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Analysis of methanol, fusel alcohols, and other volatile compounds of local specialty alcoholic beverages (wine, beer, and *soju*) in Korea

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Abstract

The most commonly consumed local specialty alcoholic beverages in South Korea are wine, beer, and *soju*. These alcoholic beverages contain a wide variety of volatile components, including methanol and fusel alcohols, due to their different raw materials, manufacturing methods, and fermentative microorganisms. GC–MS combined with solid phase micro-extraction (SPME) was utilized to establish a simultaneous analytical method for methanol, fusel alcohols, and other volatile compounds in alcoholic beverages, which included 9 wine, 10 beer, and 10 *soju* samples. A total of 221 volatile compounds were identified, consisting of 6 acetals, 14 acids, 33 alcohols, 9 aldehydes, 2 amides, 13 benzene derivatives, 5 phenols, 81 esters, 8 furans, 16 ketones, 6 sulfides, 26 terpenes, 1 pyrazine, and 1 miscellaneous. Among the three types of alcoholic beverages, wine had the highest methanol content. In case of fusel alcohols, wine, beer, and *soju* contained 25, 16, and 14 alcohol components, respectively. In general, those main volatile components of wine, beer, and *soju* were esters, alcohols, and benzene derivatives. In beer, terpenes were detected at similar levels with alcohols. Volatile compounds contributing to the distinction between *soju* and beer were some alcohols, such as propan-1-ol, 2-methylpropan-1-ol, and 3-methylbutan-1-ol, and acids, such as octanoic acid, whereas wine samples were distinguished from other types of alcoholic beverages by some esters, such as methyl acetate, ethyl (E)-but-2-enoate, ethyl 3-hydroxybutanoate, and diethyl butanedioate, and some alcohols, such as hexan-1-ol, nonan-2-ol, and nonan-1-ol.

Keywords Alcoholic beverage, Wine, Beer, *Soju*, Methanol, Fusel alcohols, Volatile compounds

Introduction

According to the Korea Agro-Fisheries and Food Trade Corporation, the size of the alcoholic beverage market has been declining since 2016 by launch price. Also, per capita alcohol consumption has been on a downward trend over the past decade through 2021. In contrast, the size of the local specialty alcohol market has been more than doubled from 2017 to 2021. As the local specialty alcohol market grows, it is important to evaluate scientifically the safety and quality of local specialty alcohol products in Korea. From 2010 to 2020, the most highly consumed alcoholic beverages in South Korea were beer, *soju*, *and* wine.

Wine are made by fermentation of raw fruits such as grapes and berries [9]. Beer is produced by brewing ingredients, such as grain, and hops, using yeast [25]. *Soju* are produced by distillation of fermented alcoholic beverages to increase the ethanol content [15]. These different types of alcoholic beverages vary by raw materials, production methods (only *soju* is distilled, whereas the

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others are fermented), microorganisms used, and ethanol content of final products. This diversity results in a variety of by-products, such as methanol, higher alcohols, and other volatile compounds.

Methanol is a volatile harmful component produced from the demethylation of pectin by pectinolytic enzymes that are naturally present in fruits such as grapes during alcohol fermentation of wine [22]. Not only can it cause a hangover symptoms after consuming alcoholic beverages [29], but it also has toxic effects on the human body, including blindness or death [11]. According to Korean Ministry of Food and Drug Safety (MFDS) regulation, the regulatory limits for methanol in alcoholic beverages are 1 mg/mL for wine and 0.5 mg/mL for other alcoholic beverages, respectively.

Fusel alcohols, also known as higher alcohols, are alcohols with two or more carbon atoms. They are generally produced from either sugar metabolism (anabolic pathway) or amino acid catabolism (Ehrlich pathway) [24]. They are harmful substances that are known to cause hangovers, which can include dizziness and headaches after consuming alcoholic beverages [6], whereas they serve as important flavor components in alcoholic beverages [35]. They are also involved as ester precursors that are critical for the flavor of alcoholic beverages [4].

Raw materials, production methods, and aging processes can contribute to the flavor characteristics of alcoholic beverages [45]. Among these factors, the formation of volatile compounds in wine can be affected primarily by microorganisms such as yeast, bacteria, and molds [24]. Beer contains a wide range of volatile components that can react with each other during the brewing process [28]. During the fermentation process, yeasts take up amino acids and sugars from the wort. Those sugars are metabolized, under anaerobic conditions, and converted primarily to ethanol and carbon dioxide [1]. As by-products of fermentation, volatile metabolites such as aldehydes, ketones, higher alcohols, organic acids, and esters are produced [1]. On the other hand, amino acids regulate the biosynthesis of flavor-active compounds formed by yeasts [12]. *Soju*, one of the most popular alcoholic beverages in Korea, is rich in volatile compounds because it is manufactured through a complex fermentation process of raw materials followed by a high-temperature distillation process (Wu, Jihong et al., 2021). The volatile components of *soju* can vary depending on the distillation conditions, aging stage, and manufacturing conditions, as well as the fermentation of the raw materials [13].

Although there have been many studies which analyzed methanol, fusel alcohols, and volatile components of alcoholic beverages [15, 21, 22], there are limited studies that have simultaneously analyzed them in alcoholic beverages. In addition, no study has been reported on volatile components of local specialty alcoholic beverages in Korea, to our knowledge. Therefore, in this study, methanol, fusel alcohols, and other volatile components were analyzed simultaneously for wine, beer, and *soju*, which are major local specialty alcoholic beverages in Korea (Table 1). In addition, volatile compositions of different alcoholic beverages were compared to determine those key volatile compounds that contribute to distinguishing between different samples. This study could provide a scientific basis for quality control of local specialty alcoholic beverages.

Methods/experimental

Samples

A total of 29 different types of alcoholic beverages distributed in Korea, including wine (n = 9), beer (n = 10), and *soju* (n = 10), were analyzed. S*oju* is distilled alcoholic beverage and the rest is classified as fermented alcoholic beverage. All samples were stored at −70 °C in amber bottles until their analysis.

Chemicals

As an internal standard for methanol analysis, ethanol-1,1,2,2,2-d_5 was purchased from Sigma Aldrich Corporation (St. Louis, MO, USA). As an internal standard for volatiles analysis, 3-pentanol was purchased from Sigma Aldrich Corporation. *n*-Alkane standards (C_7-C_{30}) and all other authentic standards were obtained from Sigma-Aldrich. For all aqueous solutions, distilled water obtained from an ultra pure water system (Human Science, Seoul, Korea) was used.

Extraction of volatile compounds in alcoholic beverages

To extract methanol and other volatile compounds from the samples, solid phase microextraction (SPME) was performed using GERSTEL MPS autosampler (GERSTEL, Mulheiman der Ruhr, Germany). 100 uL of ethanol-1,1,2,2,2-d5 (1.00 μg/μL in water) and 50 uL of 3-pentanol (0.500 μg/μL in 15% ethanol) were added as internal standards in 20 mL headspace amber screw vial (Agilent Technologies, Santa Clara, CA, USA) filled with 5 mL of alcoholic beverage. The samples were equilibrated at 50 ℃ and 250 rpm for 20 min. The adsorption was performed under the same condition with equilibration using Carboxen/polydimethylsiloxane fiber (Carboxen-PDMS, 75 μm Supelco, Bellefonte, PA, USA). The extracted analytes were desorbed in the injection port of GC at 250 ℃ for 5 min. SPME fiber was exposed to the sample headspace at penetration depth of 21 mm to adsorb the analytes, before desorbed in a GC injector at 250 ℃ for 5 min. Baking was employed for 3 min before

