

Impact of utilization of oxygen scavenger on aroma quality of Longjing tea during storage at elevated temperature

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ABSTRACT

The fresh aroma of Longjing tea is vulnerable to unfavorable storage conditions. However, limited research has addressed effective solutions apart from low-temperature storage. This study aimed to investigate the impact of oxygen scavenger on aroma quality of packaged Longjing tea samples at elevated storage temperatures. As a result, the utilization of oxygen scavenger could effectively mitigate aroma deterioration of Longjing tea caused by elevated temperature during storage, resulting in a decrease in the stale odor scores by more than 3.0. The utilization of oxygen scavenger achieved aroma-preserving effect by inhibiting the accumulation of key stale odor compounds and maintaining the levels of volatiles related to freshness aroma. The key volatile contributors to stale odor are primarily ketones and alcohols resulting from thermal carotenoids degradation/lipid oxidation, which exhibit woody or fatty odors. These findings provide essential theoretical principles for improving Longjing tea preservation technology.

1. Introduction

Longjing tea, with a history of tea culture and cultivation spanning over 1200 years, is globally recognized as one of China's prestigious green teas. In China, the total pre-sale volume of Longjing tea in JD Supermarket increased by 135 % year-on-year in 2023 according to the report of China Tea Marketing Association, implying a rapid expansion online market of Longjing tea which had occupying the offline market (Xu, 2023). Longjing tea is a geographical indication product originating from Zhejiang Province and divided into six grades according to the quality level (GB/T 18650). The distinctive sensory characteristics of Longjing tea are achieved through a sequential process consisting of spreading, pan-fixing and drying (Flaig, Qi, Wei, Yang, & Schieberle, 2020). During tea processing, the thermal treatments are employed to enhance its unparalleled quality in terms of the "emerald color, beautiful shape, fragrant aroma and mellow taste" (Tong et al., 2022).

However, the storage stability of Longjing tea is relatively low due to its susceptibility to environmental factors such as temperature, humidity and light (Wang et al., 2019). The gradual decrease in freshness flavor of Longjing tea caused by improper storage is often accompanied by gradual darkening of color and deterioration in aroma (Lee & Chambers,

2010). The undesirable off-flavor exerts a substantially negative impact on the consumption experience, nutritional value, and economic worth (Dai et al., 2019). Existing research reported the volatiles undergoing changes during storage encompass a wide range of major categories, including alcohols, esters, ketones, acids, hydrocarbons, aromatic compounds, oxygen-heterocyclic compounds and nitrogen compounds (Dai et al., 2020; Guo et al., 2020). For instance, hexanoic acid and *trans*-2-nonenal have been identified as the primary contributors to the stale odor in Longjing tea (Cao et al., 2024), while 1-penten-3-ol, 2,5-dimethylpyrazine and 1-octen-3-ol were also found to be associated with storage time (Wang, Liu, et al., 2020). To sum up, the reported variations in contributors to off-flavor during storage may differ significantly due to the distinct precursors and generation pathways induced by storage factors.

Among the environmental factors, water content and oxygen have the greatest impact on tea quality during storage (Cao et al., 2024; Xiao et al., 2022). The water content in the final tea product can be effectively reduced through the continuous optimization and improvement of tea processing techniques, as well as strict adherence to standard requirements for controlling tea moisture levels. The presence of oxygen thus emerges as the pivotal factor in triggering the deterioration of

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quality in green tea (Fan et al., 2024). Low temperature has been reported to be conducive to maintaining green tea quality during storage by inhibiting chemical reactions, with the recommended storage temperature for green tea being $-80\text{ }^{\circ}\text{C}$ or $-20\text{ }^{\circ}\text{C}$ (Dai et al., 2019). However, this storage method places significant demands on equipment performance and results in high energy consumption. The exploration of energy-efficient and streamlined storage way is therefore crucial in enhancing the sales quality of Longjing tea products. Oxygen scavenger is widely employed to prevent or decelerate oxidation in the food products by eliminating oxygen (Brody, Strpinsky, & Kline, 2001; Tewari, Jayas, Jeremiah, & Holley, 2002). The utilization of iron-based oxygen scavenger systems was predominant in commercial use (Polyakov & Miltz, 2016), with their absorption rate increasing as the temperature risen above $5\text{ }^{\circ}\text{C}$ (Charles, Sanchez, & Gontard, 2006; Galotto, Anfossi, & Guarda, 2009). The focus of this study is to investigate the utilization of oxygen scavenger in preserving aroma quality of Longjing tea during storage at elevated temperatures. The aroma quality of Longjing tea was assessed through scoring and commenting, simultaneously, their volatile profiles were quantitatively determined using a modified simultaneous distillation extraction coupled with gas chromatography–mass spectrometry (MSDE-GC–MS). We further investigated the potential key volatiles that were associated with aroma-preservation effect of oxygen scavenger on Longjing tea stored at elevated temperature, contributing to freshness-flavor and stale-flavor. The findings obtained from this study will demonstrate inhibiting effect of oxygen scavenger on aroma deterioration caused by elevated temperature while exploring key volatile contributors, which could provide an effective and energy-efficient storage way for green tea.

