## AGRICULTURAL AND FOOD CHEMISTRY

# Generation of Volatile Compounds in Litchi Wine during Winemaking and Short-Term Bottle Storage

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**ABSTRACT:** Evolution of volatile components during litchi (*Litchi chinensis* Sonn.) winemaking was monitored, and aroma profiles of litchi wines bottle aged for 5 months at ambient temperature  $(25-28 \, ^{\circ}\text{C})$  and low temperature  $(8-10 \, ^{\circ}\text{C})$  were compared via headspace solid phase microextraction (SPME) coupled with gas chromatography—mass spectrometry (GC-MS). The majority of terpenoids deriving from litchi juice decreased, even disappeared along with alcoholic fermentation, while terpenol oxides, ethers, and acetates came into being and increased. Ethyl octanote, isoamyl acetate, ethyl hexanoate, ethyl butanoate, *cis*-rose oxide, and *trans*-rose oxide had the highest odor activity values (OAVs) in young litchi wines. Six aromatic series were obtained by grouping OAVs of odor-active compounds with similar odor descriptions to establish the aroma profile for young litchi wines, and floral and fruity attributes were two major aroma series. Compared to ambient temperature when bottle aging, lower temperature benefited key aroma retention and expectantly extended the shelf life of young litchi wines.

KEYWORDS: Litchi wine, volatile compounds, alcoholic fermentation, aroma profile, bottle storage

#### ■ INTRODUCTION

Litchi (*Litchi chinensis* Sonn.) is a tropical and subtropical fruit of high commercial value belonging to the *Sapindaceae* family and has earned its popularity worldwide due mainly to its charming aroma and attractive red appearance. However, the distinctive flavor of this fruit, usually described as honey, rose-floral, and citrus-fruity,<sup>1–3</sup> is liable to lose its attractive feature and rapidly goes unpleasant once harvested from trees.<sup>4</sup> Therefore, much attention has been paid to postharvest quality preservation via various approaches<sup>5,6</sup> and further processing of this fruit, which includes the production of litchi wine, litchi juice, dried litchi, and canned litchi,<sup>7</sup> aiming to keep its excellent characteristics for a longer period.

Winemaking is a promising way of litchi deep processing. To produce litchi wine of high quality, many researchers are concerned about appropriate parameters of fermentation, such as temperature,<sup>8,9</sup> amino acids as additive,<sup>10,11</sup> yeast, and SO<sub>2</sub>.<sup>12</sup> As a crucial feature of a high-quality litchi wine, charming flavor has also been investigated, and the results showed that esters, alcohols, and terpenoids were the most abundant volatile compounds of litchi wine.<sup>13–16</sup> However, it remains unclear as to how these volatiles, especially terpenoids derived from litchi fruit, develop during litchi winemaking and bottle-aging processes.

Brewing adaptability of different litchi cultivars was preliminarily studied, and cv. Huaizhi was proved to be one of the most appropriate cultivars for winemaking.<sup>17,18</sup> In addition, our prior study<sup>19</sup> indicated that Huaizhi fruit had a characteristic volatile profile containing more terpenoids than other varieties. Therefore, it is expected that more terpenoids in this cultivar would be retained through the winemaking process and contribute to the final litchi wine flavor. In this study, the changes of volatile components during litchi winemaking from the Huaizhi cultivar and the odor profiles of litchi wines bottle-stored under different conditions for a short-term were investigated via gas chromatography—mass spectrometry (GC-MS) combined with headspace solid phase microextraction (SPME), aiming to know about the fate of volatiles in litchi wine and to provide some valuable information for inhibiting flavor deterioration in litchi wine processing.

#### MATERIALS AND METHODS

Litchi Wine. Litchi wine was made from Huaizhi cultivar (Litchi chinensis Sonn.) harvested in 2006 from Guangdong Province, China. Fresh fruits were peeled, deseeded, and crushed, then total sugar (expressed as glucose) of the litchi juice was adjusted to 190 g/L with sucrose and pH to 3.6 with tartaric acid to avoid the growth of microorganisms, and 100 mg/L SO2 was added for preservation. Physicochemical characteristics of initial juice were as follows: total sugar, 162 g/L; titratable acidity (expressed as tartaric acid), 3.1 g/L; and pH 4.2. Afterward, the modified litchi juice was transported into a 25 m<sup>3</sup> stainless steel tank and kept at 5 °C for 48 h for clarification. Clear litchi juice was separated, and 200 mg/L in advance activated industrial yeast (DV10, Lallemand Co., France) was added into the clear juice to start fermentation. The alcoholic fermentation temperature was controlled between 12 to 14 °C until the total sugar was reduced below 3 g/L. The samples were collected at four periods: litchi juice before modification (F-1), early stage (10 days after yeast addition, F-2), later stage (20 days after yeast addition, F-3), and the end of alcoholic fermentation (reducing sugar was 1.8 g/L, F-4). Physicochemical characteristics of the litchi wine were as follows: ethanol, 10.9% (v/v); titratable acidity, 6.0 g/L; volatile acidity, 0.4 g/L (expressed as acetic acid); reducing sugar, 2.1 g/L; and free SO<sub>2</sub>, 28.5 mg/L. Diatomaceous filtration was conducted after alcoholic fermentation, and subsequently, the finished litchi wine was stored in green bottles under two different temperature conditions (ambient temperature, 25-28 °C, A-5 and lower temperature, 8–10  $^{\circ}\text{C},$  L-6) for about 5 months before analysis.

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## Table 1. Purities, Manufacturers, Quantitative Ions, and Emendation Factors to 4-Methyl-2-pentanol of the Volatile Standards Used for the Characterization of Litchi Wines

