Effects of micro-oxygenation on the aroma of Cabernet Sauvignon wine from Ningxia, China

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Abstract: Micro-oxygenation (MOX) is an effective post-harvest technique for the flavor improvement of grape wine. This study investigated the effect of MOX on the aroma quality of Ningxia wine for the first time. Three sub-region Cabernet Sauvignon dry red wines were treated with different levels of oxygen before or after malolactic fermentation. The wine aroma was analyzed through gas chromatography-mass spectrometry (GC-MS) and quantitative descriptive analysis (QDA) after six months of aging. The data obtained demonstrated that the dose and timing of oxygen addition were key factors influencing the effectiveness of MOX. The most noticeable modifications in wine aroma compounds were generated by an oxygen dosage of 30 (mL/L)/month added before malolactic fermentation. Predominantly, the concentrations of 2-phenylethanol, benzaldehyde, diacetyl, and 2,3-pentanedione showed an increased pattern upon MOX treatments. The sensory analysis revealed that MOX improved the aroma quality of wine by decreasing green and animal odors, meanwhile enhancing the olfactory intensities of dried fruits, flowers, and nuts. This work confirmed that MOX was suitable for aroma modification of Cabernet Sauvignon dry red wine from Ningxia and established a preliminary MOX procedure that can serve as a reference for future applications.
Keywords: grape wine, micro-oxygenation, volatile organic compounds, aroma profiles, Ningxia
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1 Introduction

Aroma is a critical rating criterion for wine style and quality. The concentration, class, and volatility of volatile organic compounds (VOCs) define the aroma profile of grape wine. Over 1000 VOCs have been discovered, with several of them proving to have a substantial impact on wine fragrance[1]. The composition of VOCs is relevant to grape variety, climate, soil, and, most importantly, the brewing techniques (fermentation and aging)[2,3].

Oxygen plays a decisive role in wine fermentation and aging. Excessive or insufficient oxygen may result in unpleasant flavors, and an appropriate level of oxygenation is a fundamental aspect of producing high-quality wines[4]. To accurately control oxidation, micro-oxygenation (MOX) has been employed in the brewing process. MOX is the process of deliberately introducing trace amounts of oxygen into red wine, either continuously or intermittently, to accelerate maturation and improve the sensory quality of the wine. This technique was successfully implemented in southern France in the early 1990s, authorized for application in Europe by the European Commission in 1996, and has been extensively used in many wine regions in recent years[5,6].

Red wine can be treated with MOX before or soon after malolactic fermentation (MLF) or during the long-term aging stage[7]. Not only may pre- or post-MLF MOX improve wine color stability and softness[8,9], but it is also reliable to modify the aroma properties[4,10,11]. Previous research has shown that pre-MLF MOX significantly increased concentrations of succinic derivatives and long-chain esters in a Cencibel wine[12], as well as C13-norisoprenoids and terpenes in a Merlot wine[13]. When treated with post-MLF MOX, a considerable improvement of C6 alcohols, terpenes, and lactones was observed in red wines[14]. It was also demonstrated that MOX could eliminate undesirable reductive notes and diminish the vegetative odors of wine[5,15]. When combined with oak chips, MOX could mimic oak barrel aging of red wine to save cost and boost the complexity of aroma by amplifying the scents of fruits, spices, nuts, and tobaccos[5,13,16].

The Eastern Foot of Helan Mountain (EFHM) in Ningxia is a world-class wine region and one of China's Wine Geographical Indication Product Protection Areas. EFHM is located on a long and narrow plain between Helan Mountain and the Yellow River in Northwest China (105°45′E-106°27′E, 37°43′N-39°05′N)[17]. With plenty of sunshine and gravel soil, the environment of EFHM is favorable to red grape growing, and *Vitis vinifera* L. cv. Cabernet Sauvignon is the dominating cultivar. However, there is a grave issue that the aroma of Cabernet Sauvignon wine is of weak complexity and typicality. Although MOX has been widely

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utilized for aroma improvement in many other wine regions[5], to the general knowledge, a lack of research has examined the influence of MOX on the wine aroma in Ningxia. Therefore, applying MOX to the wine-producing craft is expected to be a new oenological way to fill these gaps in this region. The current study involves an investigation of the effects of different levels of MOX treatments performed before or after MLF on the VOCs and olfactory profiles of Cabernet Sauvignon dry red wines from three sub-regions of Ningxia. The findings of this work would aid in demonstrating the potential utility of MOX in this location.