2. Materials and methods

2.1. Preparation of Longjing tea samples

Longjing tea samples were produced at Hangzhou Tea Factory Co., Ltd. located in Zhejiang province, China. Fresh tea leaves were plucked at the standardized criteria of one bud with two leaves. The manufacturing procedure followed the sequential steps of spreading, first panning, cooling, second panning and final panning in accordance with the national standard of technique regulation for Longjing tea processing (DB33/T 239-2023). The harvested fresh tea leaves were placed indoors at a temperature of $25\text{ }^{\circ}\text{C} \pm 5\text{ }^{\circ}\text{C}$ and a relative humidity of 60 % for a duration of 8–12 h. Well-spread leaves were subsequently fixed through pan-firing in a roasting machine for flat tea at $180\text{--}200\text{ }^{\circ}\text{C}$ for around 4 min. The fixed leaves were rapidly cooled to ambient temperature and then stacked up to a height of approximately 10–15 cm for a duration of around 2 h. The softened leaves were subsequently subjected to second panning in a separate roasting machine for flat tea, operating within a temperature range of $170\text{--}180\text{ }^{\circ}\text{C}$ and lasting approximately 3 min. Finally, the final panning was carried out using roller-roasting machine at a temperature of $90\text{ }^{\circ}\text{C}$ for a period of 30 min till the moisture content of tea reached below 6.5 %. The tea samples were prepared for further treatment.

The samples were packaged using aluminum foil composite bags (PET/AL/PE, 6 cm \times 20 cm, Guangzhou Yijie Packaging Factory, Guangdong, China). The packaging bags has low oxygen permeability ($0.72 \pm 0.18\text{ cm}^3/(\text{m}^2 \cdot 24\text{h} \cdot 0.1\text{ MPa})$), low water vapor transmittance ($1.15\text{ g} \pm 0.03\text{ g}/(\text{m}^2 \cdot 24\text{h})$) and lighttight ($0.0 \pm 0.0\%$) according to the material analysis (QingXi Technology Research Institute). Each package contained 50 g of Longjing tea, and the bag opening was sealed after removing excess air. The samples in oxygen scavenger treatment group (OS) were individually packaged with two packs of reduced-iron oxygen scavenger (3.0 g per pack, with a total oxygen absorbed capacity of 450 mL/pack, Xinchang Qunxing Industry Co., Ltd.) prior to sealing the bags. The samples in non-oxygen scavenger treatment group (NS) were directly packed into the packaging bag and hermetically sealed with air evacuation. All the packaged Longjing tea samples were stored

in dark for a duration of 18 months. Storage temperatures were $-20\text{ }^{\circ}\text{C}$, $5\text{ }^{\circ}\text{C}$ and $25\text{ }^{\circ}\text{C}$.

2.2. Sensory evaluation

An amount of 3.0 g each tea sample was brewed with 150 mL boiling water for 4 min, then tea infusion was filtered. The standard evaluation cups with infused leaves were used for sensory evaluation. The sensory evaluation was conducted by a panel of six expert panelists specializing in tea sensory for over 10 years. All the expert panelists demonstrated a high level of expertise in assessing the aroma quality of Longjing tea. Furthermore, the perceptions training for stale odor was conducted using aged Longjing tea sample that had been exposed to ambient temperature for 5 years, which was defined as a level 9 of stale odor. The antithesis of stale odor is freshness, which can be defined as aroma quality that closely resembles the unimpaired retention of freshly processed Longjing tea. Recognition training of freshness was conducted by the aroma reminiscent of crisp green vegetables like cucumber. The aroma quality of tea samples was assessed in accordance with Methodology for Sensory Evaluation of Tea (GB/T 23776, National Standard), encompassing both aroma scoring (100-point scoring system) and description analysis. Quantitative descriptive analysis (QDA) was used for scoring the stale odor of Longjing tea samples based on a scale of 0–9 (0: absent; 9: extremely strong). The final score for stale odor was determined by calculating the mean of the sensory panelists' scores.

2.3. Volatiles extraction

The modified simultaneous distillation extraction (MSDE) was utilized for the extraction of volatile compounds (Fan et al., 2024). The ethanol-reflux condenser was connected on one end to the tea container and on the other end to the solvent evaporator. A steam generator was at the bottom of tea container. The container was loaded with a total of 15 g of tea sample. The water steam generator and extraction solvent evaporator were injected with 250 mL of water and 30 mL of ethyl ether (containing 1 ppm of butylated hydroxytoluene, BHT), and the temperatures were maintained at $100\text{ }^{\circ}\text{C}$ and $40\text{ }^{\circ}\text{C}$ respectively. The BHT serves as an internal standard to rectify errors arising from variations in volume (Yang, Lin, & Choong, 2002). Tea volatile compounds carried out by water steam, were extracted using ethyl ether steam in the extraction chamber and subsequently condensed for 120 min through an ethanol-reflux condenser. Then the collected volatiles-ethyl ether extract was dehydrated using 5.0 g of anhydrous sodium sulfate overnight, till it was concentrated to a volume of 1.0 mL for further analysis. Each experiment of tea sample was performed in triplicate.

2.4. Gas chromatography–mass spectrometry (GC–MS) analysis

The analysis of volatile compounds was performed using a Shimadzu gas chromatograph 2010-plus equipped with a triple quadrupole mass spectrometer QP 2020 (Shimadzu, Tokyo, Japan). The SH-Rxi-5Sil MS capillary column (0.25 mm \times 30 m, 0.25 μm , Shimadzu, Tokyo, Japan) was employed. GC conditions were set according to our previous study (Fan et al., 2024): injection port temperature at $250\text{ }^{\circ}\text{C}$ with split-less mode and injection volume 1.0 μL . High purity helium ($> 99.999\%$) was used as carrier gas with a flow rate of 1 mL min^{-1} . The oven temperature followed a linear gradient change: it was maintained at $50\text{ }^{\circ}\text{C}$ from 0 to 5 min, increased from $50\text{ }^{\circ}\text{C}$ to $210\text{ }^{\circ}\text{C}$ between 5 and 53.5 min, maintained at $210\text{ }^{\circ}\text{C}$ from 53.5 to 58.5 min, increased from $210\text{ }^{\circ}\text{C}$ to $230\text{ }^{\circ}\text{C}$ between 58.5 and 60.0 min and then maintained at $230\text{ }^{\circ}\text{C}$ for the last 5 min. Mass spectrometer conditions were as follows: ion source temperature was set to $250\text{ }^{\circ}\text{C}$, electron energy 70 eV, solvent delay time 3 min and full scan range from 35 to 450 amu. The raw data was deconvoluted using LabSolutions GC–MS solution (4.45 SP1, Shimadzu, Tokyo, Japan). Initially filter criteria were implemented to screen the potential volatile compounds based on retain frequency ($> 60\%$) and