Table 1 Alcoholic beverage samples used in this study

Samples	Origin	Ingredients used	Alcohol contents (%)
Wine			
w1	Gyeongsangbuk-do	Grape, yeast, potassium metabisulfite	13
w2	Gyeonggi-do	Grape, sugar, yeast, antioxidant, sulfur dioxide	13.5
w3	Gangwon-do	Grape, sugar, yeast, sulfurous acid	11.5
w4	Chungcheongnam-do	Grape, sugar, antioxidant, synthetic preservatives, potassium metabisulfite, potassium sorbate	12
w5	Jeollabuk-do	Wild grape, sugar, yeast, antioxidant, synthetic preservatives, potassium sulfite, potassium sorbate	12
w6	Gyeonggi-do	Grape, fructose, antioxidant, sulfur dioxide	9.9
w7	Chungcheongbuk-do	Grape, sugar, yeast, antioxidant, synthetic preservatives, potassium metabisulfite, potassium sorbate	12
w8	Gyeongsangbuk-do	Grape, sugar, yeast, potassium metabisulfite	12
w9	Gyeonggi-do	Wild grape, ethanol, sugar, salt, antioxidant, acidity regulator, *DL*-malic acid, sodium sulfite anhydrous, alanine	12
Beer			
b1	Jeollabuk-do	Rice, barley malt, hop pellet, yeast, carbon dioxide	5
b2	Gyeonggi-do	Rice, barley malt, hop, yeast	4.5
b3	Chungcheongnam-do	Malt, hop pellet, yeast, carbon dioxide	6
b4	Jeollanam-do	Rice, wheat, malt, hop pellet, yeast, carbon dioxide	4.5
b5	Gyeongsangbuk-do	Wheat, barley malt, hop pellet, yeast, spelt seed, oat flake, citric acid, calcium chloride, calcium sulfate	4.7
b6	Gangwon-do	Barley malt, hop pellet, yeast, carbon dioxide, acidity regulator, fortifying nutrient	4.7
b7	Gyeonggi-do	Rice, barley malt, hop pellet, yeast, acidity regulator, fortifying nutrient, lactic acid bacteria	4.5
b8	Gangwon-do	Rice, malt, hop, yeast	5.4
b9	Jeollabuk-do	Malt, hop, yeast, carbon dioxide, sodium carbonate, calcium sulfate, calcium chloride, magnesium sulfate	5
b10	Gyeongsangbuk-do	Malt, hop, yeast	6.2
Soju			
s1	Gyeongsangbuk-do	Rice, *nuruk*, yeast, lactic acid	32
s2	Gyeongsangbuk-do	Rice, *nuruk*	45
s3	Jeju Special Self-Governing Province	Rice, *nuruk*, hulled foxtail millet, hulled barley	23
s4	Gyeonggi-do	*Nuruk*, yeast, foxtail millet, sorghum	29
s5	Gangwon-do	Rice	40
s6	Gangwon-do	Rice, *nuruk,* yeast	41
s7	Chungcheongbuk-do	Distillate	24
s8	Gyeonggi-do	Rice, *ipguk,* yeast	25
s9	Gyeongsangbuk-do	Rice, *nuruk*, *koji*, yeast	25
s10	Chungcheongnam-do	Rice, *ipguk*, yeast	22

and after extraction, respectively, to prevent contamination of the fiber by remaining residues.

Analysis of volatile compounds

Volatile analysis was carried out using a 7890A gas chromatograph (Agilent Technologies)−5977B mass selective detector (Agilent Technologies) equipped with a DB-WAX column (30 m length × 0.25 mm i.d. × 0.25μm film thickness, J&W Scientific, Folsom, CA, USA). The initial GC oven temperature was maintained at 35 ℃ for 5 min, increased to 80 ℃ at a rate of 3 ℃/min, and 220 ℃ at a rate of 28 ℃/min, before held for 10 min. Helium (99.999%) was used as the carrier gas at a steady flow rate of 0.8 mL/min. Injection was conducted on split mode (10:1) with both injector and transfer line temperatures of 250 ℃. The mass spectrometer was operated in the simultaneous selected ion monitoring (SIM)/scan mode as the acquisition type, using with a scanning range of 35–350 atomic mass units (a.m.u.) in the electron impact (EI) ion source mode at 70 eV. The SIM data were used to extract the ion peaks of methanol. The scan data were used to extract the ion peaks of volatile compounds.

Identification and quantification of volatile components

Methanol in alcoholic beverages was confirmed by comparing retention times, and also both qualification and quantification ions of authentic standard compounds. The qualification was conducted by monitoring m/z 29, 31, and 32 for methanol and 33, 49, and 51 for ethanol-1,1,2,2,2-d_5, while the quantification was carried out by monitoring m/z 31 for methanol and 51 for ethanol-1,1,2,2,2-d_5.

Volatile compounds were positively determined by comparison of retention times (RT) and mass spectral data to those of authentic standard chemicals. In case where authentic standards were not available, the compounds were identified by comparing their mass spectra using the NIST Chemistry Webbook. Retention index (RI) of volatile compounds was calculated using *n*-alkanes from C_5 to C_{30} as an external standard to confirm the identification. Quantitative analysis was performed by comparing their peak areas to that of the internal standard compound. All of the samples were experimented in triplicate.

Data analysis

Principal component analysis (PCA) was performed to determine the difference in fusel alcohols and other volatile compounds, respectively, depending on different types of alcoholic beverages by using SIMCA 16 software (Umetrics, Umea, Sweden).

Results and discussion

Volatile composition

A total of 221 volatile compounds were found, including 6 acetals, 14 acids, 33 alcohols, 9 aldehydes, 2 amides, 13 benzene derivatives, 5 phenols, 81 esters, 8 furans, 16 ketones, 6 sulfides, 26 terpenes, 1 pyrazine, and 1 miscellaneous compound. The mean, standard deviation, minimum, and maximum values of volatile compounds contents are shown in Table 2. Figure 1 shows the overall compositions of volatile components in different types of alcoholic beverages. The contents of volatile components of samples, consisting of three types of alcoholic beverages, were obtained as the mean value of the peak area ratio of the analyte to that of the internal standard. The main components of wine, beer, and *soju* were esters and alcohols, followed by benzene derivatives. In the case of beer, terpenes were a major component, detected at similar levels with alcohols. *Soju* contained similar levels of esters and alcohols. In beer and soju, terpenes were found at higher levels than acids, while acids were detected more than terpenes in wine.

Methanol

The LOD and LOQ values for methanol were calculated to be 1.00 and 3.04 ug/mL, respectively, indicating that the method has sufficient sensitivity. The linearity of calibration curve was confirmed at eight points from 5.00 mg/L to 1.00 g/L by plotting the quantification ion peak ratios of analytes and internal standards. The equation of calibration curve was $y = 0.0373x + 0.183$. The correlation coefficient (r^2), which represents a linearity value, was 0.9999. The accuracy and precision for methanol were in the range of 99.2–102% (101%) and 0.570–1.40% (1.11%), respectively. Figure 2 indicates the distribution of methanol contents in three different types of alcoholic beverages. The methanol contents of alcoholic beverages varied significantly according to the types of alcoholic beverages, with the highest content in wine, followed by *soju* and beer. The methanol contents ranged from 43.6 to 396 mg/L in wine, N.D. to 13.6 mg/L in beer, and 1.45 to 25.6 mg/L in *soju*, illustrating more varied distribution in wine compared to beer and *soju*. Fruit wine is usually associated with high methanol levels [22]. In particular, methanol formation can be affected by the amount of pectin in the grape skins [22]. The detection rate (%) of methanol in wine, beer, and *soju was* 100, 41.7, and 100 (Table 3), respectively.

Higher alcohols

Higher alcohols were the second most abundant volatile compounds in all kinds of alcoholic beverages, which was attributed to the relatively high levels of 3-methylbutan-1-ol; in particular, its content was highest in *soju*. 3-Methylbutan-1-ol can be formed through deamination and decarboxylation reactions from isoleucine during fermentation [30]. Of 33 alcohols detected in alcoholic beverages, 25 alcohols were found in wine, while only less than half were detected in beer (16) and *soju* (14). Wine showed relatively high content of hexan-1-ol, while beer had comparatively high content of linalool. Hexan-1-ol, which contributes to fruity odors [31], can have a positive effect on the aroma of wine [27]. Linalool, which is the most abundant in beer compared to other alcoholic beverages, can be produced by the oxidation of myrcene, which is the main component of many hop varieties [36] during brewing [34]. Propan-1-ol, 2-methylpropan-1-ol, 3-methylbutan-1-ol, hexan-1-ol, heptan-1-ol, butane-2,3-diol, and octan-1-ol were found in all three types of alcoholic beverages. 2-Methylpropan-1-ol is formed from sugar and valin via Ehrlich pathway [18]. 2,3-Butanediol is produced via diacetyl metabolic pathway by yeast [14]. Undecan-2-ol, which is a yeast-derived compound, was only detected in the beer samples.