				emendat	ion factor <sup>b</sup>
standard	purity	manufacturer <sup>a</sup>	quantitative ion	1%	11%
ethyl acetate	99.9%	Supelco	43	10.359	15.352
ethanol	99.9%	Sigma-Aldrich	45	11995.4	9520.96
isobutyl acetate	99.5%	Sigma-Aldrich	43	_ <sup>c</sup>	1.278
1-propanol	99.8%	Sigma-Aldrich	31	-	220.67
isobutanol	99.9%	Supelco	43	23.637	19.460
isoamyl acetate	99.5%	Sigma-Aldrich	70	1.419	3.355
1-butanol	99.1%	Supelco	56	-	21.514
$\beta$ -myrcene	95.0%	Fluka	93	4.657	10.449
limonene	99.5%	Dr. Ehrenstorfer GmbH	68	4.130	2.083
isopentanol	99.0%	Sigma-Aldrich	70	27.685	14.213
ethyl hexanoate	99.5%	Sigma-Aldrich	88	1.503	1.547
1-pentanol	99.5%	Dr. Ehrenstorfer GmbH	70	-	17.462
hexyl acetate	99.7%	Fluka	43	-	0.885
<i>p</i> -cymene	99.5%	Supelco	119	10.227	-
terpinolene	97.0%	Fluka	136	10.429	14.405
acetoin	96.0%	Aldrich	45	534.968	-
4-methyl-1-pentanol	97.0%	Sigma-Aldrich	56	-	2.955
2-heptanol	99.0%	Dr. Ehrenstorfer GmbH	45	0.591	-
ethyl lactate	98.0%	Aldrich	45	93.248	66.971
1-hexanol	99.0%	Dr. Ehrenstorfer GmbH	56	1.092	3.079
rose oxide <sup>d</sup>	99.0%	Fluka	139	0.164	0.313
heptyl acetate	98.0%	Aldrich	45	-	0.741
methyl octanoate	99.5%	Dr. Ehrenstorfer GmbH	74	-	0.320
nonanal	98.0%	Supelco	57	1.002	-
(E)-2-hexen-1-ol	96.0%	Sigma-Aldrich	57	2.854	-
2-octanol	99.0%	Dr. Ehrenstorfer GmbH	45	0.535	-
ethyl octanoate	99.5%	Dr. Ehrenstorfer GmbH	88	7.107	0.865
linalool oxide <sup>e</sup>	97.0%	Fluka	59	6.741	-
1-octen-3-ol	98.0%	Fluka	57	0.380	0.552
1-heptanol	99.5%	Dr. Ehrenstorfer GmbH	70	1.062	1.161
isoamyl hexanoate	98.0%	Aldrich	70	-	0.199
acetic acid	99.0%	Supelco	60	2217.59	2638.31
furfural	99.5%	Fluka	96	-	2.432
2-ethyl-1-hexanol	99.5%	Dr. Ehrenstorfer GmbH	57	0.241	0.327
benzaldehyde	99.0%	Dr. Ehrenstorfer GmbH	106	0.928	0.514
meso-2,3-butanediol	99.0%	Sigma-Aldrich	45	1139.81	3249.70
linalool	96.5%	Aldrich	93	0.900	0.798
1-octanol	99.5%	Fluka	84	1.701	0.916
allyl methyl sulfide	98.0%	Sigma-Aldrich	88	-	191.02
ethyl decanoate	99.5%	Dr. Ehrenstorfer GmbH	88	1.883	1.956
diethyl succinate	99.5%	Dr. Ehrenstorfer GmbH	129	-	6.046
α-terpineol	90.0%	Sigma-Aldrich	136	1.608	0.835
methionol	98.0%	Sigma-Aldrich	106	-	269.869
citral <sup>f</sup>	95.0%	Sigma-Aldrich	69	1.661	-
geranyl acetate	99.0%	Fluka	69	-	0.779
$\beta$ -citronellol	95.0%	Sigma-Aldrich	69	0.952	2.947
nerol	97.0%	Sigma-Aldrich	69	0.860	2.237
2-phenethyl acetate	99.0%	Fluka	104	0.536	0.653
ethyl dodecanoate	99.5%	Dr. Ehrenstorfer GmbH	88	12.369	11.344
geraniol	99.5%	Fluka	69	2.224	7.988
hexanoic acid	99.0%	Dr. Ehrenstorfer GmbH	60	20.543	15.194

#### Table 1. Continued

				emendation factor $^{b}$	
standard	purity	manufacturer <sup>a</sup>	quantitative ion	1%	11%
geranylacetone <sup>g</sup>	65.0%	Sigma-Aldrich	69	1.502	3.451
benzyl alcohol	99.8%	Supelco	108	35.872	-
2-phenylethanol	99.0%	Fluka	91	15.084	14.430
butylated hydroxytoluene	99.5%	Sigma-Aldrich	205	0.325	1.227
1-dodecanol	98.0%	Dr. Ehrenstorfer GmbH	70	7.558	19.172
octanoic acid	99.0%	Dr. Ehrenstorfer GmbH	60	14.947	14.315
decanoic acid	99.0%	Dr. Ehrenstorfer GmbH	60	33.222	86.275

<sup>*a*</sup> Manufacturers: Supelco, Bellefonte, PA; Sigma-Aldrich, St. Louis, MO; Fluka, Buchs, Switzerland; Dr. Ehrenstorfer GmbH, Augsburg, Germany; Aldrich, Milwaukee, WI. <sup>*b*</sup> Emendation factors to 4-methyl-2-pentanol (internal standard) of each volatile standards were measured in synthetic wines containing 1% and 11% (v/v) ethanol separately. <sup>*c*</sup> Not employed in this study. <sup>*d*</sup> Mixtures of *cis*- and *trans*-rose oxide. <sup>*e*</sup> Mixtures of *cis*- and *trans*-furan linalool oxide. <sup>*f*</sup> Mixtures of geranial and neral. <sup>*g*</sup> Mixtures containing 35% nerylacetone.

**Chemicals.** DL-Malic acid ( $\geq$ 98%, capillary GC) was purchased from Sigma-Aldrich (China sector), and NaOH (analytical grade) was purchased from Beijing Chemical Works. All volatile standards used for identification and quantification in this study were of HPLC quality or GC grade, and their purities and commercial sources are listed in Table 1.

**Extraction of Volatile Compounds.** The SPME manual device equipped with a 50/30- $\mu$ m DVB/CAR/PDMS fiber (Supelco, Bellefonte, PA., USA) was employed for the extraction of volatile compounds in litchi wine. Aliquots (5 mL) of litchi wine and 10  $\mu$ L of 4-methyl-2-pentanol (1.039 mg/mL water, internal standard) were blended in a 15-mL airtight vial containing a magnetic stirrer. After being equilibrated at 40 °C for 30 min, the sample was extracted with DVB/CAR/PDMS fiber (conditioned at 270 °C for 1 h prior to use) for 30 min with continued heating and agitation.<sup>19</sup> Afterward, the fiber was inserted into the GC injector for 25 min to desorb analytes. Each litchi wine sample was extracted in triplicate.

**GC-MS Analysis.** An Agilent 6890 GC coupled with an Agilent 5975 MS and equipped with a 60 m  $\times$  0.25 mm id HP-INNOWAX capillary column with 0.25  $\mu$ m film thickness (J&W Scientific, Folsom, Calif., USA) was applied to separate and identify the volatile compounds. The conditions of GC-MS in this study were previously reported.<sup>19</sup> Retention indices were calculated using the C7–C24 *n*-alkane series (Supelco, Bellefonte, PA) under the same chromatographic conditions. Identification was based on retention indices of reference standards and mass spectra matching in the standard NIST 05 library. When reference standards were not available, tentative identification was made according to mass spectra matching in the standard NIST 08 library and a comparison of retention indices sourced in NIST Standard Reference Database.<sup>20</sup>

**Quantification.** Quantification procedure was carried out using the internal standard quantification method, and 4-methyl-2-pentanol was employed as the internal standard (10  $\mu$ L of a 1.039 mg/mL solution in water of this internal standard was added to each sample and standard solution). Considering that high ethanol content greatly affected the absorption ability of SPME fiber for other volatiles,<sup>21</sup> two synthetic matrixes with 1% and 11% (v/v) ethanol were prepared in distilled water, each contained 3.0 g/L malic acid, and the pH was adjusted to 3.6 with NaOH. Volatile standards dissolved in 1% and 11% synthetic matrixes at concentrations typically found in litchi juice<sup>19</sup> and grape wine,<sup>22,23</sup> respectively. Volatile standards were then extracted and analyzed under the same conditions as litchi wine samples to obtain their emendation factors to the internal standard, and they are listed in Table 1. Quantitative data of the identified volatile compounds were calculated through the following formula:

analyte's concentration = (analyte's area/4-methyl-2-pentanol's area) × emendation factor to 4-methyl-2-pentanol × 4-methyl-2-pentanol's concentration