variability coefficient (< 25 %). Identification of volatile compounds were achieved by comparing the mass spectra with the National Institute of Standards and Technology (NIST) library (a similarity threshold set above 70 %) and the retention index. The identified volatiles were quantified by an internal standard BHT as follow:

$$\text{Peak area ratio (PAR)} = \frac{\text{Peak area of target}}{\text{Peak area of BHT}}$$

2.5. Statistical analysis

All analyses were replicated thrice. The principal components analysis (PCA), hierarchical cluster analysis (HCA) and heatmap drawing were performed using the OmicShare tool, an online platform for data analysis (<https://www.omicshare.com/tools>). Venn diagram was created using Adobe photoshop 2023. Multiple factor analysis (MFA) was employed for integrating datasets to investigate the associations between aroma quality and key volatile compounds, using XLSTAT 2019 (Addinsoft, New York, NY, USA). The correlation index was calculated by Spearman's correlation method.

3. Results

3.1. Storage temperature and oxygen scavenger impacted the aroma quality of Longjing teas

The aroma characteristics and scores of Longjing tea varied significantly under various storage conditions, as demonstrated in Table 1. After being stored at -20°C for 18 months, Longjing tea maintained a high level of freshness and stale aroma quality, with the score of 82.5. The deterioration of aroma quality in Longjing tea was significantly accelerated during storage at 5°C and 25°C . The aroma quality average scores of Longjing tea packaged with oxygen scavenger were 81.0 and 79.8, whereas the sample packaged without oxygen scavenger achieved average scores of 77.5 and 76.7 after a storage period of 18 months at 5°C and 25°C . The stale odor average scores of 5°C-NS and 25°C-NS were significantly higher at 6.0 and 7.8, respectively, compared to those of 5°C-OS (1.5) and 25°C-OS (4.3). Comparative analysis revealed a positive correlation between storage temperature and the intensity of stale odor during the same storage duration. The findings were consistent with previous research indicating that tea stored at -20°C maintained its freshness aroma, while a noticeable decline in tea freshness occurs when stored at room temperature (Dai et al., 2019). It was interesting that the aroma quality of Longjing tea packaged with oxygen scavenger and stored at temperature of 5°C , exhibited remarkable consistency compared to those stored at -20°C . The stale odor scores of 5°C-OS samples were below 2.0. The 25°C-OS samples exhibited a moderate intensity of stale odor (average 4.3), which was significantly lower than the stale odor intensities observed in the 5°C-NS (6.0) and 25°C-NS (7.8). The findings indicated the utilization of oxygen scavenger could effectively mitigate aroma deterioration caused by elevated temperature, resulting in a decrease in the stale odor scores by more than 3.0.

3.2. Overall determination of volatiles in Longjing teas

A total of 158 volatile metabolites belong to 12 categories were

identified from all samples, including oxygen heterocyclic compounds, esters, alkenes, ketones, acids, aldehydes, phenols, aryl compounds, alcohols, pyrroles and others (Fig. 1A). The alcohols and oxygen heterocyclic compounds exhibited the highest proportion of relative peak areas, ranging from 25.88 % to 39.54 % and 13.47 % to 18.17 %. The findings was consistent with previous research indicating that alcohol is an important category of volatile compounds and plays a crucial role in the aromatic profile of Longjing tea. However, the composition of alcohols is complex and its contribution varies noticeably due to cultivar, processing and other factors (Gong et al., 2017; Wang, Liu, et al., 2020; Wang, Ma, et al., 2020). After being stored for 18 months, a significant increase in the proportions of alcohols, ketones and oxygen heterocyclic compounds was observed when the storage temperature exceeded 0°C . Conversely, a decrease in the proportion of acids was noted. The proportions of pyrroles and oxygen heterocyclic compounds were found to be higher in 5°C-OS & 25°C-OS compared to 5°C-NS & 25°C-NS respectively, whereas the proportions of alcohols, aldehydes and alkanes were observed to be higher in 5°C-NS & 25°C-NS than in 5°C-OS & 25°C-OS .

Venn diagram (Fig. 1B) was employed for illustrating the differences in volatile compounds of Longjing tea samples, investigating the impact of storage temperature and presence/absence oxygen scavenger on the aroma compounds. There were 79, 122 and 122 volatile compounds identified in Longjing teas storage at -20°C , 5°C and 25°C respectively. Samples storage at different temperatures shared 58 volatile compounds, while 92 volatile compounds were co-detected in samples stored at temperature of 5°C and 25°C . This suggested the storage temperature exerted a significant influence on the volatile compound profiles. The 5°C-OS shared 70 volatile compounds with 5°C-NS , while the 25°C-OS and 25°C-NS shared 56 volatile compounds. This finding indicated that the utilization of oxygen scavenger had a significant impact on altering the volatile profiles of Longjing tea.