Table 2 Volatile components identified in three types of alcoholic beverages distributed in Korea

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
Acetals												
ac1	893	1,1-diethoxyethane	0.418 ± 0.966	0	2.90	0.0137 ± 0.0355	0	0.112	2.34 ± 2.51	0	6.49	A
ac2	967	2,4,5-trimethyl-1,3-dioxolane	N.D[v]	0	0	0.0438 ± 0.138	0	0.438	N.D	0	0	B
ac3	1063	1,1-diethoxy-2-methylbutane	N.D	0	0	N.D	0	0	0.0101 ± 0.0319	0	0.101	B
ac4	1109	1-(1-ethoxyethoxy)pentane	N.D	0	0	N.D	0	0	0.0178 ± 0.0563	0	0.178	B
ac5	1110	2-propan-2-yl-1,3-dioxolane	0.0378 ± 0.113	0	0.340	N.D	0	0	N.D	0	0	B
ac6	1303	1,1,3-triethoxypropane	N.D	0	0	N.D	0	0	0.131 ± 0.414	0	1.31	B
Acids												
c1	1453	acetic acid	2.37 ± 1.33	0.343	4.09	0.193 ± 0.264	0	0.879	0.722 ± 0.698	0.0201	1.76	A
c2	1571	2-methylpropanoic acid	N.D	0	0	0.000855 ± 0.00270	0	0.00855	0.0354 ± 0.0878	0	0.281	A
c3	1630	butanoic acid	0.0104 ± 0.0311	0	0.0934	N.D	0	0	0.00136 ± 0.00431	0	0.0136	A
c4	1675	3-methylbutanoic acid	N.D	0	0	0.00949 ± 0.0206	0	0.0583	0.00796 ± 0.0252	0	0.0796	A
c5	1958	heptanoic acid	N.D	0	0	0.0436 ± 0.138	0	0.436	N.D	0	0	A
c6	1853	hexanoic acid	0.143 ± 0.428	0	1.28	0.203 ± 0.350	0	1.07	0.0477 ± 0.108	0	0.323	A
c7	2067	octanoic acid	1.30 ± 1.13	0.349	4.03	2.04 ± 0.983	0.305	3.53	0.0183 ± 0.0295	0	0.0646	A
c8	2158	(2E,4E)-hexa-2,4-dienoic acid	1.00 ± 1.83	0	5.01	N.D	0	0	N.D	0	0	A
c9	2172	nonanoic acid	N.D	0	0	0.0319 ± 0.0602	0	0.193	N.D	0	0	A
c10	2278	decanoic acid	0.450 ± 0.278	0	0.941	0.518 ± 0.529	0	1.43	N.D	0	0	A
c11	2340	dec-9-enoic acid	N.D	0	0	0.0155 ± 0.0490	0	0.155	N.D	0	0	A
c12	2398	4-ethoxy-4-oxobutanoic acid	0.154 ± 0.220	0	0.550	N.D	0	0	N.D	0	0	A
c13	2466	benzoic acid	0.128 ± 0.178	0	0.407	0.00460 ± 0.0146	0	0.0460	0.165 ± 0.197	0	0.576	A
c14	2489	dodecanoicacid	N.D	0	0	0.0201 ± 0.0439	0	0.125	N.D	0	0	A
Alcohols												
a1	1029	butan-2-ol	0.0164 ± 0.0492	0	0.147	N.D	0	0	0.00607 ± 0.0192	0	0.0607	A
a2	1041	propan-1-ol	0.159 ± 0.122	0	0.438	0.0576 ± 0.0303	0.0192	0.120	0.983 ± 0.506	0.360	1.82	A
a3	1098	2-methylpropan-1-ol	1.44 ± 0.763	0.663	3.26	0.425 ± 0.105	0.300	0.600	8.76 ± 4.76	3.20	18.3	A
a4	1110	prop-2-en-1-ol	N.D	0	0	N.D	0	0	0.00763 ± 0.0241	0	0.0763	A
a5	1149	butan-1-ol	3.64 ± 10.7	0	32.1	N.D	0	0	0.187 ± 0.287	0	0.932	A
a6	1215	3-methylbutan-1-ol	33.7 ± 16.7	10.7	68.1	11.5 ± 2.95	8.14	17.0	92.2 ± 58.8	0.0959	216	A
a7	1240	hexan-1-ol	3.48 ± 1.91	0	7.00	0.143 ± 0.148	0.0268	0.511	0.440 ± 0.372	0.0477	1.27	A
a8	1329	4-methylpentan-1-ol	0.00492 ± 0.0107	0	0.0309	N.D	0	0	N.D	0	0	A
a9	1336	heptan-2-ol	0.0147 ± 0.0184	0	0.0439	0.0110 ± 0.0253	0	0.0765	N.D	0	0	A
a10	1343	3-methylpentan-1-ol	0.00904 ± 0.0141	0	0.0358	N.D	0	0	N.D	0	0	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
a11	1376	(E)-hex-3-en-1-ol	0.00271 ± 0.00814	0	0.0244	N.D	0	0	N.D	0	0	B
a12	1385	3-ethoxypropan-1-ol	0.00756 ± 0.0227	0	0.0680	N.D	0	0	0.0159 ± 0.0338	0	0.106	A
a13	1391	(Z)-hex-3-en-1-ol	0.203 ± 0.449	0	1.39	N.D	0	0	N.D	0	0	A
a14	1399	octan-3-ol	0.00146 ± 0.00439	0	0.0132	N.D	0	0	N.D	0	0	A
a15	1410	(E)-hex-4-en-1-ol	0.0169 ± 0.0508	0	0.152	N.D	0	0	N.D	0	0	A
a16	1411	(E)-hex-2-en-1-ol	0.0117 ± 0.0350	0	0.105	N.D	0	0	N.D	0	0	A
a17	1422	cyclohexanol	0.00474 ± 0.0100	0	0.0281	N.D	0	0	N.D	0	0	A
a18	1434	4-methylhexan-1-ol	N.D	0	0	N.D	0	0	0.0104 ± 0.0328	0	0.104	B
a19	1459	oct-1-en-3-ol	N.D	0	0	0.0716 ± 0.153	0	0.493	N.D	0	0	A
a20	1464	heptan-1-ol	0.0547 ± 0.0899	0	0.234	0.0778 ± 0.140	0	0.456	0.00872 ± 0.0276	0	0.0872	A
a21	1471	6-methylhept-5-en-2-ol	N.D	0	0	0.00564 ± 0.0178	0	0.0564	N.D	0	0	A
a22	1496	2-ethylhexan-1-ol	0.0804 ± 0.0734	0	0.235	N.D	0	0	0.0622 ± 0.0693	0	0.161	A
a23	1525	nonan-2-ol	N.D	0	0	0.270 ± 0.360	0	0.889	N.D	0	0	A
a24	1547	butane-2,3-diol	0.0776 ± 0.103	0	0.270	1.83 ± 5.79	0	18.3	0.0153 ± 0.0484	0	0.153	A
a25	1553	2,6-dimethylheptan-4-ol	0.0272 ± 0.0817	0	0.245	N.D	0	0	0.218 ± 0.668	0	2.12	A
a26	1555	3,7-dimethylocta-1,6-dien-3-ol (Linalool)	0.226 ± 0.658	0	1.98	2.54 ± 4.28	0.121	15.6	N.D	0	0	A
a27	1567	octan-1-ol	0.169 ± 0.240	0	0.770	0.783 ± 2.18	0	7.68	0.0222 ± 0.0467	0	0.136	A
a28	1625	decan-2-ol	N.D	0	0	0.0624 ± 0.197	0	0.624	N.D	0	0	A
a29	1635	2-(2-ethoxyethoxy)ethanol	N.D	0	0	0.00936 ± 0.0296	0	0.0936	N.D	0	0	A
a30	1670	nonan-1-ol	0.436 ± 0.440	0	1.03	N.D	0	0	N.D	0	0	A
a31	1693	(Z)-non-3-en-1-ol	0.0304 ± 0.0912	0	0.274	N.D	0	0	N.D	0	0	A
a32	1727	undecan-2-ol	N.D	0	0	0.192 ± 0.320	0	0.910	N.D	0	0	A
a33	1774	decan-1-ol	0.0118 ± 0.0355	0	0.106	0.0473 ± 0.100	0	0.257	N.D	0	0	A
Aldehydes												
d1	700	acetaldehyde	0.312 ± 0.839	0	2.55	0.0404 ± 0.0819	0	0.267	0.0785 ± 0.0480	0.0182	0.157	A
d2	840	prop-2-enal	N.D	0	0	N.D	0	0	0.00606 ± 0.0192	0	0.0606	B
d3	913	3-methylbutanal	N.D	0	0	N.D	0	0	0.0687 ± 0.102	0	0.250	A
d4	1050	(E)-but-2-enal	0.0394 ± 0.118	0	0.355	N.D	0	0	0.0270 ± 0.0704	0	0.223	A
d5	1076	hexanal	N.D	0	0	N.D	0	0	0.123 ± 0.311	0	0.979	A
d6	1085	(E)–2-methylbut-2-enal	N.D	0	0	N.D	0	0	0.0369 ± 0.117	0	0.369	A
d7	1185	3-ethoxypropanal	N.D	0	0	N.D	0	0	0.0986 ± 0.312	0	0.986	B
d8	1394	nonanal	0.0136 ± 0.0325	0	0.0976	0.0181 ± 0.0435	0	0.135	0.0495 ± 0.113	0	0.362	A
d9	2058	2-methylbutanal	N.D	0	0	N.D	0	0	0.0137 ± 0.0434	0	0.137	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
Amides												
n1	1392	N,2-dimethylpropanamide	N.D	0	0	0.00249 ± 0.00786	0	0.0249	N.D	0	0	C
n2	1454	N,N-dimethylpropanamide	0.0151 ± 0.0454	0	0.136	N.D	0	0	N.D	0	0	A
Benzenederivatives												
bn1	1031	toluene	0.0385 ± 0.0539	0	0.143	0.0283 ± 0.0246	0	0.0652	0.190 ± 0.321	0	1.04	A
bn2	1118	ethylbenzene	N.D	0	0	N.D	0	0	0.0268 ± 0.0846	0	0.268	A
bn3	1392	ethyl benzoate	N.D	0	0	0.407 ± 1.27	0	4.03	N.D	0	0	C
bn4	1441	1,2,4,5-tetramethylbenzene (Durene)	4.09 ± 12.3	0	36.8	N.D	0	0	N.D	0	0	A
bn5	1529	benzaldehyde	0.758 ± 1.89	0	5.75	0.0144 ± 0.0278	0	0.0855	0.337 ± 0.483	0	1.59	A
bn6	1658	2-phenylacetaldehyde	N.D	0	0	0.00501 ± 0.0125	0	0.0392	0.0544 ± 0.143	0	0.454	A
bn7	1732	1,1,6-trimethyl-2H-naphthalene	0.0394 ± 0.0616	0	0.171	N.D	0	0	N.D	0	0	B
bn8	1779	naphthalene	N.D	0	0	N.D	0	0	0.0159 ± 0.0503	0	0.159	A
bn9	1812	methyl 2-hydroxybenzoate	0.0442 ± 0.132	0	0.397	0.138 ± 0.322	0	0.982	N.D	0	0	A