When a volatile standard was a mixture of two isomers (indicated in Table 1), the sum of areas of these two isomers was employed to

calculate the emendation factor to the internal standard, and this emendation factor was then used to measure the amounts of each isomer in samples. The emendation factors obtained in the 1% standard solution were used for the quantification of volatiles in litchi juice (F-1), and those obtained in the 11% standard solution were used for volatiles in litchi wine samples (F-2, F-3, F-4, A-5, and L-6). The concentration of volatile compounds for which there was no pure reference available was estimated by using the same emendation factor (obtained in the same standard solution) as one of the compounds with the most similar chemical structure.<sup>22</sup>

**Statistical Analysis.** All statistical procedures were performed through SPSS, version 16.0, statistical package for Windows (SPSS Inc., USA). A one-way analysis of variance (ANOVA) was used to analyze the OAV result in finished and bottle-aged litchi wines (F-4, A-5, and L-6) employing Duncan's multiple range tests at a level of p < 0.05. The volatile amounts and OAVs were presented as the mean  $\pm$  SD of triplicate measurements.

#### RESULTS AND DISCUSSION

Generation of Litchi Wine Volatiles during Alcoholic Fermentation. All volatile chemicals detected in litchi juice and wine samples in this study were listed in Table 2. In litchi juice (F-1), 87 volatile chemicals were observed, including 36 terpenoids, 25 alcohols, 10 esters, 4 aliphatic acids, 3 aldehydes, 2 ketones, 2 volatile phenols, 2 alkenes, 2 ethers, and 1 sulfur compound. Alcohols, esters, aliphatic acids, and terpenoids constituted major volatile components of litchi juice. Some terpenoids, to our knowledge, such as geranyl methyl ether,  $\beta$ -gurjunene,  $\alpha$ -cyclogeraniol,  $\tau$ -muurolol,  $\alpha$ -cadinol, cadalene, and geranic acid, have never been reported as litchi fruit or litchi juice constituents but are tentatively identified in this study.

Terpenoids were thought to play a key role in imparting characteristic flavor to litchi fruit.<sup>1–3,19</sup> During alcoholic fermentation of litchi wine, most terpenoids, 25 in 36, were found to have disappeared gradually, and these compounds were, namely,  $\beta$ -myrcene, limonene, (Z)- $\beta$ -ocimene, (E)- $\beta$ -ocimene, p-cymene, terpinolene, (E,Z)-alloocimene, (E,E)-alloocimene, cis-furan linalool oxide, *trans*-furan linalool oxide,  $\alpha$ -copaene,  $\beta$ -gurjunene,  $\gamma$ -elemene, neral,  $\gamma$ -muurolene,  $\alpha$ -cyclogeraniol, borneol,  $\alpha$ -muurolene, geranial,  $\gamma$ -geraniol, *cis*-isogeraniol, *trans*-isogeraniol, *p*-cymen-8-ol, geranylacetone, and geranic acid. Additionally, another 6 relatively abundant terpenols, linalool, 4-terpineol,  $\alpha$ -terpineol,  $\beta$ -citronellol, nerol, and geraniol decreased rapidly throughout alcoholic fermentation but all could