3.3. The effect of storage temperature on volatile compounds in Longjing tea

There were 65 volatile compounds found to be associated with storage temperature based on the analysis of relative contents of volatile compounds at different temperature. PCA and HCA were performed based on these 65 volatile compounds. In PCA score plot (Fig. 2A), the Longjing tea samples stored at -20°C , 5°C and 25°C were distinctly distributed in separate quadrants. According to the result of HCA (Fig. 2B), 25°C-NS , 5°C-NS and -20°C-NS were separated into two groups with 25°C-NS and 5°C-NS grouped together, while -20°C-NS segregated. Both the PCA score plot and HCA dendrogram indicated that storage temperature could significantly alter the volatile compounds, thereby influencing the aroma quality performance of Longjing tea.

A heatmap (Fig. 2C) was plotted to discriminate Longjing tea samples stored at -20°C , 5°C and 25°C based on the relative content of these volatile compounds. The color of the scale bar in the heatmap represented the volatiles levels ranging from highest (pink) to lowest (green), which demonstrated variations among sample groups. As shown in Fig. 2C, all the 65 volatile compounds were grouped into four groups. The level of volatile compounds were higher in 25°C storage samples, including 8 ketones, 5 alcohols, 6 alkanes, 4 others, 2 esters, 1 oxygen heterocyclic compound, 1 phenol, 1 alkene and 1 aryl compound.

Table 1
Sensory properties of Longjing teas under different storage conditions.

Item	-20°C-NS	5°C-OS	5°C-NS	25°C-OS	25°C-NS
Aroma scores	82.5 ± 1.0^a	81.0 ± 1.3^{ab}	77.5 ± 0.5^c	79.8 ± 1.2^b	76.7 ± 1.0^c
Stale odor score	0.2 ± 0.3^e	1.5 ± 0.5^d	6.0 ± 0.5^b	4.3 ± 0.4^c	7.8 ± 0.5^a
Sensory description	Fresh and pure aroma	Fresh and pure aroma	Obvious off-flavo Obvious stale odor	Little fresh aroma Little stale odor	Obvious off-flavor Serious stale odor

Note: Values are given as mean ($n = 6$). Means with different letters represent significant difference ($p < 0.05$).

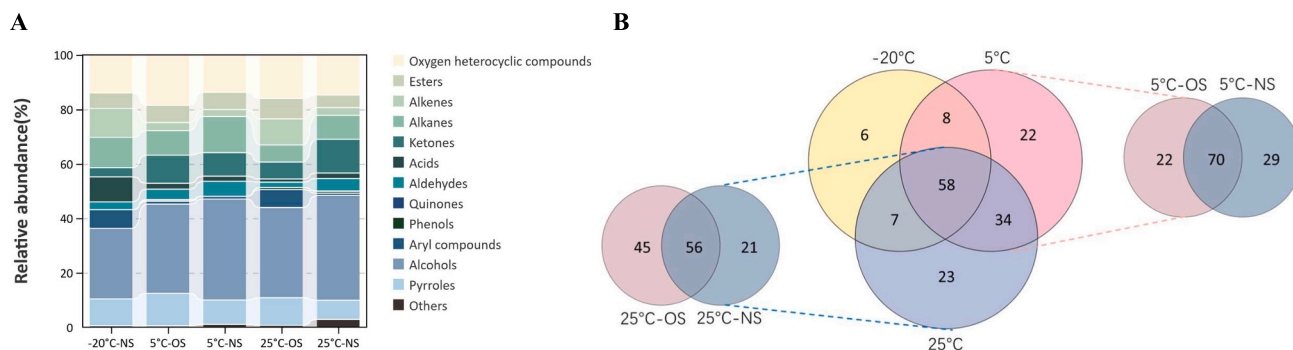


Fig. 1. Comparison of composition (A) and venn diagram (B) of aroma compounds in Longjing tea under different treatments.