bn10	1903	phenylmethanol	0.0913 ± 0.210	0	0.632	N.D	0	0	N.D	0	0	A
bn11	1943	2-phenylethanol	13.3 ± 8.48	5.68	31.6	5.53 ± 2.65	2.39	11.6	8.35 ± 5.77	0.651	17.8	A
bn12	2042	(E)–3-phenylprop-2-enal	N.D	0	0	N.D	0	0	0.686 ± 1.55	0	4.67	A
bn13	2284	methyl 2-aminobenzoate	0.0247 ± 0.0430	0	0.122	N.D	0	0	N.D	0	0	A
Phenols												
ph1	1888	2-methoxyphenol (Guaiacol)	N.D	0	0	0.00372 ± 0.0118	0	0.0372	N.D	0	0	A
ph2	1927	2,6-ditert-butyl-4-methylphenol	0.0487 ± 0.0785	0	0.192	0.0132 ± 0.0291	0	0.0836	0.412 ± 0.979	0	3.14	A
ph3	2025	phenol	N.D	0	0	0.00686 ± 0.0217	0	0.0686	N.D	0	0	A
ph4	2226	4-ethenyl-2-methoxyphenol	N.D	0	0	0.0684 ± 0.0831	0	0.247	0.0109 ± 0.0238	0	0.0678	A
ph5	2372	4-ethenylphenol	N.D	0	0	0.0200 ± 0.0634	0	0.200	N.D	0	0	A
Esters												
e1	813	methyl acetate	0.0166 ± 0.0201	0	0.0419	N.D	0	0	N.D	0	0	A
e2	870	ethyl acetate	22.7 ± 13.8	4.95	46.2	3.82 ± 2.01	0.854	7.01	29.2 ± 47.1	2.83	155	A
e3	932	propyl acetate	0.0385 ± 0.0516	0	0.139	0.0507 ± 0.0730	0	0.232	0.0563 ± 0.0907	0	0.199	A
e4	954	ethyl propanoate	0.145 ± 0.202	0	0.574	0.0749 ± 0.0741	0.0277	0.272	0.110 ± 0.281	0	0.900	A
e5	959	ethyl 2-methylpropanoate	0.135 ± 0.143	0	0.483	0.0845 ± 0.146	0	0.466	0.0352 ± 0.0820	0	0.250	A
e6	985	butyl acetate	N.D	0	0	0.0127 ± 0.0321	0	0.101	N.D	0	0	A
e7	988	2-methylpropyl acetate	0.112 ± 0.111	0	0.270	0.181 ± 0.166	0	0.479	0.389 ± 0.507	0	1.52	A
e8	1040	ethyl butanoate	0.654 ± 0.267	0.291	1.04	0.720 ± 0.263	0.344	1.18	0.331 ± 0.500	0	1.67	A
e9	1048	ethyl 2-methylbutanoate	0.0691 ± 0.0819	0	0.234	0.0317 ± 0.0745	0	0.240	0.00800 ± 0.0253	0	0.0800	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
e10	1050	propyl butanoate	N.D	0	0	0.0183 ± 0.0579	0	0.183	N.D	0	0	A
e11	1053	ethyl3-methylbutanoate	0.167 ± 0.147	0	0.434	0.0372 ± 0.0715	0	0.229	0.0432 ± 0.136	0	0.432	A
e12	1078	2-methylpropyl propanoate	N.D	0	0	0.0349 ± 0.0683	0	0.209	N.D	0	0	A
e13	1092	2-methylpropyl2-methylpropanoate	N.D	0	0	0.789 ± 1.63	0	5.07	N.D	0	0	A
e14	1092	2-methylpropyl 2-methylbutanoate	N.D	0	0	0.105 ± 0.206	0	0.620	N.D	0	0	B
e15	1127	3-methylbutyl acetate	4.37 ± 1.57	1.02	6.35	17.0 ± 11.2	3.68	35.3	27.0 ± 52.4	0.100	172	A
e16	1134	ethyl pentanoate	N.D	0	0	0.0531 ± 0.0625	0	0.195	0.0139 ± 0.0441	0	0.139	A
e17	1159	ethyl (E)-but-2-enoate	1.26 ± 1.27	0	3.35	N.D	0	0	N.D	0	0	B
e18	1176	3-methylbutyl propanoate	N.D	0	0	0.558 ± 1.29	0	4.10	N.D	0	0	A
e19	1177	methyl hexanoate	0.0114 ± 0.0343	0	0.103	N.D	0	0	N.D	0	0	A
e20	1184	2-methylpropyl 3-methylbutanoate	N.D	0	0	0.0322 ± 0.102	0	0.322	N.D	0	0	A
e21	1188	pentyl propanoate	N.D	0	0	N.D	0	0	0.00833 ± 0.0263	0	0.0833	A
e22	1193	pentyl 2-methylpropanoate	N.D	0	0	0.410 ± 0.885	0	2.45	N.D	0	0	B
e23	1195	2-methylbutyl 2-methylpropanoate	N.D	0	0	2.05 ± 4.88	0	15.7	N.D	0	0	A
e24	1196	3-methylbutyl 2-methylpropanoate	N.D	0	0	3.39 ± 10.3	0	32.7	N.D	0	0	A
e25	1234	ethyl hexanoate	22.1 ± 11.8	7.40	39.1	11.8 ± 5.38	1.80	19.3	9.13 ± 11.7	0.0356	39.4	A
e26	1247	ethyl 2-oxopropanoate	0.00882 ± 0.0265	0	0.0794	0.00524 ± 0.0166	0	0.0524	N.D	0	0	A
e27	1251	hexyl acetate	0.457 ± 0.837	0	2.66	0.256 ± 0.219	0	0.750	0.0225 ± 0.0474	0	0.116	A
e28	1264	3-methylbutyl butanoate	N.D	0	0	0.00434 ± 0.0137	0	0.0434	N.D	0	0	A
e29	1265	heptan-2-yl acetate	N.D	0	0	0.0371 ± 0.117	0	0.371	N.D	0	0	B
e30	1277	3-methylbutyl 2-methylbutanoate	N.D	0	0	0.0337 ± 0.106	0	0.337	N.D	0	0	A
e31	1281	2-methylbutyl 2-methylbutanoate	N.D	0	0	0.252 ± 0.486	0	1.54	N.D	0	0	A
e32	1486	3-methylbutyl 3-methylbutanoate	N.D	0	0	0.294 ± 0.621	0	1.55	N.D	0	0	A
e33	1296	2-methylbutyl 3-methylbutanoate	N.D	0	0	0.258 ± 0.813	0	2.57	N.D	0	0	A
e34	1296	pentyl 3-methylbutanoate	N.D	0	0	0.0244 ± 0.0651	0	0.206	N.D	0	0	B
e35	1301	ethyl (E)-hex-3-enoate	N.D	0	0	0.112 ± 0.288	0	0.910	N.D	0	0	A
e36	1343	methyl 4-methylhex-2-enoate	N.D	0	0	0.0146 ± 0.0461	0	0.146	N.D	0	0	B
e37	1345	ethyl heptanoate	0.114 ± 0.0886	0	0.235	0.993 ± 1.90	0	6.15	9.42 ± 28.9	0	91.7	A
e38	1420	hexyl butanoate	N.D	0	0	0.00470 ± 0.0149	0	0.0470	N.D	0	0	A
e39	1354	hexyl 2-methylpropanoate	N.D	0	0	0.0206 ± 0.0651	0	0.206	N.D	0	0	B
e40	1355	ethyl 2-hydroxypropanoate	3.35 ± 3.16	0	10.2	0.134 ± 0.424	0	1.34	3.01 ± 5.05	0	13.3	A
e41	1360	ethyl (E)-hex-2-enoate	N.D	0	0	0.00494 ± 0.0156	0	0.0494	N.D	0	0	B
e42	1347	hexyl formate	0.932 ± 2.79	0	8.38	N.D	0	0	N.D	0	0	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
e43	1381	heptyl acetate	N.D	0	0	0.0518 ± 0.0724	0	0.243	N.D	0	0	A
e44	1384	ethyl 3-ethoxypropanoate	N.D	0	0	N.D	0	0	0.0432 ± 0.137	0	0.432	A
e45	1386	oct-1-en-3-yl acetate	N.D	0	0	0.0105 ± 0.0332	0	0.105	N.D	0	0	A
e46	1389	octyl acetate	N.D	0	0	0.0189 ± 0.0573	0	0.182	N.D	0	0	A
e47	1393	methyl octanoate	0.195 ± 0.212	0	0.638	N.D	0	0	N.D	0	0	A
e48	1403	ethyl 2-hydroxy-3-methylbutanoate	N.D	0	0	N.D	0	0	0.0123 ± 0.0388	0	0.123	B
e49	1441	ethyl octanoate	50.9 ± 47.0	0	142	26.0 ± 24.6	0.00961	63.8	19.9 ± 18.7	0.104	53.7	A
e50	1451	3-methylbutyl hexanoate	N.D	0	0	N.D	0	0	2.22 ± 7.02	0	22.2	A
e51	1505	ethyl (2E,4E)-hexa-2,4-dienoate	0.965 ± 1.87	0	5.74	N.D	0	0	N.D	0	0	A
e52	1525	ethyl 3-hydroxybutanoate	0.133 ± 0.148	0	0.380	N.D	0	0	N.D	0	0	A
e53	1543	ethyl nonanoate	0.0952 ± 0.129	0	0.364	0.474 ± 0.751	0	2.44	0.686 ± 0.811	0	2.38	A
e54	1551	ethyl 2-hydroxy-4-methylpentanoate	0.0267 ± 0.0577	0	0.166	N.D	0	0	N.D	0	0	A
e55	1555	ethyl 2-hydroxyethyl hexanoate	N.D	0	0	N.D	0	0	0.0462 ± 0.101	0	0.306	C
e56	1561	2-methylpropyl octanoate	N.D	0	0	0.00656 ± 0.0207	0	0.0656	0.0674 ± 0.213	0	0.674	A
e57	1575	(5-methyl-2-propan-2-ylcyclohexyl)acetate	0.531 ± 1.59	0	4.78	N.D	0	0	N.D	0	0	A
e58	1578	3-methylbutyl 2-hydroxypropanoate	0.0338 ± 0.0834	0	0.251	N.D	0	0	0.00699 ± 0.0221	0	0.0699	A
e59	1600	[(1S,2R,4S)−1,7,7-trimethyl-2-bicyclo[2.2.1] heptanyl] acetate	N.D	0	0	N.D	0	0	0.000572 ± 0.00181	0	0.00572	B
e60	1650	ethyl decanoate	15.1 ± 11.8	3.40	36.4	9.15 ± 11.0	0.322	29.6	14.9 ± 22.4	0	73.9	A
e61	1670	3-methylbutyl octanoate	0.0314 ± 0.0941	0	0.282	0.211 ± 0.257	0	0.810	0.149 ± 0.437	0	1.39	A
e62	1675	ethyl (E)-dec-4-enoate	N.D	0	0	0.653 ± 1.20	0	3.71	0.00832 ± 0.0263	0	0.0832	A
e63	1675	(E)-dec-4-enoate	0.0513 ± 0.154	0	0.462	N.D	0	0	N.D	0	0	A