volatile chemicals	RI	ID	F-1	F-2	F-3	F-4	A-5	L-6
ethyl acetate	878	А	6092.0 ± 351.6	58006.8 ± 9042.2	53360.9 ± 6141.5	42266.5 ± 2935.8	47755.4 ± 5563.5	49958.4 ± 1550.0
ethanol (g/L)	925	А	$8.7 \pm 1.1$	$84.2 \pm 15.0$	$80.6\pm9.0$	$83.7\pm0.2$	$82.6\pm7.2$	$84.1 \pm 5.2$
isobutyl acetate	1013	А	nd	$265.3\pm37.1$	$226.3\pm46.5$	$234.9\pm38.9$	$226.0\pm11.2$	$192.1 \pm 2.7$
ethyl butanoate	1047	Α	nd	$1669.8 \pm 247.1$	$1503.6 \pm 154.8$	$1381.9\pm80.5$	$1357.7 \pm 47.6$	$1453.1\pm31.7$
1-propanol	1057	Α	nd	$25013.7 \pm 396.3$	$24938.3 \pm 951.1$	$15924.6 \pm 2761.5$	$17546.9 \pm 2211.2$	$15774.5 \pm 364.9$
isobutanol	1111	А	$134.0 \pm 21.7$	$14283.6 \pm 549.0$	14451.7 ± 1350.0	$7599.9 \pm 1389.0$	9359.4 ± 1259.3	$10778.7 \pm 284.7$
isoamyl acetate	1122	Α	$7.1\pm0.6$	$9430.6 \pm 1701.9$	$8390.7 \pm 1029.3$	$6812.7\pm237.9$	$7337.8 \pm 157.3$	$8291.6\pm91.5$
1-butanol	1158	Α	nd	$511.8\pm5.0$	$524.2\pm3.2$	$357.4\pm9.3$	$432.8\pm78.3$	$390.3\pm5.1$
$\beta$ -myrcene	1164	А	$441.3\pm27.9$	$109.4\pm68.6$	nd	nd	nd	nd
limonene	1193	Α	$76.7\pm12.6$	$7.3 \pm 2.7$	$4.0 \pm 1.2$	nd	nd	nd
4-pentenyl acetate	1199	В	nd	$9.0 \pm 3.0$	$7.0 \pm 1.1$	nd	nd	nd
isopentanol	1220	А	$604.5\pm40.0$	$26114.2\pm14.1$	$26639.6 \pm 401.3$	$19830.0 \pm 177.6$	$22672.2 \pm 1182.4$	$23128.2 \pm 189.0$
ethyl hexanoate	1232	Α	$3.6\pm0.3$	$1910.5\pm318.7$	$1629.1\pm185.0$	$1854.8\pm5.8$	$1396.2\pm94.1$	$1410.8\pm174.6$
(Z)- $\beta$ -ocimene	1240	В	$40.3\pm11.7$	nd	nd	nd	nd	nd
1,3,5-trimethylbenzene	1248	В	$124.4\pm4.2$	$141.3\pm46.4$	$87.6 \pm 16.2$	$117.3\pm25.5$	$101.5\pm14.2$	$114.9\pm7.2$
1-pentanol	1255	А	nd	$36.5\pm3.3$	$51.8\pm0.7$	$39.8 \pm 13.7$	$39.3 \pm 4.9$	$36.8\pm 6.5$
3-methyl-3-butenol	1255	В	$24.5\pm1.5$	$16.1\pm0.1$	$15.7\pm0.3$	$12.6\pm0.2$	$13.5\pm1.0$	$14.4\pm0.3$
3-methyl-2-butenyl acetate	1256	В	$6.4\pm0.4$	$6.0\pm0.5$	$5.6\pm0.5$	$5.8\pm0.1$	$3.9\pm0.3$	$4.4\pm0.3$
(E)- $\beta$ -ocimene	1257	В	$89.2 \pm 26.4$	nd	nd	nd	nd	nd
<i>p</i> -cymene	1271	А	$154.9 \pm 12.7$	nd	nd	nd	nd	nd
hexyl acetate	1273	А	nd	$49.4\pm8.0$	$44.1\pm5.5$	$49.4\pm0.6$	$39.8 \pm 0.7$	$43.2\pm2.0$
terpinolene	1290	А	$47.2\pm2.2$	$25.3\pm 6.3$	nd	nd	nd	nd
acetoin	1298	А	$107252\pm9998$	nd	nd	nd	nd	nd
2,4-dithiapentane	1299	В	$10.2\pm0.5$	$2.1\pm0.4$	nd	nd	nd	nd
2-heptanol	1319	Α	$17.5\pm1.1$	nd	nd	nd	nd	nd
3-methyl-2-butenol	1322	В	$147.9\pm9.2$	nd	nd	nd	nd	nd
3-methyl-1-pentanol	1328	Α	nd	$10.6\pm0.1$	$10.4\pm1.0$	$9.9\pm0.3$	$11.2\pm0.2$	$12.8\pm1.1$
6-methyl-5-hepten-2-one	1342	Α	$9.8\pm1.6$	nd	nd	nd	nd	nd
2,6-dimethyl-4-heptanol	1344	С	d	d	d	d	d	d
ethyl lactate	1350	А	$3506.3\pm214.2$	$841.8\pm46.0$	$903.3\pm49.1$	$850.6\pm188.3$	$5458.7\pm357.3$	$\textbf{3493.4} \pm \textbf{18.5}$
1-hexanol	1355	А	$51.4\pm4.1$	$15.1\pm1.7$	$13.1\pm0.01$	$12.1\pm2.5$	$14.7\pm1.2$	$13.0\pm1.5$
cis-rose oxide	1356	А	$3.2\pm1.6$	$10.3\pm0.9$	$10.2\pm0.4$	$12.5\pm0.02$	$15.5\pm0.1$	$18.9\pm0.6$
trans-rose oxide	1370	Α	$1.3\pm0.6$	$3.4\pm0.3$	$3.4\pm0.2$	$4.1\pm0.07$	$3.1\pm0.1$	$5.1\pm0.1$
heptyl acetate	1373	А	nd	$4.0\pm0.9$	$3.5\pm0.6$	$3.9\pm0.5$	nd	nd
(E,Z)-alloocimene	1378	В	$48.1 \pm 13.4$	nd	nd	nd	nd	nd
2-ethylhexyl acetate	1378	В	nd	$2.58\pm0.41$	$2.60\pm0.03$	$4.04\pm0.06$	$1.58\pm0.09$	$1.70\pm0.37$
3-ethoxy-1-propanol	1380	А	nd	$26.3\pm0.5$	$30.7\pm3.0$	nd	nd	nd
methyl octanoate	1390	А	nd	$7.0 \pm 0.8$	$6.5\pm0.2$	$8.0 \pm 0.7$	$3.7\pm0.1$	$4.5 \pm 0.2$
3-octanol	1392	В	$22.6 \pm 4.1$	nd	nd	nd	nd	nd
nonanal	1394	А	$1.5 \pm 0.6$	nd	nd	nd	nd	nd
( <i>E,E</i> )-alloocimene	1400	В	$44.0 \pm 9.3$	nd	nd	nd	nd	nd
(E)-2-hexen-1-ol	1409	А	$26.4 \pm 3.3$	nd	nd	nd	nd	nd
2-octanol	1417	Α	$5.0 \pm 1.2$	nd	nd	nd	nd	nd
4-methyl cyclohexene	1418	С	nd	d	d	d	d	d
ethyl octanoate	1437	А	$42.1 \pm 1.0$	$4370.5 \pm 677.4$	$4148.7 \pm 44.1$	$4739.3 \pm 26.0$	$2698.9 \pm 73.4$	$3054.1 \pm 55.7$
cis-furan linalool oxide	1445	А	$9.3 \pm 1.3$	nd	nd	nd	nd	nd
$p,\alpha$ -dimethyl styrene	1445	В	$44.8 \pm 4.0$	nd	nd	nd	nd	nd
1-octen-3-ol	1451	А	$74.2 \pm 12.3$	$9.2 \pm 1.9$	$7.8 \pm 0.5$	$8.6 \pm 2.1$	$5.87 \pm 0.01$	$5.6 \pm 0.5$
1-heptanol	1455	Α	$3.8 \pm 0.3$	$7.3 \pm 3.2$	$7.2 \pm 0.6$	$8.6 \pm 1.7$	$7.5 \pm 0.4$	$8.4 \pm 0.5$
isoamyl hexanoate	1458	Α	nd	$2.6 \pm 0.3$	$2.26 \pm 0.03$	$2.4 \pm 0.1$	$1.2 \pm 0.2$	$0.82 \pm 0.05$
acetic acid	1463	А	9666 ± 394	$192136 \pm 32560$	$201886 \pm 21488$	$168381 \pm 30258$	$151482 \pm 4503$	$128804 \pm 24987$
6-methyl-5-hepten-2-ol	1464	Α	$12.5 \pm 1.9$	nd	nd	nd	nd	nd

## Table 2. Concentrations of Volatile Components in Litchi Wine Samples from Winemaking and Short-Term Bottle Storage Processes $(\mu g/L)^a$