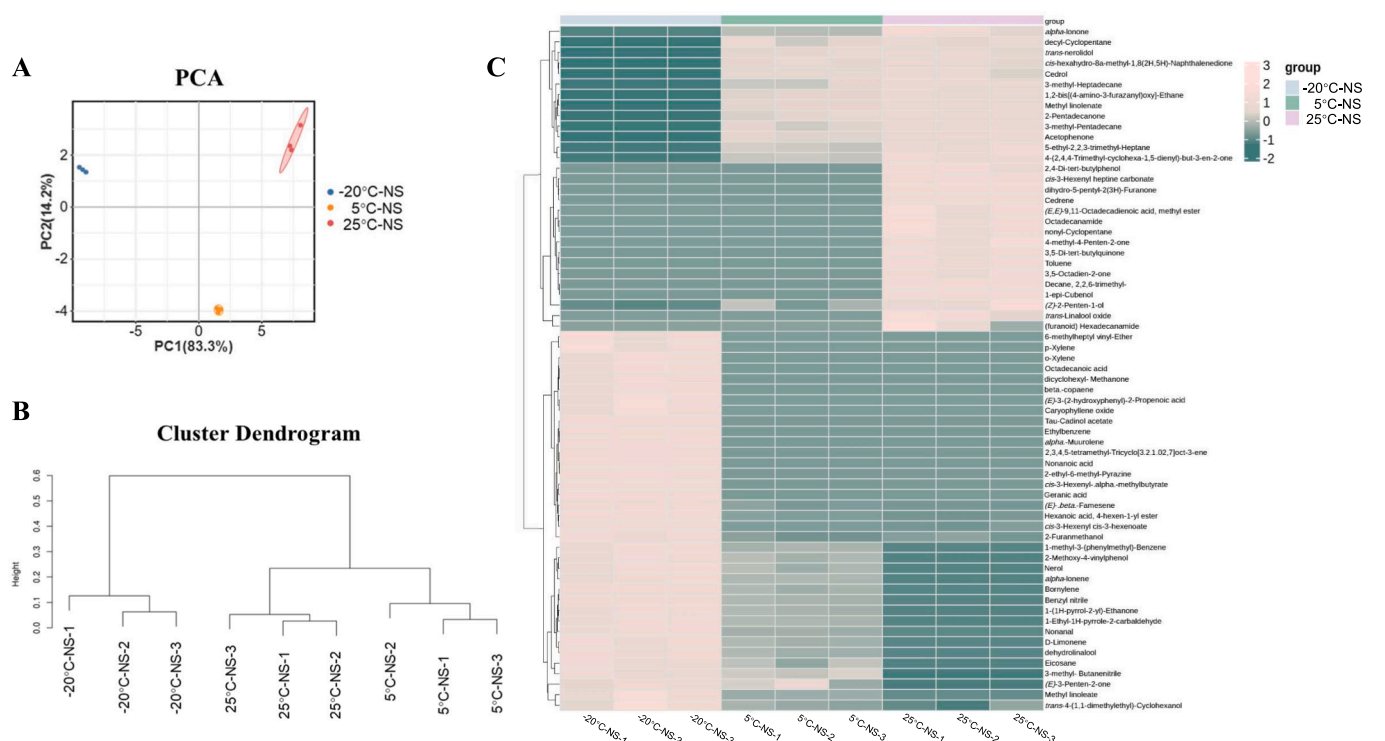


Fig. 2. Multivariate analysis of Longjing tea stored at different temperatures. (A) Heatmap of volatile compounds related storage temperatures. (B) Principal component analysis (PCA) of Longjing tea samples based on volatiles related storage temperatures. (C) Cluster dendrogram of samples based on volatiles related storage temperatures.

Longjing tea stored at -20°C exhibited higher level of volatiles including 7 alkenes, 4 esters, 4 acids, 4 aryl compounds, 3 oxygen heterocyclic compounds, 3 ketones, 3 pyrroles, 3 alcohols, 2 others, 1 aldehyde, 1 alkane and 1 phenol. The results indicated that elevated temperature reduced the levels of alkenes in Longjing tea such as *.beta.-copaene*, *.alpha.-muurolene*, *(E)-.beta.-famesene*, *alpha-ionene*, which are characterized by floral and fruity aroma (Li, Zhu, & Wang, 2017; Zhang et al., 2022). However, the levels of ketones including *alpha-ionone*, 2-pentadecanone, acetophenone, dihydro-5-pentyl-2(3H)-furanone and 3,5-octadien-2-one, as well as alcohols such as *trans-nerolidol*, *cedrol*, 1-*epi-cubenol*, *(Z)*-2-penten-1-ol and *trans-Linalool oxide* (furanoid) were observed to increase with rising storage temperatures. Most of these volatile compounds are identified as woody or fatty note (Dai et al., 2020; Shi et al., 2021). These findings partially support previous researches indicating that the increase of ketones and alcohols contributes to the development of stale odor during storage (Liu et al., 2023).

3.4. The effect of oxygen scavenger on temperature-induced volatile compounds in Longjing tea

There were 29 volatile compounds responding to the presence/absence of oxygen scavenger screened from storage temperature-related volatile compounds. Fig. 3 showed the relative peak area of these volatile compounds in Longjing tea samples with/without oxygen scavenger stored at 5°C and 25°C . The presence of oxygen scavenger in packaged Longjing tea obviously maintained a higher level of alkenes (*(E)-.beta.-famesene* and *.alpha.-muurolene*), acids (nonanoic acid, geranic acid and *(E)*-3-(2-hydroxyphenyl)-2-propenoic acid), pyrroles (1-ethyl-1H-pyrrole-2-carbaldehyde and 1-(1H-pyrrol-2-yl)-Ethanone), as well as oxygen heterocyclic compounds (2-furanmethanol, 2-methoxy-4-vinylphenol and caryophyllene oxide). Whereas the concentrations of alkanes (decane, 2,2,6-trimethyl- and 5-ethyl-2,2,3-trimethyl-heptane), esters (*cis*-3-hexenyl heptene carbonate and *(E,E)*-9,11-Octadecadienoic acid, methyl ester), as well as amides (hexadecanamide and octadecanamide) were found to be higher in samples packaged without