e64	1667	ethyl (Z)-dec-4-enoate	N.D	0	0	N.D	0	0	0.0350 ± 0.111	0	0.350	B
e65	1688	diethyl butanedioate	9.07 ± 5.05	2.67	15.6	0.0618 ± 0.183	0	0.582	2.74 ± 2.89	0.483	8.57	A
e66	1697	ethyl dec-9-enoate	0.199 ± 0.452	0	1.38	0.181 ± 0.422	0	1.35	0.00464 ± 0.0147	0	0.0464	B
e67	1706	methyl (2E)−3,7-dimethylocta-2,6-dienoate	N.D	0	0	1.09 ± 2.19	0	6.79	N.D	0	0	B
e68	1733	(1-butoxy-1-oxopropan-2-yl) butanoate	0.0125 ± 0.0374	0	0.112	N.D	0	0	N.D	0	0	A
e69	1750	ethyl undecanoate	0.00919 ± 0.0276	0	0.0827	N.D	0	0	N.D	0	0	A
e70	1812	ethyl 2-phenylacetate	0.165 ± 0.218	0	0.686	N.D	0	0	0.0646 ± 0.103	0	0.258	A
e71	1841	2-phenylethylacetate	0.166 ± 0.159	0	0.476	0.731 ± 1.20	0	3.52	0.472 ± 0.566	0	1.65	A
e72	1844	2-phenylethyl propanoate	N.D	0	0	0.0300 ± 0.0950	0	0.300	N.D	0	0	A
e73	1853	ethyl dodecanoate	2.07 ± 1.63	0	5.23	1.58 ± 2.53	0	5.75	0.934 ± 1.26	0	4.04	A
e74	1878	3-methylbutyl decanoate	0.0265 ± 0.0796	0	0.239	0.00563 ± 0.0178	0	0.0563	N.D	0	0	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
e75	1915	ethyl 3-phenylpropanoate	N.D	0	0	N.D	0	0	0.00309 ± 0.00976	0	0.0309	A
e76	2058	ethyl tetradecanoate	0.133 ± 0.126	0	0.308	0.116 ± 0.223	0	0.561	0.700 ± 1.31	0	4.38	A
e77	2172	ethyl(E)−3-phenylprop-2-enoate	0.00853 ± 0.0256	0	0.0768	N.D	0	0	N.D	0	0	A
e78	2261	ethyl hexadecanoate	0.488 ± 0.262	0.0713	0.840	0.327 ± 0.534	0	1.75	0.285 ± 0.224	0	0.589	A
e79	2267	ethyl (E)-hexadec-9-enoate	N.D	0	0	0.0155 ± 0.0489	0	0.155	N.D	0	0	B
e80	2415	2-phenylethyl octanoate	N.D	0	0	0.0134 ± 0.0424	0	0.134	N.D	0	0	A
e81	2466	ethyl octadecanoate	N.D	0	0	0.0192 ± 0.0435	0	0.134	N.D	0	0	A
Furans												
f1	893	2-methylfuran	N.D	0	0	0.00694 ± 0.0149	0	0.0412	N.D	0	0	A
f2	946	2,5-dimethylfuran	N.D	0	0	0.0213 ± 0.0282	0	0.0795	N.D	0	0	A
f3	1063	2-ethenylfuran	N.D	0	0	0.00225 ± 0.00713	0	0.0225	N.D	0	0	B
f4	1230	2-pentylfuran	N.D	0	0	N.D	0	0	0.260 ± 0.821	0	2.60	A
f5	1422	3-(4-methylpent-3-enyl)furan	N.D	0	0	0.0867 ± 0.182	0	0.594	N.D	0	0	B
f6	1468	furan-2-carbaldehyde (Furfural)	0.293 ± 0.434	0	1.41	0.00997 ± 0.0315	0	0.0997	1.43 ± 2.54	0	6.85	A
f7	1539	2,6,6-trimethyl-10-methylidene-1-oxaspiro[4.5]dec-8-ene	0.202 ± 0.337	0	0.873	N.D	0	0	N.D	0	0	B
Ketones												
k1	810	propan-2-one	0.00156 ± 0.00468	0	0.0140	0.00190 ± 0.00423	0	0.0124	0.000879 ± 0.00278	0	0.00879	A
k2	899	butan-2-one	N.D	0	0	0.00270 ± 0.00855	0	0.0270	N.D	0	0	A
k3	976	pentan-2-one	0.0485 ± 0.146	0	0.437	0.00790 ± 0.0250	0	0.0790	N.D	0	0	A
k4	974	butane-2,3-dione	0.00786 ± 0.0236	0	0.0707	N.D	0	0	0.00682 ± 0.0216	0	0.0682	A
k5	994	2-methylpentan-3-one	N.D	0	0	0.00829 ± 0.00901	0	0.0192	N.D	0	0	A
k6	1004	4-methylpentan-2-one	N.D	0	0	0.0489 ± 0.0958	0	0.232	N.D	0	0	A
k7	1177	heptan-2-one	N.D	0	0	0.00478 ± 0.0151	0	0.0478	N.D	0	0	A
k8	1280	3-hydroxybutan-2-one (Acetoin)	0.143 ± 0.264	0	0.775	0.0128 ± 0.0404	0	0.128	0.0460 ± 0.128	0	0.409	A
k9	1282	octan-2-one	N.D	0	0	0.0144 ± 0.0320	0	0.0937	N.D	0	0	A
k10	1391	nonan-2-one	N.D	0	0	0.378 ± 1.18	0	3.72	0.00729 ± 0.0178	0	0.0555	A
k11	1497	decan-2-one	N.D	0	0	0.204 ± 0.429	0	1.27	N.D	0	0	A
k12	1605	undecan-2-one	0.0531 ± 0.159	0	0.478	0.742 ± 1.16	0	3.32	0.0518 ± 0.164	0	0.518	A
k13	1670	1-phenylethanone	N.D	0	0	N.D	0	0	0.0167 ± 0.0365	0	0.104	A
k14	1825	tridecan-2-one	N.D	0	0	0.0497 ± 0.123	0	0.387	N.D	0	0	A
k15	2006	1-(1H-pyrrol-2-yl)ethanone	N.D	0	0	0.00397 ± 0.0126	0	0.0397	N.D	0	0	A
k16	2458	chromen-2-one (Coumarin)	N.D	0	0	N.D	0	0	0.0233 ± 0.0737	0	0.233	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
Sulfides												
su1	740	methylsulfanylmethane	0.001000 ± 0.00300	0	0.00900	0.0137 ± 0.0194	0	0.0526	N.D	0	0	A
su2	963	ethyl 5-methylthiadiazole-4-carboxylate	N.D	0	0	N.D	0	0	0.0187 ± 0.0592	0	0.187	C
su3	1065	(methyldisulfanyl)methane	N.D	0	0	0.00916 ± 0.0225	0	0.0702	0.0228 ± 0.0722	0	0.228	A
su4	1083	2-methylthiophene	0.0298 ± 0.0895	0	0.268	N.D	0	0	N.D	0	0	A
su5	1558	ethyl 3-methylsulfanylpropanoate	N.D	0	0	N.D	0	0	0.0180 ± 0.0570	0	0.180	A
su6	1738	3-methylsulfanylpropan-1-ol (Methionol)	0.111 ± 0.185	0	0.552	N.D	0	0	0.0276 ± 0.0873	0	0.276	A
Terpenes												
t1	1054	2,2-dimethyl-3-methylidenebicyclo[2.2.1] heptane (Camphene)	0.0214 ± 0.0641	0	0.192	N.D	0	0	N.D	0	0	A
t2	1154	2-methyl-5-propan-2-ylcyclohexa-1,3-diene (α-Phellandrene)	N.D	0	0	0.0414 ± 0.0747	0	0.192	N.D	0	0	A
t3	1158	7-methyl-3-methylideneocta-1,6-diene (Myrcene)	N.D	0	0	12.9 ± 30.0	0	96.7	N.D	0	0	A
t4	1171	1-methyl-4-propan-2-ylcyclohexa-1,3-diene (α-Terpinene)	0.0192 ± 0.0576	0	0.173	N.D	0	0	N.D	0	0	A
t5	1191	1-methyl-4-prop-1-en-2-ylcyclohexene (Limonene)	0.101 ± 0.190	0	0.546	0.0864 ± 0.239	0	0.765	0.404 ± 1.22	0	3.88	A
t6	1199	1,3,3-trimethyl-2-oxabicyclo[2.2.2]octane (Eucalyptol)	0.142 ± 0.298	0	0.921	N.D	0	0	N.D	0	0	A
t7	1249	styrene	0.545 ± 1.18	0	3.56	6.41 ± 10.2	0	32.4	0.0188 ± 0.0593	0	0.188	A
t8	1249	(3E)−3,7-dimethylocta-1,3,6-triene (Ocimene)	N.D	0	0	0.0537 ± 0.170	0	0.537	N.D	0	0	A
t9	1282	1-methyl-4-propan-2-ylcyclohexa-1,4-diene (γ-Terpinene)	N.D	0	0	0.00965 ± 0.0305	0	0.0965	N.D	0	0	A
t10	1476	(1R,2S,6S,7S,8S)−1,3-dimethyl-8-propan-2-yltricyclo[4.4.0.02,7]dec-3-ene (Copaene)	N.D	0	0	N.D	0	0	0.502 ± 1.59	0	5.02	B
t11	1541	(4R)−1-methyl-4-propan-2-ylcyclohexene	0.0291 ± 0.0874	0	0.262	N.D	0	0	N.D	0	0	C
t12	1599	(1S,2S,4R)−1-ethenyl-1-methyl-2,4-bis(prop-1-en-2-yl) cyclohexane (β-Elemene)	N.D	0	0	N.D	0	0	0.0331 ± 0.105	0	0.331	A
t13	1615	(1R,4E,9S)−4,11,11-trimethyl-8-methylidenebicyclo[7.2.0] undec-4-ene (Caryophyllene)	N.D	0	0	0.463 ± 1.43	0	4.54	0.0718 ± 0.162	0	0.482	A
t14	1620	4-methyl-1-propan-2-ylcyclohex-3-en-1-ol (4-Terpineol)	0.116 ± 0.165	0	0.437	N.D	0	0	N.D	0	0	A
t15	1631	5-methyl-2-propan-2-ylcyclohexan-1-ol (Menthol)	0.145 ± 0.434	0	1.30	N.D	0	0	N.D	0	0	A
t16	1695	(1E,4E,8E)−2,6,6,9-tetramethylcycloundeca-1,4,8-triene (Humulene)	N.D	0	0	1.11 ± 3.10	0	9.86	0.0463 ± 0.147	0	0.463	A
t17	1715	2-(4-methylcyclohex-3-en-1-yl)propan-2-ol (Terpineol)	0.194 ± 0.459	0	1.38	N.D	0	0	N.D	0	0	A
t18	1770	1-methyl-4-(6-methylhept-5-en-2-yl)benzene (Curcumene)	N.D	0	0	N.D	0	0	0.0297 ± 0.0939	0	0.297	B
t19	1774	3,7-dimethyloct-6-en-1-ol (Citronellol)	N.D	0	0	0.141 ± 0.244	0	0.629	N.D	0	0	A