#### Table 2. Continued

volatile chemicals	RI	ID	F-1	F-2	F-3	F-4	A-5	L-6
menthone	1467	В	nd	nd	nd	nd	nd	$15.0\pm0.2$
geranyl methyl ether	1468	С	d	d	d	d	d	d
trans-furan linalool oxide	1474	А	$29.5\pm4.9$	nd	nd	nd	nd	nd
furfural	1474	А	nd	nd	nd	nd	$31.8\pm1.9$	$11.3\pm1.0$
nerol oxide	1474	В	$1.5\pm0.7$	$8.3\pm1.6$	$7.3\pm0.7$	$10.3\pm0.4$	$11.6\pm0.5$	$14.3\pm0.7$
octyl acetate	1475	В	nd	$13.2\pm1.1$	$12.6\pm1.2$	$19.2\pm2.5$	$4.6\pm0.4$	$8.5\pm0.8$
2-ethyl-1-hexanol	1490	А	$8.9\pm1.5$	nd	nd	nd	$0.806\pm0.003$	$0.9\pm0.1$
methyl phenethyl ether	1491	С	d	d	d	d	d	d
α-copaene	1494	В	$1.8\pm1.0$	nd	nd	nd	nd	nd
(E)-2-hepten-1-ol	1511	В	$5.5\pm1.4$	nd	nd	nd	nd	nd
geranyl ethyl ether	1512	В	nd	$2.7\pm0.5$	$3.6\pm1.3$	$5.6\pm1.4$	$19.6\pm1.6$	$20.4\pm0.2$
propyl octanoate	1513	В	nd	$0.32\pm0.06$	$0.35\pm0.06$	nd	nd	nd
benzaldehyde	1534	А	$2.1\pm0.6$	nd	$0.75\pm0.04$	nd	nd	$0.97\pm0.05$
levo-2,3-butanediol	1542	В	$16405\pm2039$	$483685 \pm 33469$	$572558 \pm 17762$	$447893 \pm 63842$	$419286 \pm 62169$	$468136 \pm 83232$
linalool	1547	А	$32.9\pm8.6$	$8.6\pm1.1$	$7.4\pm0.8$	$13.4\pm1.2$	$25.2\pm2.2$	$23.7\pm1.3$
isobutyl octanoate	1551	В	nd	$0.35\pm0.02$	$0.4\pm0.1$	$0.5\pm0.2$	$0.25\pm0.04$	nd
1-octanol	1557	А	$2.9\pm1.2$	$1.4\pm0.3$	$1.2\pm0.1$	$2.2\pm0.2$	$1.8\pm0.1$	$2.3\pm0.1$
meso-2,3-butanediol	1579	А	$14172\pm1701$	$130859\pm8495$	$160857\pm3183$	$125566 \pm 11133$	$114413\pm 6650$	$137574 \pm 27300$
methyl decanoate	1591	А	nd	$1.2\pm0.3$	$1.3\pm0.5$	$1.5\pm0.4$	$0.37\pm0.03$	$0.93\pm0.04$
eta-gurjunene	1598	В	$1.6 \pm 1.0$	nd	nd	nd	nd	nd
4-terpinenol	1607	В	$10.8\pm2.5$	$2.4\pm0.3$	$2.0 \pm 0.2$	$4.0 \pm 0.3$	$2.9\pm0.3$	$3.59\pm0.06$
(E)-2-octen-1-ol	1614	В	$5.4 \pm 1.4$	nd	nd	nd	nd	nd
ethyl furoate	1631	В	nd	nd	nd	nd	nd	$16.4 \pm 1.1$
allyl methyl sulfide	1632	А	nd	$2447.3 \pm 157.1$	$2077.7\pm90.3$	$3926.3\pm48.0$	$2122.8\pm195.6$	$3016.4 \pm 117.4$
ethyl decanoate	1639	А	$19.5\pm4.7$	$2232.1 \pm 169.1$	$2492.4\pm865.3$	$2638.4\pm369.5$	$886.1\pm26.7$	$1447.3\pm26.2$
γ-elemene	1645	В	$3.7 \pm 1.4$	$12.8\pm0.2$	nd	nd	nd	nd
isoamyl octanoate	1659	В	nd	$1.6\pm0.1$	$1.9\pm0.7$	$1.7 \pm 0.3$	$0.6\pm0.1$	$1.2\pm0.1$
1-nonanol	1661	В	$3.2 \pm 1.8$	nd	nd	nd	nd	nd
citronellyl acetate	1662	В	nd	$22.3\pm2.3$	$21.3\pm8.2$	$42.6\pm7.5$	$6.2 \pm 0.7$	$20.9\pm0.2$
diethyl succinate	1682	А	nd	nd	nd	$11.3 \pm 0.2$	$54.5 \pm 4.6$	$53.3 \pm 4.9$
neral	1689	А	$3.8 \pm 1.0$	nd	nd	nd	nd	nd
ethyl 9-decenoate	1693	В	nd	$101.2 \pm 6.3$	$121.8\pm48.5$	$167.0 \pm 39.8$	$58.8\pm3.1$	$95.8 \pm 1.7$
$\gamma$ -muurolene	1697	В	$1.9 \pm 0.9$	nd	nd	nd	nd	nd
α-cyclogeraniol	1702	С	d	nd	nd	nd	nd	nd
α-terpineol	1703	А	$13.6 \pm 3.1$	$1.9 \pm 0.2$	$1.7 \pm 0.1$	$4.3 \pm 0.1$	$3.1 \pm 0.4$	$3.5 \pm 0.2$
borneol	1709	А	$2.7 \pm 0.3$	nd	nd	nd	nd	nd
(Z)-6-nonen-1-ol	1720	В	$4.1 \pm 0.6$	nd	nd	nd	nd	nd
3-methylthiopropanol	1726	Α	nd	$3068.7 \pm 270.0$	$3167.9 \pm 21.1$	$3507.4 \pm 426.1$	$2796.0 \pm 280.9$	3494.7 ± 582.7
neryl acetate	1729	A	nd	$6.0 \pm 0.9$	$5.5 \pm 1.9$	$13.6 \pm 3.2$	$2.2 \pm 0.5$	$5.6 \pm 0.4$
α-muurolene	1734	В	$7.0 \pm 2.2$	nd	nd	nd	nd	nd
2-acetylphenol	1734	В	nd	$61.1 \pm 6.2$	$54.5 \pm 21.0$	$136.1 \pm 31.1$	$26.6 \pm 4.5$	$55.2 \pm 4.2$
benzyl acetate	1735	В	nd	nd	nd	nd	nd	$5.0 \pm 0.3$
geranial	1741	A	$43.6 \pm 8.8$	nd	nd	nd	nd	nd
geranyl acetate	1760	A	$2.1 \pm 1.1$	$26.8 \pm 4.0$	$23.2 \pm 8.4$	$62.7 \pm 12.7$	$8.7 \pm 2.2$	$18.8 \pm 1.2$
p-citronellol	17/0	A	$68.5 \pm 23.9$	$24.3 \pm 9.6$	$19.0 \pm 1.7$	$56.1 \pm 3.6$	$25.8 \pm 6.0$	$52.1 \pm 0.8$
γ-geranioi	1793	D	$2/.8 \pm 8.7$	nd				
etnyl pnenylacetate	1/98	D	$0.13 \pm 0.05$		$0.331 \pm 0.000$	$0.8 \pm 0.2$	$2.1 \pm 0.2$	$1.964 \pm 0.002$
neroi	1808	A D	$29.3 \pm 8.3$	$2.1 \pm 0.8$	$1.0 \pm 0.2$	$4.3/\pm 0.04$	$2.2 \pm 0.2$	$2.87 \pm 0.06$
trans isogerania <sup>1</sup>	1010	D	$2.3 \pm 0.7$	nd	nd	nd	nd	nd
2 phonothyl acetate	1019	D	$10.0 \pm 3.7$	$\frac{110}{974 \pm 64}$	11u 70 5 ± 5 0	10	110	10
2-prienemyr acetate	1030	л ^	$1.0 \pm 0.2$	$07.4 \pm 0.4$	$70.3 \pm 3.8$	$2+3.4 \pm 32.2$	$72.1 \pm 20.3$	$133.3 \pm 3.9$
geraniol	1048	л л	$11.10 \pm 0.00$ $1053.2 \pm 249.4$	$201.0 \pm 9.4$ 121.8 $\pm$ 62.2	$\pm \pm 10.7 \pm 218.3$	$513.7 \pm 220.8$ 105.1 $\pm$ 15.2	$137.1 \pm 01.3$ $13.8 \pm 2.2$	$52.7 \pm 2.1$
bovanoic acid	1000	л л	$1033.2 \pm 248.4$	$131.0 \pm 03.3$	$\pm 2.0 \pm 23.4$	$173.1 \pm 13.3$ $2262.6 \pm 150.5^{-1}$	$\pm 2.0 \pm 2.3$	$77.2 \pm 0.7$
inchantoic actu	1000	л	JT.U 4.1	1070.7 ± 04.1	10/T.T ± 0./	2000.0 ± 130.30	12/0.1 ± 100.9	1720.7 1 247.0