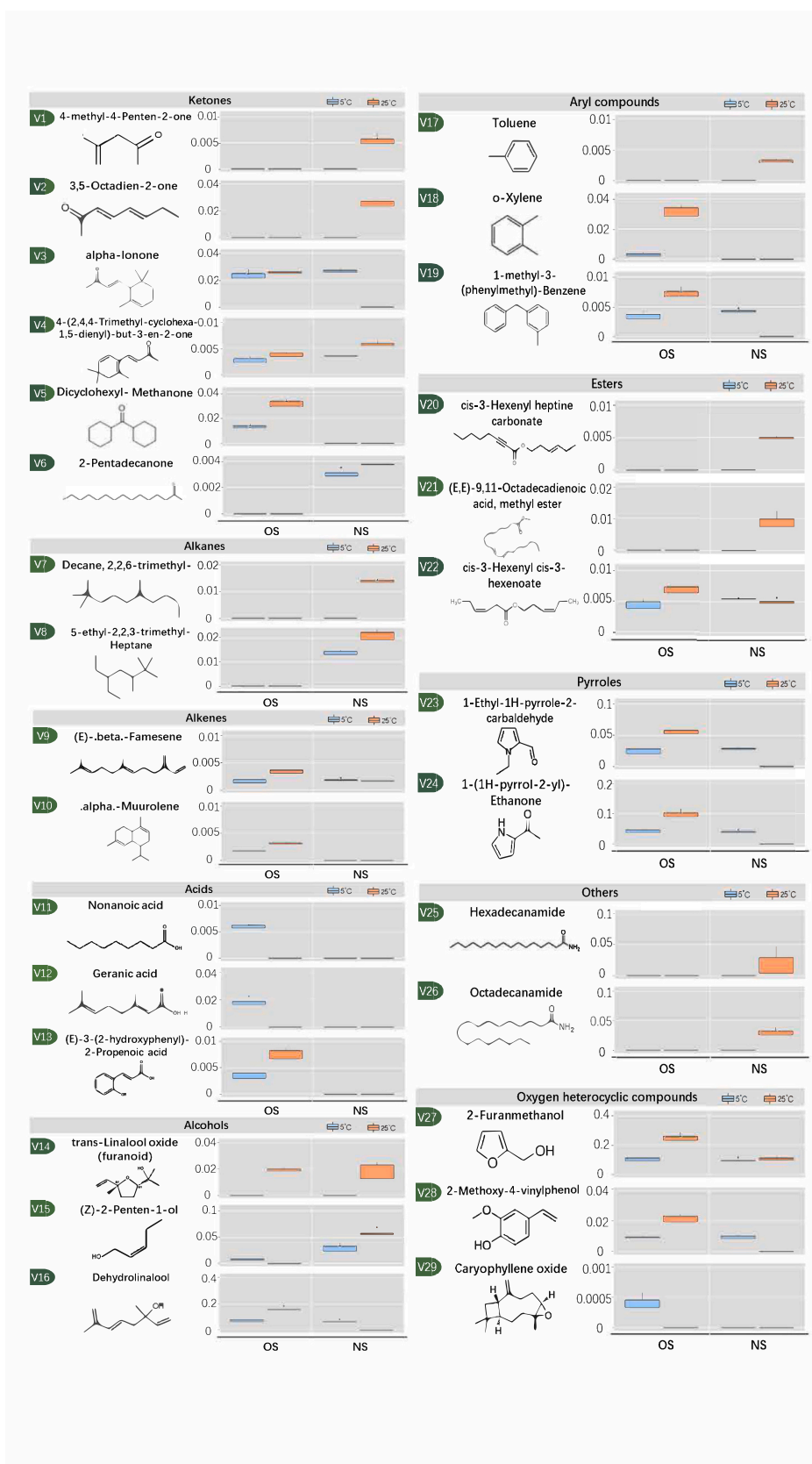


Fig. 3. The Box-plot illustrating the levels of key volatiles in Longjing tea samples packaged at the presence/absence of oxygen scavenger.

oxygen scavenger. Among the six key ketones, the level of dicyclohexylmethanone was higher in the presence of oxygen scavenger in package, while other ketones (4-methyl-4-penten-2-one, 3,5-octadien-2-one, 4-(2,4,4-trimethyl-cyclohexa-1,5-dienyl)-but-3-en-2-one and 2-pentadecanone) showed the opposite trend. Furthermore, in the case of alcohols, the presence of oxygen scavenger could effectively enhance the retention of dehydrolinalool, while simultaneously reducing the levels of *trans*-Linalool oxide (furanoid) and (*Z*)-2-Penten-1-ol. The findings indicated that the utilization of oxygen scavengers effectively mitigated the decline in alkenes, pyrroles and oxygen heterocyclic compounds induced by elevated temperature, while also restraining the increase in alkane, ester, and ketone contents when subjected to elevated storage temperature. PCA score plot, based on these 29 key volatile compounds (Fig. 4), demonstrated that the principal components account for a total of 86.6 % of variances, with PC1 explaining 62.8 % and PC2 explaining 23.8 %. The samples of 5 °C-NS, 5 °C-OS and 25 °C-OS were located on the negative axis of PC1; however, 25 °C-NS samples were positioned along the positive axis of PC1. Therefore, the utilization of oxygen scavenger exerted a significant influence on the volatile compounds at the same storage temperature, resulting in a notable alteration in the aroma substance profile of the sample.

3.5. The association between aroma sensory traits and oxygen scavenger-related volatile compounds

MFA was employed to investigate the correlations among utilization of oxygen scavenger, aroma sensory score, stale odor score and key oxygen scavenger-related volatile compounds. The loading plot of MFA showed 90.95 % of the total variation explained by the first two factors ($F_1 = 75.63\%$, $F_2 = 15.33\%$). As shown in Fig. 5A, oxygen scavenger-OS and aroma sensory scores were positioned on the positive end of the horizontal axis, whereas oxygen scavenger-NS and stale odor score were situated towards the negative end of the horizontal axis. The majority of the key volatile compounds were distributed around sensory traits, among which approximately 83 % of the volatiles exhibited an absolute value above 0.5 in the direction of F_1 . This finding indicated a strong association between the majority of the volatiles related to utilization of oxygen scavenger and the sensory traits. Furthermore, the correlation between sensory traits and key volatile compounds, as indicated by the Pearson correlation coefficient, is illustrated in Fig. 5B. All the screened volatile compounds related to utilization of oxygen scavenger showed significant ($p < 0.05$) correlations with sensory traits. Four volatile compounds, including 4-(2,4,4-Trimethyl-cyclohexa-1,5-dienyl)-but-3-en-2-one, *alpha*-ionone, 2-pentadecanone and 5-ethyl-2,2,3-trimethylheptane, exhibited extremely significant ($p < 0.001$) positive correlations with stale odor score but negative correlations with aroma sensory