Table 2 (continued)

No[i]	RI[ii]	Volatilecompounds	Wine			Beer			Soju			ID[iii]
			Average[iv]	Min	Max	Average	Min	Max	Average	Min	Max	
t20	1785	(1S,8aR)−4,7-dimethyl-1-propan-2-yl-1,2,3,5,6,8a-hexahydronaphthalene (D-Cadinene)	N.D	0	0	0.0467 ± 0.148	0	0.467	N.D	0	0	A
t21	1819	(6,6-dimethyl-2-bicyclo[3.1.1]hept-2-enyl)methanol (Myrtenol)	N.D	0	0	N.D	0	0	0.00116 ± 0.00368	0	0.0116	A
t22	1841	(1S,4S)−1,6-dimethyl-4-propan-2-yl-1,2,3,4-tetrahydronaphthalene (Calamenene)	0.0281 ± 0.0488	0	0.119	0.0602 ± 0.111	0	0.329	0.999 ± 2.60	0	8.29	B
t23	1876	2,5-dimethyl-8-propan-2-yl-1,2,3,4-tetrahydronaphthalene (Calamene)	N.D	0	0	N.D	0	0	0.0497 ± 0.136	0	0.433	B
t24	1915	(1S)−4,7-dimethyl-1-propan-2-yl-1,2-dihydronaphthalene (α-Calacorene)	N.D	0	0	N.D	0	0	0.00658 ± 0.0208	0	0.0658	B
t25	2024	(6E)−3,7,11-trimethyldodeca-1,6,10-trien-3-ol (Nerolidol)	N.D	0	0	0.00228 ± 0.00720	0	0.0228	N.D	0	0	A
t26	2275	1,6-dimethyl-4-propan-2-ylnaphthalene (Cadalene)	N.D	0	0	N.D	0	0	0.0738 ± 0.138	0	0.417	B
Pyrazines												
p1	1408	2-ethyl-3-methylpyrazine	0.00325 ± 0.00975	0	0.0292	N.D	0	0	N.D	0	0	A
Miscellaneous												
mc1	1344	3-ethenoxyprop-1-ene	N.D	0	0	0.00309 ± 0.00976	0	0.0309	N.D	0	0	B