#### Table 2. Continued

volatile chemicals	RI	ID	F-1	F-2	F-3	F-4	A-5	L-6
p-cymen-8-ol	1862	В	$2.9\pm0.5$	nd	nd	nd	nd	nd
geranylacetone	1864	А	$0.8\pm0.1$	nd	nd	nd	$3.5\pm0.1$	$11.9\pm4.4$
benzyl alcohol	1892	А	$214.6\pm26.3$	nd	nd	nd	nd	nd
2-phenylethanol	1928	А	$429.9\pm54.8$	$1649.3\pm159.9$	$1699.4\pm59.7$	$3291.4\pm92.8$	$1858.0\pm83.9$	$2561.9\pm381.2$
butylated hydroxytoluene	1925	А	$1.2\pm0.8$	$11.8\pm2.6$	$15.2\pm9.5$	$35.9\pm5.1$	$22.8\pm0.6$	$31.6\pm2.0$
1-dodecanol	1974	А	$2.7\pm1.0$	$22.2\pm3.3$	$22.9\pm5.1$	$80.5\pm27.1$	$30.4\pm7.2$	$46.3\pm3.1$
eugenyl methyl ether	2027	В	$0.12\pm0.01$	nd	nd	nd	nd	nd
octanoic acid	2075	А	$39.8 \pm 11.3$	$1845.9\pm67.0$	$3754.3 \pm 2360.9$	$8339.0\pm794.2$	$3157.8\pm549.9$	$4820.4\pm509.4$
3-tert-butyl-4-hydroxyanisole	2111	С	d	d	d	d	d	d
au-muurolol	2185	В	nd	$4.5\pm1.3$	$4.4\pm0.9$	$28.9\pm3.4$	nd	nd
α-cadinol	2210	В	nd	nd	nd	$5.0 \pm 0.3$	nd	nd
cadalene	2242	В	nd	$12.1\pm2.0$	$14.6\pm5.3$	$27.2\pm10.8$	nd	nd
decanoic acid	2292	А	$23.2\pm7.1$	$1420.5\pm208.7$	$2002.9\pm169.6$	$10711.9 \pm 3499.5$	$3993.7 \pm 1061.7$	$3582.0\pm249.4$
geranic acid	2350	В	$3.3\pm2.6$	$375.0\pm320.5$	nd	nd	nd	nd
dibutylhydroxybenzaldehyde	>2400	С	d	d	d	nd	nd	nd

<sup>*a*</sup> Concentration values (mean  $\pm$  SD, *n* = 3) of the same compounds followed by different letters are significantly different (*P* < 0.05); RI, retention indices on HP-innowax column; ID, reliability of the identification proposal: A, identified; mass spectrum and RI agree with standards; B, tentatively identified; mass spectrum agrees with the mass spectral database, and RI agrees with literature data (ref 20); C, tentatively identified; mass spectrum agrees with the mass spectral database; F-1, F-2, F-3, F-4, A-5, and L-6 represent litchi juice before modification, early stage, later stage, and the end of alcohol fermentation of litchi wine, litchi wines bottle-stored at ambient temperature, and at lower temperature for 5 months, respectively. nd, not detected; d, detected.

### Table 3. Odor Thresholds, Odor Activity Values, Odor Description, and Assignment to Aromatic Series of Potent Odorants in Young Litchi Wines

				odor activity value <sup>b</sup>		
potent odorant	odor threshold <sup><i>a</i></sup> ( $\mu$ g/L)	odor description	aromatic series	F-4	A-5	L-6
ethyl acetate	7500 (31)	pineapple, fruity, varnish, balsamic	chemical, fruity	$5.6\pm0.4$ a	$6.4\pm0.7~a$	$6.7\pm0.2$ a
ethyl butanoate	20 (32)	strawberry, apple, banana	fruity	$69.1\pm4.0$ a	$67.9\pm2.4~\mathrm{a}$	$72.7\pm1.6$ a
isoamyl acetate	30 (32)	banana, fruity, sweet	sweet, fruity	$227.1\pm7.9~\mathrm{a}$	$244.6\pm5.2\;a$	$276.4\pm3.1~\text{b}$
isopentanol	30000 (32)	alcohol, nail polish	chemical	$0.7\pm0.0~a$	$0.8\pm0.0\ b$	$0.8\pm0.0\ b$
ethyl hexanoate	14 (32)	fruity, green apple, banana, brandy	fruity	$132.5\pm0.4~\text{b}$	$99.7\pm6.7~a$	$100.8\pm12.5~a$
cis-rose oxide	0.2 (31)	rose	floral	$62.7\pm0.1~a$	$77.7\pm0.7~b$	$94.7\pm2.8~c$
trans-rose oxide	0.2 (31)	rose	floral	$20.6\pm0.4~b$	$15.6\pm0.6~\mathrm{a}$	$25.4\pm0.5\ c$
ethyl octanoate	5 (32)	floral, fruity, banana, pear, brandy	floral, fruity	$947.9\pm5.2~c$	$539.8\pm14.7$ a	$610.8\pm11.2~\text{b}$
acetic acid	200000 (32)	vinegar, pungent	fatty	$0.8\pm0.2\;a$	$0.8\pm0.0\;a$	$0.6\pm0.1~a$
levo-2,3-butanediol	150000 (33)	fruity	fruity	$3.7\pm0.5~a$	$3.5\pm0.5\;a$	$3.9\pm0.7~\text{a}$
linalool	25 (32)	citrus, floral, sweet, grape-like	floral, sweet, fruity	$0.5\pm0.1\;a$	$1.0\pm0.1~b$	$0.9\pm0.1~b$
meso-2,3-butanediol	150000 (33)	Fruity	fruity	$1.1\pm0.1~\mathrm{a}$	$1.0\pm0.1~\text{a}$	$1.2\pm0.2~\text{a}$
ethyl decanoate	200 (32)	brandy, fruity, grape	fruity	$13.2\pm1.8~\mathrm{b}$	$4.4\pm0.1~\text{a}$	$7.2\pm0.1$ a
methionol	1000 (32)	cooked potato, garlic	herbaceous	$3.5\pm0.4~a$	$2.8\pm0.3\;a$	$3.5\pm0.6\;a$
$\beta$ -citronellol	100 (31)	rose	floral	$0.6\pm0.0~b$	$0.3\pm0.1\;a$	$0.5\pm0.0\ b$
2-phenethyl acetate	250 (32)	flowery, fruity	floral, fruity	$1.0\pm0.1~b$	$0.4\pm0.1~\text{a}$	$0.5\pm0.0\;a$
geraniol	30 (32)	rose, floral	floral	$6.5\pm0.5~b$	$1.5\pm0.1~\text{a}$	$2.5\pm0.2\;a$
hexanoic acid	420 (32)	fatty, cheese, rancid	fatty	$5.6\pm0.4b$	$3.0\pm0.4~\mathrm{a}$	$4.6\pm0.6~b$
octanoic acid	500 (32)	fatty, cheese, rancid	fatty	$16.7\pm1.6~\mathrm{b}$	$6.3\pm1.1$ a	$9.6\pm1.0$ a
decanoic acid	1000 (32)	fatty, rancid	fatty	$10.7\pm3.5~b$	$4.0\pm1.1$ a	$3.6\pm0.2~\text{a}$