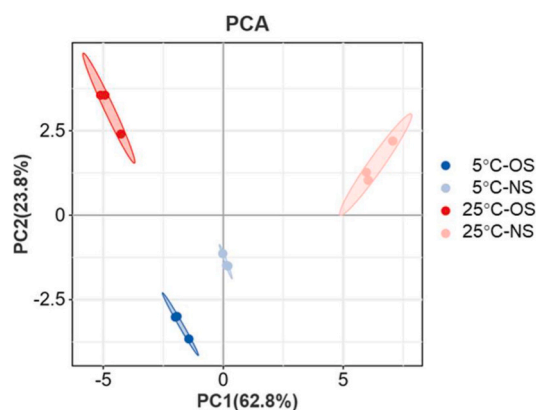


Fig. 4. Principal component analysis (PCA) of Longjing tea samples based on volatiles responding to the presence/absence of oxygen scavengers screened from storage temperature-related volatile compounds.

score. The stale odor score also showed a significant positive association ($p < 0.01$) with the volatile compounds (*Z*)-2-penten-1-ol, octadecanamide, 4-methyl-4-penten-2-one, toluene, 3,5-octadien-2-one, decane, 2,2,6-trimethyl- as well as *cis*-3-hexenyl heptene carbonate. In contrast, there were nine volatile compounds revealed extremely significant ($p < 0.001$) positive correlations with aroma sensory score, including 1-(1H-pyrrol-2-yl)-ethanone, caryophyllene oxide, *alpha*-muurolene, nonanoic acid, geranic acid, (*E*)-3-(2-hydroxyphenyl)-2-propenoic acid, 1-methyl-3-(phenylmethyl)-benzene, dehydrolinalool and 1-ethyl-1H-pyrrole-2-carbaldehyde. The caryophyllene oxide, *alpha*-muurolene, nonanoic acid and geranic acid were found to be negatively ($p < 0.001$) associated with stale odor score.

4. Discussion

Temperature is a key determinant of flavor deterioration of green tea during storage (Dai et al., 2019; Dai et al., 2020), as many chemical transformations are temperature-dependent (Li, Taylor, & Mauer, 2011). According to reports, the recommended storage temperature for green tea is $-20\text{ }^{\circ}\text{C}$ and below, as it effectively preserves a comparable freshness level of the tea prior to storage (Dai et al., 2019). Whereas an obvious decrease on tea freshness flavor was found in tea stored above $0\text{ }^{\circ}\text{C}$, particularly at higher temperatures where the deterioration occurred more faster (Dai et al., 2019). In the present study, the same trend was found in Longjing tea that storing tea especially at a temperature of $25\text{ }^{\circ}\text{C}$ resulted in a significant decline in aroma sensory scores and an increase in stale odor scores compared to storage at $-20\text{ }^{\circ}\text{C}$. The higher aroma scores and lower stale odor scores observed in $5\text{ }^{\circ}\text{C}$ -OS and $25\text{ }^{\circ}\text{C}$ -OS, compared to $5\text{ }^{\circ}\text{C}$ -NS and $25\text{ }^{\circ}\text{C}$ -NS respectively, along with the closer sensory scores obtained from two sets of comparison samples ($-20\text{ }^{\circ}\text{C}$ -NS & $5\text{ }^{\circ}\text{C}$ -OS, $5\text{ }^{\circ}\text{C}$ -NS & $25\text{ }^{\circ}\text{C}$ -OS; Table 1), suggested that the utilization of oxygen scavenger effectively mitigated aroma deterioration caused by elevated temperature. For volatile compounds, the elevated storage temperature obviously led to an increase the levels of specific ketones and alcohols in Longjing tea, while the utilization of oxygen scavenger in the packaging significantly reduced the levels of these volatile compounds in this study. Five ketones were identified as being associated with the stale odor, namely 4-(2,4,4-Trimethyl-cyclohexa-1,5-dienyl)-but-3-en-2-one, *alpha*-ionone, 2-pentadecanone, 4-methyl-4-penten-2-one and 3,5-octadien-2-one. The volatile *alpha*-ionone, which possesses a woody aroma, has been identified as one of the thermal degradation products derived from carotenoids (Ho, Zheng, & Li, 2015; Meng et al., 2021). The increasing of 2-pentadecanone (greasy) and 3,5-octadien-2-one (fatty) during storage are usually associated with lipid oxidation (Ho et al., 2015; Kakko, Damerau, Rios, Laaksonen, & Yang, 2023; Xi et al., 2023). The alcohol compound (*Z*)-2-penten-1-ol (green) was identified as another stale odor-related volatile compound, which increase during storage at elevated temperature due to lipid oxidation. It has been reported to be one of oxidative markers used for distinguish the level of oxidation (Grilo & Wang, 2021; Liu et al., 2023). In conclusion, the utilization of oxygen scavenger in the packaging of Longjing tea effectively inhibits the accumulation of stale odor volatiles by mitigating thermal oxidation/degradation.