[i] All volatile components are listed by the order of their RI values in chemical class

[ii] Retention indices were determined using n-alkanes C5–C30 as external standards

[iii] Identification of the compounds was based on the following: A, the mass spectrum and retention index agreed with those of the authentic compounds under the same conditions (positive identification); B, the mass spectrum and retention index were consistent with those from the NIST database (tentative identification); and C, the mass spectrum was consistent with that of W9N08 (Wiley and NIST) and manual interpretation (tentative identification)

[iv] Mean values of the relative peak area to that of the internal standard ± standard deviation

[V] N.D. = not detected

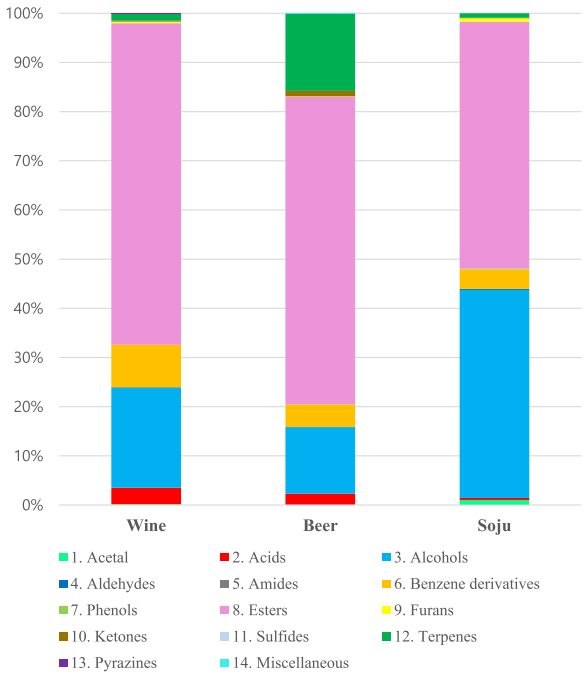


Fig. 1 Relative compositions of identified volatile components in the three types of alcoholic beverages according to chemical functional groups. Each stacked bar is 100% wide

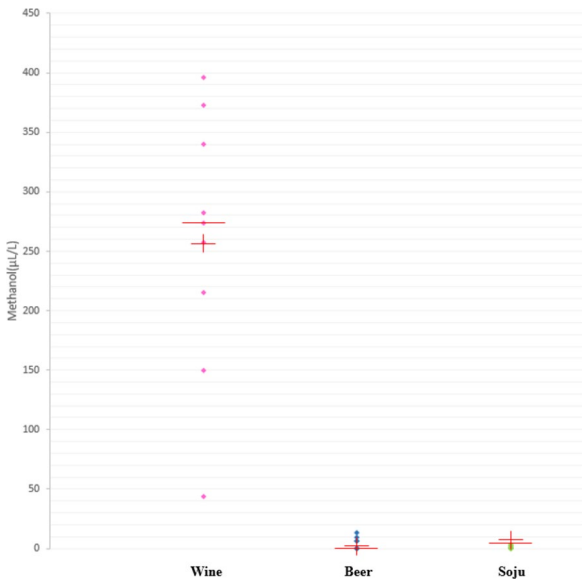


Fig. 2 Distribution of methanol contents in the three types of alcoholic beverages. Each dot corresponds to the samples. The cross indicates the mean concentration of the compound and the line indicates the median concentration of the compound

Other volatile compounds

Esters were the most dominant group of volatile compounds in all kinds of alcoholic beverages with significant amounts of ethyl acetate, 3-methylbutyl acetate (isoamyl acetate), ethyl hexanoate, ethyl octanoate, and ethyl decanoate. Those ester compounds found in all types of alcoholic beverages were 2-methylpropyl acetate, hexyl acetate, diethyl butanedioate, and ethyl hexadecanoate. Ethyl acetate is a typical metabolite of yeast [5]. The contents of diethyl succinate were higher in wine compared to other alcoholic beverages. Butanedioic acid, an organic acid produced by yeast in the early stages of alcoholic fermentation [40], has been shown to be involved in the formation of this ester compound [10]. Terpenes were major volatile compounds in beer, while their contents were low in wine and *soju*. In particular, in beer, terpenes showed the similar levels with higher alcohols. Terpenes can be biosynthesized only by higher plants, algae, fungi, and some yeasts such as *Kluyveromyceslactis, Torulasporadelbrueckii* (*Saccharomyces fermentanti*) and *Ambrosiozymamonospora* [19]. They are initiated by condensation of three molecules of acetyl-CoA. Further reactions lead to the formation of geranyl pyrophosphate (GPP), a precursor for monoterpenes. In addition, sesquiterpenes and diterpenes are synthesized from farnesyl diphosphate (FPP) and geranylgeranyl diphosphate (GGPP), respectively [19]. The content of terpenes was high due to the high contents of myrcene, styrene and humulene in beer samples. Myrcene, which is synthesized by the mevalonate (MVA) pathway in the cytoplasm of yeast [34], is the most abundant component of hop oil, comprising over 80%. Styrene, which can be formed by bacteria, fungi, and microbial metabolism as well as metabolism of raw materials [2, 26, 39], is derived from phenylalanine [16]. This compound is generally recognized as safe, but can cause off-flavor at detectable levels [3]. Humulene is derived from 2-C-methyl-D-erythritol 4-phosphate (MEP) pathway in plastids of hops [34]. On the other hand, propan-2-one, 3-hydroxybutan-2-one, and undecane-2-one were the only ketones detected in all types of alcoholic beverages. Benzaldehyde and 2-phenylethanol were also detected in all alcoholic beverages. In particular, 2-phenylethanol is a metabolite formed through the catabolism of phenylalanine via the Ehrlich pathway, mainly contributing to the desirable floral-like aromas [23, 30, 42]. Benzaldehyde is primarily formed by the action of microorganisms on aromatic amino acids or by the oxidation of benzyl alcohol [46]. Acetal, detected in all types of alcoholic beverages, can be synthesized by the reaction of aldehydes with alcohols during fermentation and ripening, which can affect the sensory properties of alcoholic beverages [37, 41]. Diacetyl was found at small amounts in wine and *soju*, while, 2-undecanone, well-known as a component of hop oil, was most abundant in beer compared to other alcoholic beverages [17]. 1-Phenylethanone and chromen-2-one (coumarin) were found