<sup>*a*</sup> The reference from which the value has been taken is given in parentheses. In ref 31, the matrix was a 10% (w/w) water/ethanol solution; in ref 32, the matrix was a 11% water/ethanol solution containing 7 g/L glycerol and 5 g/L tartaric acid, with the pH adjusted to 3.4 with 1 M NaOH; in ref 33, the matrix was a 10% (v/v) water/ethanol solution with the pH adjusted to 3.5 with tartaric acid. <sup>*b*</sup> OAVs of the same compounds followed by different letters are significantly different (P < 0.05); F-4, A-5, and L-6 represent the litchi wine samples at the end of alcohol fermentation, bottle-stored at ambient temperature, and at lower temperature for 5 months, respectively.

be detected in the final litchi wine (F-4). Geraniol was reported to be present as glycosides in litchi pulp,<sup>24</sup> and this

glycoconjugate precursor was likely to be released by endogenous or/and exogenous glycosidase enzymes or chemical acid



**Figure 1.** Aromatic series in the young litchi wines stored under different conditions. F-4, A-5, and L-6 represent the litchi wine samples at the end of alcohol fermentation, bottle-stored at ambient temperature, and at lower temperature for 5 months, respectively. Asterisks show that at least two samples showed significant differences in the aromatic series.

during the winemaking procedure.<sup>23,25</sup> However, in the present study, a sharp drop in the amount of this important flavorcontributor was observed from 1053.2  $\mu$ g/L in litchi juice (F-1) to 195.1  $\mu$ g/L at the end of fermentation (F-4). Probably, this reduction is attributed to yeast metabolism through which geraniol, to some extent, might be converted into other terpenols,<sup>26</sup> and these terpenols, together with geraniol, were then partially esterified with acetyl CoA,23 or etherified with ethanol. Actually in this study, geranyl ethyl ether, nerol oxide, citronellyl acetate, neryl acetate, and geranyl acetate did increase gradually with the process of fermentation, and geranyl ethyl ether, citronellyl acetate, and neryl acetate were not detected in litchi juice. In addition, another two key odorants, cis-rose oxide and trans-rose oxide, also increased in the fermentation process, which might be because of reductive yeast metabolism and precursor hydrolysis.<sup>27</sup> Interestingly, two sesquiterpenes,  $\tau$ -muurolol and cadalene, newly generated and increased during alcoholic fermentation, while another one,  $\alpha$ -cadinol, was only observed at the end of alcoholic fermentation (F-4) (Table 2).

Twelve alcohols were just detected in litchi juice, namely, benzyl alcohol, (Z)-6-nonen-1-ol, 1-nonanol, (E)-2-octen-1-ol, (E)-2-hepten-1-ol, 2-ethyl-1-hexanol, 6-methyl-5-hepten-2-ol, 2-octanol, (E)-2-hexen-1-ol, 3-octanol, 3-methyl-2-buten-1-ol, and 2-heptanol. These alcohols faded away during alcoholic fermentation. Another 3 alcohols, 3-methyl-3-buten-1-ol, 1-hexanol, and 1-octen-3-ol, decreased but were all detected at the end of fermentation. On the contrary, some alcohols, such as 1-propanol, 3-methyl-1-pentanol, 1-pentanol, 1-butanol, isobutanol, isopentanol, 1-heptanol, levo-2,3-butanediol, meso-2,3-butanediol, 2-phenylethanol, and 1-dodecanol, increased along with the fermentation. The so-called higher alcohols, especially 1-propanol, 1-butanol, isobutanol, isopentanol, and 2-phenylethanol, were produced by yeast from branched-chain amino acids, the Ehrlich pathway, or from intermediates of sugar metabolism and could contribute desirable complexity to wine aroma at concentrations below 300 mg/L, but negative flavor at concentrations above 400 mg/L.<sup>23,25</sup> In this study, increase in 2,3-butanediol concentration may be closely related to the sharp drop of acetoin, owing to the reduction of diacetyl and acetoin through activities

of lactic acid bacteria and yeast.<sup>23</sup> Otherwise, 3-ethoxy-1-propanol was only detected in the middle of alcoholic fermentation.

Esters are one of the most important byproducts of alcoholic fermentation produced by yeast as secondary products of sugar metabolism and impart fruity flavor to alcoholic beverages.<sup>23,25</sup> Here, 14 esters were newly generated during alcoholic fermentation, including ethyl 9-decenoate, isoamyl octanoate, methyl decanoate, isobutyl octanoate, propyl octanoate, octyl acetate, isoamyl hexanoate, methyl octanoate, 2-ethylhexyl acetate, heptyl acetate, hexyl acetate, 4-penten-1-yl acetate, ethyl butanoate, and isobutyl acetate. Among these components, 4-penten-1-yl acetate and propyl octanoate rapidly disappeared at the end of fermentation. All esters detected in litchi juice showed increasing trends during fermentation except 3-methyl-2-buten-1-yl acetate and ethyl lactate. In the finished litchi wine, the most abundant esters (>0.5 mg/L) were ethyl acetate, isoamyl acetate, ethyl octanoate, ethyl decanoate, ethyl hexanoate, ethyl butanoate, ethyl lactate, and ethyl dodecanoate (Table 2).

Notably, 2,4-dithiapentane, the unique sulfur compound detected in litchi juice (F-1), was not observed in litchi fruits of the same cultivar (Huaizhi) in our prior study.<sup>19</sup> While 2, 4-dithiapentane diminished and disappeared rapidly, and another two sulfur compounds, allyl methyl sulfide and methionol, generated during alcoholic fermentation, of which methionol derived from methionine metabolism of microorganism elicited cauliflower and cabbage odors.<sup>23</sup>

Four volatile fatty acids, acetic acid, hexanoic acid, octanoic acid, and decanoic acid, were monitored in the present study and these acids were extremely important to wine flavor not only for their odor characters but also for their contribution to the synthesis of volatile ethyl esters.<sup>23</sup> These four volatile acids went up along with alcoholic fermentation, and this should be as a result of fatty acid metabolism of yeast and bacteria.<sup>23,25</sup> As for acetic acid, acetic and lactic acid bacteria played a key role in altering its content in wine.<sup>23</sup>

In addition, volatile compounds, such as 6-methyl-5-hepten-2-one, eugenyl methyl ether,  $p,\alpha$ -dimethyl styrene, and nonanal, gradually decreased and were not detected at the end of alcoholic fermentation. The increasing trends in the process of alcoholic fermentation were seen for butylated hydroxytoluene and 1,3, 5-trimethylbenzene (Table 2).