On the contrary, the utilization of oxygen scavenger effectively preserved the volatiles in Longjing tea that are related to aroma sensory scores, including alkenes such as *alpha*-muurolene and caryophyllene oxide, acids like nonanoic acid, geranic acid and (*E*)-3-(2-hydroxyphenyl)-2-propenoic acid, alcohols like dehydrolinalool, as well as pyrroles such as (*E*)-3-(2-hydroxyphenyl)-2-propenoic acid and 1-ethyl-1H-pyrrole-2-carbaldehyde. Among these volatile compounds, (*E*)-3-(2-hydroxyphenyl)-2-propenoic acid (also called (*E*)-2-hydroxycinnamic acid) and *alpha*-muurolene exhibit a distinct wakame-like flavor and woody aroma respectively, which is associated with the maturity of tea shoots (Oyaizu, Shimoda, Matsumoto, & Goto, 2002; Zheng et al., 2024). 1-(1H-pyrrol-2-yl)-ethanone (nut flavor), 1-ethyl-1H-pyrrole-2-carbaldehyde (roasted aroma), geranic acid (green flavor) and

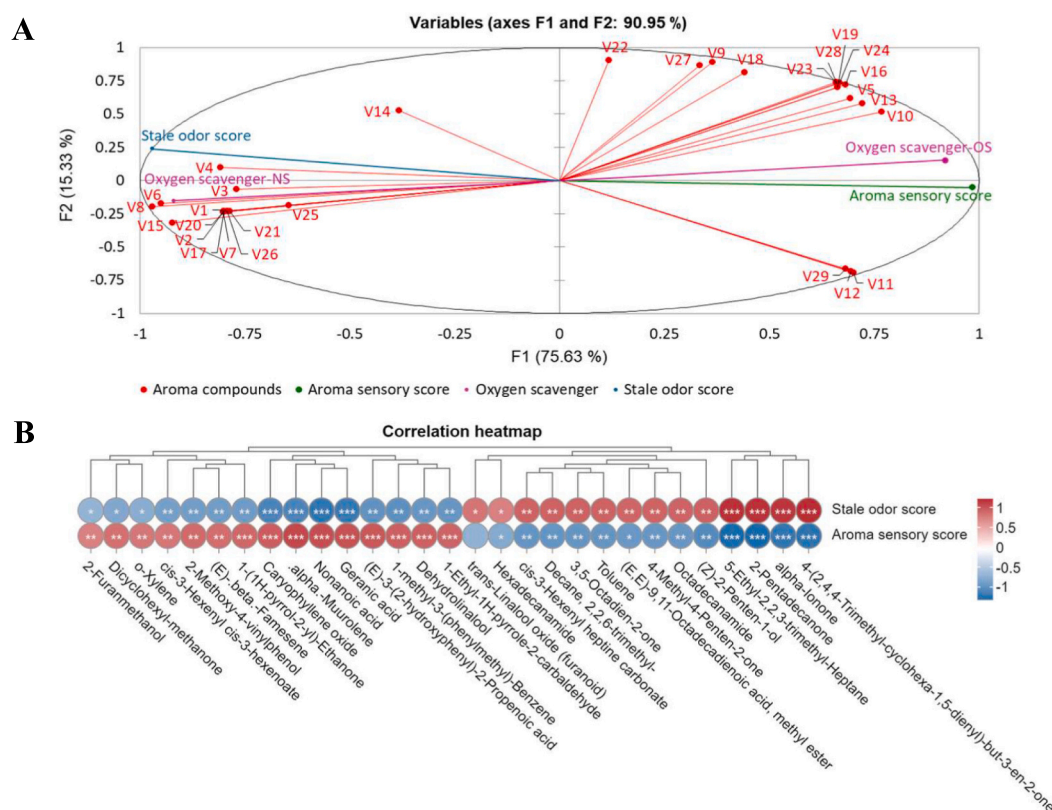


Fig. 5. Multiple factor analysis (MFA) loading plot (A) and correlation map (B) of sensory traits and key volatile compounds. *Correlation is significant between 0.01 and 0.05, ** Correlation is significant between 0.001 and 0.01, and ***correlation is significant at the 0.001 level for Pearson correlation coefficient.

dehydrolinalool (lavender-like fragrance) are formed during the drying process (Li & Wang, 2020; Wen et al., 2023; Yang, Qian, Deng, Yuan, & Jiang, 2022; Zhou, Zhang, Liu, Zhang, & Wu, 2023). These volatile compounds, derived from fresh leaves and processing technology, provided important material basis for the freshness aroma of Longjing tea.

5. Conclusions

Longjing teas are vulnerable to aroma deterioration during storage, especially when exposed to elevated temperatures. The utilization of oxygen scavenger could significantly enhance the aroma quality scores and decrease the scores for stale odor, when stored at 5 °C and 25 °C. A total of 29 volatile compounds were identified as responsive to the presence/absence of oxygen scavenger during storage at elevated temperatures. Volatile compounds resulting from thermal carotenoids degradation/lipid oxidation, including 4-(2,4,4-trimethyl-cyclohexa-1,5-dienyl)-but-3-en-2-one, *alpha*-ionone, 2-pentadecanone, 3,5-octadien-2-one, 4-methyl-4-penten-2-one and (*Z*)-2-penten-1-ol were identified as key contributors to stale odor of Longjing tea. These compounds were found to increase when stored at elevated temperatures but were significantly inhibited by utilization of oxygen scavenger. However, the key contributors to aroma sensory, including 1-(1H-pyrrol-2-yl)-ethanone, caryophyllene oxide, *alpha*-murolene, nonanoic acid, geranic acid, (*E*)-3-(2-hydroxyphenyl)-2-propenoic acid, dehydrolinalool and 1-ethyl-1H-pyrrole-2-carbaldehyde, exhibited higher concentration when stored with oxygen scavenger compared to storage without oxygen scavenger. The oxygen scavenger achieved aroma-preserving effect by inhibiting key stale odor compounds and maintain aroma quality related volatiles.

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CRediT authorship contribution statement

Jiawei Wang: Writing – original draft, Formal analysis. **Yingxin Xu:** Methodology. **Chang Xu:** Methodology. **Hongying You:** Resources. **Tonghua Xie:** Investigation. **Haowei Guo:** Data curation. **Ping Chen:** Investigation. **Qiang Chu:** Data curation. **Shuying Gong:** Supervision, Conceptualization. **Fangyuan Fan:** Writing – review & editing, Project administration.

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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