2 Materials and methods

2.1 Winemaking

Three single-variety dry red wines (2018 vintage) were elaborated from Cabernet Sauvignon grapes harvested from three representational sub-regions in Ningxia, labeled Ganchengzi (GCZ, 105°88′E, 38°08′N, a 5-year vineyard), Yinchuan (YC, 106°05′E, 38°56′N, a 4-year vineyard), and Yongning (YN, 105°97′E, 38°28′N, a 6-year vineyard). Meteorological conditions were reported for the GCZ, YC, and YN sub-regions in 2018, with the annual sunshine duration of 2705.4 h, 2800.1 h, and 2864.7 h, the accumulated temperature during the growing season (≥10°C) of 3617 ℃, 3849 ℃, and 3951.4 ℃, and the annual precipitation of 162.3 mm, 280.2 mm, and 256.3 mm, respectively (The data are from the Ningxia Institute of Meteorological Sciences).

The grapes were fermented in a 200 L stainless steel tank through the same vinification procedure at Ningxia University. After destemming and crushing, grapes were treated with 40 mg/L sulfur dioxide and 20 mg/L pectinases (Vinozym Vintage FCE, Lamothe-Abiet, Bordeaux, France). An alcoholic fermentation (AF) was then performed at 28 ℃ for seven days by 200 mg/L of a commercial yeast strain of *Saccharomyces cerevisiae* (Excellence XR, Lamothe-Abiet, Bordeaux, France). The fermentation course was monitored by measuring the specific gravity of the broth every 12 h (Figure S1). AF was usually considered to be completed when the specific gravity reached below 0.995 and residual sugar below 4.0 g/L. Following AF, each sub-region wine was purged with pure nitrogen gas for 15 min to remove carbon dioxide, then homogenized in a 5 L narrow-mouth glass container filled with nitrogen and carefully avoided oxygen incorporation. The containers were tightly closed by silicone stoppers with vacuum grease around the margins and sealed with screw covers.

2.2 Micro-oxygenation treatment

MOX treatment was carried out either before or after MLF. As part of the pre-MLF MOX therapy, oxygen doses of 10, 20, and 30 (mL/L)/month were supplied for ten days at 20 ℃. During this time, 200 mg/L lysozymes were used to impede the natural development of MLF. MLF was initiated with 10 mg/L of a commercial lactic acid bacteria strain of *Oenococcus oeni* (*Oeno* 1, Lamothe-Abiet, Bordeaux, France) and was tracked by measuring malic acid and lactic acid. The oxygen dosages for the post-MLF MOX treatments were 1, 5, and 8 (mL/L)/month for two weeks at the same ambient temperature of 20 ℃. The control wine received no oxygen treatment before or after MLF. Table 1 depicts the experimental design.

Pechamat et al.[18] developed the method of oxygen addition. With a syringe, measured ultrahigh-purity oxygen was introduced into the containers by puncturing the silicone stopper. After that, gently shake the wine to guarantee complete oxygen dissolution, then tighten the screw covers under nitrogen protection. Oxygen was added one time every two days. Each treatment was performed in triplicate. After the MOX treatment, all the wines were sulfated to reach a total sulfur dioxide concentration of 60 mg/L and were bottled under nitrogen protection. The bottled wines were aged in the cellar for six months at an optimum temperature (16±2) ℃ and humidity (65±5)% before being analyzed.

Table 1 Dose and timing of oxygen addition

Treatments	Oxygen dose/ (mL·L^{-1}) month^{-1}		Duration/d
	Before MLF	After MLF	
NM	0	0	0
B10	10	0	10
B20	20	0	10
B30	30	0	10
A1	0	1	14
A5	0	5	14
A8	0	8	14

Note: NM represents the control group without MOX treatment; B10, B20, and B30, the oxygen doses of 10, 20, and 30 (mL/L)/month before MLF; A1, A5, and A8, the oxygen doses of 1, 5, and 8 (mL/L)/month after MLF; MLF: Malolactic fermentation; MOX: Micro-oxygenation.