Evolution of Litchi Wine Volatiles during Bottle Aging. To investigate how volatile odorants develop during the first 5 months of bottle aging, we analyzed and compared volatile profiles between young litchi wine at the end of alcoholic fermentation (F-4) and those stored under ambient temperature (A-5) and lower temperature conditions (L-6). The results showed that most volatiles changed significantly along five kinds of trends. First, those volatile components decreased or disappeared in both bottle-aged litchi wines (A-5 and L-6) including geraniol, nerol, cadalene,  $\tau$ -muurolol,  $\alpha$ -cadinol, geranyl acetate, neryl acetate, citronellyl acetate, hexyl acetate, heptyl acetate, 3-methyl-2-butenyl acetate, 2-ethylhexyl acetate, octyl acetate, 2-phenethyl acetate, isoamyl hexanoate, isobutyl octanoate, ethyl hexanoate, ethyl octanoate, ethyl decanoate, octanoic acid, decanoic acid, and allyl methyl sulfide. Second, some components diminished only in the ambient-temperature-stored samples but hardly changed in the lower-temperature-stored samples, and they are  $\beta$ -citronellol,  $\alpha$ -terpineol, 4-terpinenol, hexanoic acid, methyl octanoate, isoamyl octanoate, ethyl 9-decenoate, 2-phenylethanol, and butylated hydroxytoluene. Similar results were also reported in a prior investigation of Riesling

wines<sup>28</sup> that most acetate esters decreased during short-time bottle aging, and compared with ambient temperature, lowertemperature storage could slow down the decreasing rate of some important volatile components such as isoamyl acetate. In the present investigation, isoamyl acetate together with isopentanol changed little at ambient temperature but showed an increase at lower temperature. This might be ascribed to abundant leucine in litchi juice, which persistently supplies isopentanol for esterification during bottle storage. Besides isoamyl acetate and isopentanol that evolved along the third trend, nerol oxide and 3-methyl-1-pentanol showed an increase as well, and menthone, benzaldehyde, phenylmethyl acetate, and ethyl furoate came into being only in the lower-temperature-stored litchi wines (L-6). Fourth, the components that increased during bottle aging regardless of storage temperature included cis-rose oxide, linalool, geranyl ethyl ether, geranylacetone, 1-propanol, isopentanol, 2-ethyl hexanol, furfural, diethyl succinate, ethyl lactate, and ethyl phenylacetate. Fifth, trans-rose oxide decreased in litchi wines stored at ambient temperature (A-5) but increased in lowtemperature-stored young litchi wine (L-6).

Regarding the two bottle-stored litchi wines, the concentrations of volatile odorants in the lower-temperature-stored samples (L-6) were significantly higher than, if not equal to, those in the ambient- temperature-stored ones (A-5) except for ethyl lactate and isobutyl octanoate. In detail, the lower-temperaturestored litchi wines, compared with the ambient-temperaturestored ones, possessed higher concentrations of nerol oxide, *cis*rose oxide, *trans*-rose oxide,  $\beta$ -citronellol, nerol, 4-terpineol, menthone, citronellyl actate, isoamyl acetate, ethyl octanote, phenylmethyl acetate, ethyl furoate, benzaldehyde, and allyl methyl sulfide. This finding was in good agreement with those of two prior investigations.<sup>29,30</sup> Therefore, it is reasonable to conclude that low temperature storage benefits aroma retention and hopefully extends the shelf life of young litchi wines.

Aroma Profiles of Young Litchi Wines. To better understand the aroma profiles of litchi wines, odor activity values (OAVs) of the most potent odorants, especially those with much contribution to grape wine and litchi fruit flavors in prior investigations,  $^{1-3,19,23,25}$ were calculated by dividing concentrations by their odor thresholds, and Table 3 lists the odor thresholds, odor descriptions from the literature,  $^{31-34}$  and OAVs of those aroma-active odorants. It should be noted that six odor-active volatiles with OAVs higher than 15 in young litchi wine were ethyl octanoate, isoamyl acetate, ethyl hexanoate, ethyl butanoate, cis-rose oxide, and trans-rose oxide. Of the six odor-active volatiles, cis-rose oxide and trans-rose oxide also had higher OAVs in litchi juice and imparted characteristic flavor to litchi fruit according to prior studies.<sup>2,19</sup> The following odorants with relatively high OAVs were ethyl acetate, ethyl decanoate, hexanoic acid, octanoic acid, decanoic acid, geraniol, methionol, and levo-2,3-butanediol (Table 3). Linalool, meso-2,3-butanediol, and phenethyl acetate also reached or were close to their odor thresholds, and generally all of these above-mentioned volatiles were considered to significantly contribute to litchi wine flavor.

The odor of a compound is described in terms of several descriptors agreed upon by experts, and an aromatic series could be defined as a group of volatile compounds with similar odor descriptors. By grouping in aromatic series the OAVs of the volatiles exhibiting similar odor descriptions, researchers could obtain the aroma profile of the wine studied to interpret the instrument data.<sup>33,34</sup> Accordingly, the OAVs for the compounds that showed the similar olfactory sensation based on their odor

description were grouped, and six aromatic series of odors were established (Table 3): fatty, floral, chemical, sweet, fruity, and herbaceous. Aroma profiles for litchi wines were then obtained and shown in Figure 1. As can be seen in Figure 1, floral and fruity were the most intense odor characteristics of a litchi wine followed by sweet. The floral attribute should be due chiefly to ethyl octanoate, cis-rose oxide and trans-rose oxide, and the fruity attribute is mainly owing to ethyl octanoate, isoamyl acetate, ethyl hexanoate, and ethyl butanoate, of which isoamyl acetate also was the main contributor of sweet character. Low odor intensities were observed for chemical, fatty, and herbaceous. An ANOVA was performed (p < 0.05) to differentiate these litchi wines between the aromatic series, and no significant differences were observed in the chemical and herbaceous series. The other four aromatic series, floral, fruity, sweet, and fatty, were marked with an asterisk in Figure 1, indicating that at least two litchi wines showed significant differences. Fruity and floral attributes declined along with bottle aging, and these trends were more obvious in litchi wine stored at ambient temperature than at low temperature. Sweet character was remarkably strengthened during bottle storage at low temperature, and no significant change was observed in litchi wine stored at ambient temperature. Fatty character weakened after 5 months of bottle storage, and no significant difference was observed between ambient-temperature-stored and lower-temperature-stored litchi wines. As a whole, the young litchi wine bottle aged at low temperature exhibited more fruity, floral, and sweet character.

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