QQ-NLS was selected for aroma improvement. As shown in Fig. 2, the sour-Chen aroma needs to be improved most in QQ-NLS. In other words, the aroma needed to be strengthened. According to the quantitative results in Table 2, these compounds with OAV greater than 1 had higher contents in HX-NLS than those in QQ-NLS, such as ethyl isobutyrate, 3-methylbutanal, ethyl hexanoate, ethyl butyrate, ethyl acrylate, ethyl 2-hydroxy-4-methylvalerate, ethyl lactate, 2-methyl-1-propanol, 1-propanol, butyric acid, phenylacetaldehyde, acetic acid, hexanoic acid,  $\gamma$ -nonanolactone, 1-hexanol, octanoic acid and 2-methyl-1-butanol. According to the results of penalty analysis, the compounds without floral and fruity aroma were selected, including 3-methylbutanal, ethyl acrylate, ethyl 2-hydroxy-4-methylvalerate, 2-methyl-1-propanol, 1-propanol, butyric acid, acetic acid, hexanoic acid, 1-hexanol, octanoic acid. The permutation and combination (combined according to the factorial of “n”) of these compounds in the two NLS was added to QQ-NLS according to the content difference in Table 2. Then, the sensory evaluators (20 people) conducted sensory test on sour-Chen aroma by triangular test (including two cups of HX-NLS and one cup of added samples). The results were shown in Table 4. After several experiments, it was finally found that when acetic acid, butyric acid, hexanoic acid, octanoic acid and 3-methylbutanal were added to QQ-NLS, the aroma of sour-Chen in QQ-NLS could be significantly enhanced. Then, the above penalty analysis test was repeated for the sour-Chen aroma of the improved samples, and the final results were shown in Fig. 2 (b). It was seen from Fig. 2 (b) that the sour-Chen aroma of QQ-NLS after adjustment is significantly improved, and the satisfaction is also significantly enhanced. This indicated that acetic acid, butyric acid, hexanoic acid, octanoic acid and 3-methylbutanal played an important role in the sour-Chen aroma of NLS, and their contents determined the intensity of sour-Chen aroma.

## 4. Conclusions

In summary, this study presents a novel approach by integrating penalty analysis testing with food sensory omics to investigate NLS consumer preferences and aroma regulation. Through the utilization of two different extraction methods, LLE and HS-SPME, along with GC-OMs, a comprehensive analysis detected a total of 46 aromatic active compounds. Notably, Ethyl isohexanoate and 2,4-nonadienal were identified for the first time in NLS. By combining GC-O, qualitative, and quantitative analyses, a total of 35 aroma compounds with OAV greater than 1 were identified. Subsequently, penalty analysis experiments were conducted with 111 consumers, and the results, combined with OAV findings, enabled targeted improvements to the aroma attributes with lower overall preferences in QQ-NLS. As a result, the aroma intensity was successfully enhanced, leading to improved consumer satisfaction. The key compounds (acetic acid, butyric acid, hexanoic acid, octanoic acid, and 3-methylbutanal) of sour-Chen aroma in NLS were first determined. It was the first time to identify the aroma compounds that affected the sour-Chen flavor of NLS, which laid a foundation for the future study on the sour-Chen aroma of all light flavor Baijiu. This study provides a new method for enterprises to regulate the aroma of liquor in the future, and can also be an important supplement to traditional Baijiu blending methods. It is of great significance to improve liquor sales and develop a liquor that meets consumer satisfaction.

### CRedit authorship contribution statement

**Zhen Wang:** Investigation, Methodology, Software, Writing – original draft, Writing – review & editing. **Wenjun Hao:** Investigation, Methodology, Project administration. **Jinwang Wei:** Resources. **Minquan Huang:** Investigation, Methodology, Writing – review & editing. **Xinan Zeng:** Writing – review & editing. **Ying Wang:** Resources. **Jihong Wu:** Resources. **Boru Chen:** Resources.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.fochx.2023.100852>.

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