2.3 Conventional analysis of grape must and wine

Total sugar (g/L), titratable acidity (expressed as g/L of tartaric acid), alcoholicity (%, v/v), dry extract (g/L), and volatile acidity (expressed as g/L of acetic acid) were measured according to the OIV Compendium of International Methods of Wine and Must Analysis (2008). pH was measured with a PHS-2F pH meter (INESA, Shanghai, China). Total phenol (expressed as g/L of gallic acid) and anthocyanins were determined through a Y15 Enzymatic Auto-analyzer (Biosystems, Barcelona, Spain). Tannin was analyzed as previously reported by Chira et al.[19]. The color intensity and hue were determined using a 1 mm quartz cell.

2.4 GC-MS analysis of VOCs

A CTC PAL autosampler (CTC Analytics, Zwingen, Switzerland) and an Agilent 7890B chromatograph equipped with an Agilent 7000D tripe-quad mass selective detector (Agilent Technologies, Santa Clara, CA, USA) were adopted to analyze VOCs in wines.

The headspace solid-phase micro-extraction coupled with gas chromatography-mass spectrometry (HS-SPME-GC-MS) method was based on previous research with modifications[20-22]. Briefly, a 5 mL aliquot of undiluted wine was pipetted into a 20 mL headspace vial containing 1.5 g sodium chloride. 10 μL of 4-methyl-2-pentanol (1.0083 g/L, TCI, Shanghai, China) was added as the internal standard, with an in-vial concentration of 2.01 mg/L. The vial was sealed with a magnetic PTFE/Sil cap and incubated at 40 ℃ for 5 min. VOCs were extracted in headspace with a DVB/CAR/PDMS fiber (50/30 μm coating, 1 cm, Supelco, Bellefonte, PA, USA) at 40 ℃ for 30 min, at a continuous stirring velocity of 250 r/min, and desorbed at 240 ℃ for 10 min in split-less mode. The GC oven temperature was originally held at 40 ℃ for 3 min, then elevated to 97 ℃ at a rate of 3 ℃/min for 7 min, then ramped at 2 ℃/min up to 120 ℃, then 3 ℃/min up to 150 ℃, and lastly 8 ℃/min up to 220 ℃, before holding for 10 min. The temperature of the transfer line was 230 ℃. The MS electron impact mode was applied, with an electron ionization source temperature of 230 ℃ and electron energy of 70 eV. The solvent delay was 4.4 min.

VOCs were identified through the NIST 17 standard spectral library and further verified with retention indices (RIs) of Alkanes C8 to C20 (Sigma-Aldrich, Shanghai, China) on the DB-Wax

column (30 m×250 μm×0.25 μm). 49 VOCs were analyzed in the selected ion monitoring (SIM) mode (the SIM qualifying ions are listed in Table S1) by a semi-quantitative method (Equation (1))[23,24]:

$$C_{VOC} = \frac{A_{VOC}}{A_{IS}} \times C_{IS} \quad (1)$$

where, C_{VOC} is the concentration of each VOC; A_{VOC} is the peak area of each VOC; A_{IS} is the peak area of the internal standard; C_{IS} is the concentration of the internal standard.

2.5 Sensory analysis of aroma profiles

A panel of 18 judges (eight females and ten males, 20-30 years of age, with at least two years of wine-tasting experience) were trained with the 'Le Nez du Vin' aroma kit (Ease Scent, Beijing, China) over a period of four weeks before the formal sensory evaluation. The training was carried out twice a week for 60 min. At the end of the fourth week, a copy of MOX-treated wine samples was served to the panelists for smelling and discussion. Ten categories of aroma descriptors, including green, fresh fruit, dried fruit, floral, spicy, mushroom, mesothelium, nutty, animal, and woody scents, were determined for quantitative descriptive analysis (QDA). Furthermore, six randomly selected wine samples were scored by the judges in duplicate. The accuracy and repeatability of each person were assessed through the Panel Check software (Version 1.4.2), and finally, 14 panelists (seven females and seven males) passed the checking (Figure S2).