Table 3 Concentrations of methanol in three types of alcoholic beverages

Standard curve			Linearity(R^2)	LOD	LOQ
y = 0.0373x + 0.1834			0.9999	1.00	3.04
Concentration (mg/L)[1)]					
wine		beer		Soju	
w1	274 ± 0.437	b1	N.D	s1	5.10 ± 1.34
w2	396 ± 0.708	b2	N.D	s2	25.6 ± 0.252
w3	373 ± 2.76	b3	6.31 ± 0.717	s3	12.2 ± 2.25
w4	150 ± 3.46	b4	13.6 ± 0.892	s4	21.8 ± 1.14
w5	340 ± 2.67	b5	N.D	s5	trace
w6	257 ± 4.58	b6	N.D	s6	trace
w7	282 ± 4.17	b7	6.08 ± 0.317	s7	6.91 ± 1.11
w8	215 ± 2.13	b8	N.D	s8	11.5 ± 2.22
w9	43.6 ± 3.79	b9	N.D	s9	trace
		b10	9.65 ± 1.21	s10	trace
Detection rate (%)					
100		41.7		100	

[1] Mean values ± standard deviation

only in *soju* samples. Coumarin is a secondary metabolite originating from the phenylpropanoid pathway via cinnamic acid [43]. Among aldehydes, nonanal was detected in all three types of alcoholic beverages. 2-Methylfuran, 2,5-dimethylfuran, and 3-(4-methylpent-3-enyl)furan were observed only in beer. 2-Methylfuran, which contributes to the sweet chocolate aroma, has been known to be found in brewing barley [7, 38]. 2,5-Dimethylfuran and 3-(4-methylpent-3-enyl)furan have also been known to be detected in beer [32, 38]. 2-Ethyl-3-methylpyrazine was found in one sample of wine. Pyrazines, which are considered as a group of volatile heterocyclic nitrogen-containing compounds, are regarded as crucial flavor compounds found in various foods [47]. In particular, pyrazines play an important role in the flavor of cereal brewed beers [33]. Pyrazine production levels are chosen as indicators of sensory food quality, primarily arising from the non-enzymatic Maillard reaction between amino acids and reducing sugars during heating. Ren et al. reported that monitoring pyrazine levels in wine is crucial because, even at low concentrations, it can organically affect wine quality. Pyrazine derivatives can be classified into three types, including natural pyrazines, alkyl pyrazines, and pyrazines produced by microorganisms [33]. In wine, most monocyclic alkylpyrazines are produced by enzymatic reactions mediated by microorganisms [33]. Compared to other alcoholic beverages, *soju* had higher content of furans, which was attributed to its relatively high levels of furfural. Furfural, which had a high content in *soju*, is mainly produced by the pyrolysis of polypentose in the hulls of grains [44] and is generated in the later stages of distillation [20]. This compound has the sweet aroma note of almonds [44]. Methionol was detected in all alcoholic beverages, except for beer. It is a compound derived from the catabolism of methionine via the Ehrlich pathway during fermentation and usually contributes to the off-flavor [8].

Principal component analysis

In Fig. 3, the first three principal components (PCs) explained 50.8% of the variance 22.1%, 17.0%, and 11.7% for PC1, PC2, and PC3, respectively. Those compounds responsible for the discrimination of *soju* and beer can

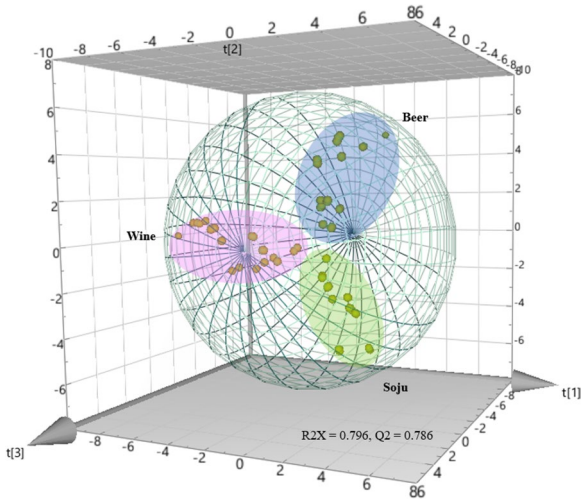


Fig. 3 PCA 3D score plots of volatile compounds of wine, beer, and *soju*. Each color indicates as follows: Pink, wine; Blue, beer; Green, *soju*

be attributed to some alcohol components, such as propan-1-ol, 2-methylpropan-1-ol, and 3-methylbutan-1-ol, and acids, such as octanoic acid, in the negative direction of PC1. Wine was distinguished from other alcoholic beverages by some esters, such as methyl acetate, ethyl (E)-but-2-enoate, ethyl 3-hydroxybutanoate, and diethyl butanedioate, and alcohols, such as hexan-1-ol, nonan-2-ol, and nonan-1-ol, in the positive direction of PC2.

A simultaneous analytical method for methanol, fusel alcohols, and other volatile compounds was established to analyze wine, beer, and *soju* samples. The LOD and LOQ were calculated to be 1.00 and 3.04 ug/mL, respectively, which shows that the method has sufficient sensitivity. The methanol content was higher in wine than in beer and *soju*, with concentrations ranging from 43.6 to 396 mg/L in wine, 0 to 13.6 mg/L in beer, and 1.45 to 25.6 mg/L in *soju*.

To compare volatile components of the three types of alcoholic beverages, a multivariate statistical approach, such as principal component analysis (PCA), was applied to distinguish the different sample groups, depending on volatile components. This study found some volatile compounds that contribute to the discrimination of three types of alcoholic beverages. We also provided volatiles profiles for three types of alcoholic beverages, possibly suggesting quality indicators for alcoholic beverages.

Abbreviations

GC–MS	Gas chromatography-mass spectrometry
SPME	Solid phase micro-extraction
MFDS	Korean ministry of food and drug safety
Carboxen-PDMS	Carboxen/polydimethylsiloxane fiber
SIM	Selected ion monitoring
EI	Electron impact
RT	Retention times
LOD	Limit of detection
LOQ	Limit of quantification
GPP	Geranyl pyrophosphate
FPP	Farnesyl diphosphate
GGPP	Geranylgeranyl diphosphate
MVA	Mevalonate
MEP	2-C-methyl-D-erythritol 4-phosphate
PCA	Principal component analysis
PLS-DA	Partial least square-discrimination analysis

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Author contributions
Dayoung Jeong: Methodology and formal analysis, Data curation, Writing-original Draft, Visualization. Young-Suk Kim: Conceptualization, Data curation, Writing-review & editing, Supervision. All authors have read and agreed to the published version of the manuscript.

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Availability of data and material
All data generated and analyzed for this study are included in this published.

Declarations

Competing interests
The researcher claims no conflicts of interest.

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