The formal sensory analysis was performed in a standard tasting room (ISO 8589-1998) at room temperature (20 ℃). Wine samples were served in random order in covered tasting glasses (ISO 3591-1997). Each of the fourteen panelists was asked to score the intensity of each descriptor on a 0-4 scale: (0) imperceptible; (1) exist but hardly recognized; (2) recognizable, but weak; (3) recognizable, but not strong enough; (4) very strong. Modified frequency (MF) was introduced to evaluate the data (Equation (2))[25]:

$$MF = \sqrt{FI} \quad (2)$$

where, F is the perceived frequency of each descriptor, %; I is the average intensity of each descriptor, %. To obtain reliable results, F (%)<20% was regarded as an invalid value.

2.6 Statistical analysis

All statistical analyses were performed by R packages (R Foundation for Statistical Computing, Vienna, Austria) and Origin 2018 software (OriginLab Corporation, Northampton, MA, USA). One-way ANOVA and Duncan test were applied to determine the variance of physicochemical indices. A non-metric multidimensional scaling (NMDS) plot and a partial least squares discrimination analysis (PLS-DA) plot were created to illustrate the separations of different treatments. The analysis of similarities (ANOSIM) and the permutational multivariate analysis of variance (PERMANOVA) were used to test whether the divergence among different treatments was greater than that among every three duplicates and whether the discrimination was significant or not. A heatmap based on normalized data was drawn to visualize the clustering and changes of VOCs. Kruskal-Wallis H test was employed to reveal the effect of MOX on each VOC. Mantel test and correlation analysis were performed to investigate the relationship between VOCs and aroma descriptors.

3 Results

3.1 Effects of MOX on oenological parameters

Conventional oenological parameters are important judgments for evaluating the basic properties of wine. As shown in Table 2, there was no significant difference in alcohol content, dry matter, reducing sugar, titratable acidity, volatile acidity, or pH between the MOX-treated wines and the control group (NM) after six months of aging.

Polyphenols determine the color, mouthfeel, and antioxidant capacity of wine. It was found that the contents of total phenols, tannins, and free anthocyanins were lower in MOX-treated wines than in the control group (NM), and higher doses of oxygen (i.e., B20, B30, A5, and A8) caused significant decrements of them (Table 2). Different levels of MOX were able to significantly improve the color intensity of the wine, except for YN-B10 and YN-A1, in which no statistical difference was detected. The pre-MLF MOX treatments improved the hue values of the wine, whereas the post-MLF treatments came with the opposite results.

Table 2 Oenological parameters of three sub-region wines treated or untreated with MOX

Parameters	Sub-regions	NM	Before MLF			After MLF		
			B10	B20	B30	A1	A5	A8
Alcohol (%, v/v)	GCZ	15.20±0.10^a	15.30±0.10^a	15.30±0.00^a	15.30±0.10^a	15.20±0.00^a	15.20±0.10^a	15.20±0.10^a
	YC	15.10±0.10^a	15.10±0.20^a	15.10±0.10^a	15.40±0.10^a	15.10±0.20^a	15.20±0.10^a	15.30±0.00^a
	YN	14.30±0.00^a	14.40±0.10^a	14.30±0.20^a	14.40±0.10^a	14.30±0.10^a	14.30±0.10^a	14.30±0.10^a
Dry matter/g Ł$^{-1}$	GCZ	33.30±0.20^a	33.30±0.10^a	33.20±0.10^a	33.10±0.20^a	33.20±0.30^a	33.00±0.20^a	33.20±0.20^a
	YC	30.60±0.00^a	30.50±0.20^a	30.70±0.30^a	30.60±0.10^a	30.50±0.10^a	30.50±0.30^a	30.70±0.00^a
	YN	29.90±0.20^a	29.80±0.10^a	29.80±0.10^a	29.60±0.10^a	29.70±0.10^a	29.70±0.30^a	29.70±0.20^a
Reducing sugars/g Ł$^{-1}$	GCZ	0.72±0.01^a	0.72±0.01^a	0.72±0.00^a	0.72±0.01^a	0.71±0.01^a	0.71±0.01^a	0.72±0.01^a
	YC	0.69±0.01^a	0.69±0.01^a	0.70±0.01^a	0.69±0.00^a	0.69±0.01^a	0.69±0.01^a	0.70±0.01^a
	YN	0.65±0.01^a	0.64±0.00^a	0.63±0.02^a	0.63±0.01^a	0.64±0.01^a	0.64±0.00^a	0.64±0.01^a
Titratable acidity/g Ł$^{-1}$	GCZ	4.80±0.10^a	4.90±0.00^a	4.80±0.00^a	4.90±0.00^a	4.80±0.10^a	4.80±0.10^a	4.80±0.10^a
	YC	5.90±0.10^a	5.80±0.20^a	5.90±0.00^a	5.90±0.10^a	5.80±0.10^a	5.90±0.10^a	6.00±0.10^a
	YN	5.10±0.00^a	5.10±0.10^a	5.10±0.00^a	5.10±0.10^a	5.10±0.10^a	5.10±0.10^a	5.20±0.10^a
Volatile acidity/g Ł$^{-1}$	GCZ	0.41±0.03^a	0.40±0.01^a	0.41±0.00^a	0.42±0.02^a	0.39±0.02^a	0.41±0.02^a	0.41±0.02^a
	YC	0.42±0.03^a	0.43±0.00^a	0.42±0.02^a	0.43±0.04^a	0.43±0.01^a	0.43±0.05^a	0.44±0.01^a
	YN	0.50±0.01^a	0.50±0.02^a	0.50±0.03^a	0.51±0.01^a	0.51±0.02^a	0.50±0.03^a	0.51±0.02^a
pH	GCZ	3.98±0.00^a	3.98±0.00^a	3.99±0.00^a	3.98±0.00^a	3.98±0.00^a	3.98±0.00^a	3.99±0.00^a
	YC	3.83±0.00^a	3.83±0.01^a	3.83±0.00^a	3.84±0.01^a	3.84±0.01^a	3.83±0.00^a	3.83±0.00^a
	YN	3.89±0.00^a	3.88±0.00^a	3.89±0.00^a	3.89±0.00^a	3.89±0.00^a	3.89±0.00^a	3.89±0.00^a

Parameters	Sub-regions	NM	Before MLF			After MLF		
			B10	B20	B30	A1	A5	A8
Total phenols/g Ł$^{-1}$	GCZ	2.07±0.01^{a}	2.06±0.02^{ab}	2.04±0.01^{ab}	2.03±0.02^{b}	2.06±0.02^{a}	2.05±0.02^{ab}	2.04±0.01^{ab}
	YC	1.86±0.02^{a}	1.82±0.04^{abc}	1.80±0.02^{bc}	1.80±0.01^{c}	1.85±0.03^{a}	1.84±0.02^{ab}	1.85±0.02^{a}
	YN	1.70±0.04^{a}	1.66±0.02^{ab}	1.65±0.01^{b}	1.64±0.01^{b}	1.66±0.02^{ab}	1.65±0.05^{ab}	1.66±0.02^{ab}
Total tannins/g Ł$^{-1}$	GCZ	2.64±0.01^{a}	2.56±0.06^{a}	2.54±0.05^{a}	2.53±0.09^{a}	2.57±0.04^{a}	2.53±0.05^{a}	2.53±0.15^{a}
	YC	2.13±0.00^{a}	2.07±0.08^{a}	2.08±0.06^{a}	2.04±0.06^{a}	2.12±0.01^{a}	2.09±0.02^{a}	2.11±0.03^{a}
	YN	1.96±0.02^{a}	1.95±0.00^{a}	1.92±0.03^{a}	1.81±0.01^{b}	1.93±0.02^{a}	1.94±0.05^{a}	1.92±0.03^{a}
Free anthocyanins/mg Ł$^{-1}$	GCZ	484.00±10.00^{a}	477.00±11.00^{a}	474.00±6.00^{ab}	463.00±5.00^{b}	481.00±6.00^{a}	473.00±9.00^{ab}	474.00±5.00^{ab}
	YC	370.00±22.00^{a}	366.00±7.00^{a}	348.00±2.00^{b}	343.00±5.00^{b}	358.00±4.00^{ab}	347.00±2.00^{b}	348.00±3.00^{b}
	YN	368.00±19.00^{a}	355.00±12.00^{ab}	351.00±4.00^{ab}	340.00±2.00^{b}	367.00±4.00^{a}	357.00±6.00^{a}	356.00±4.00^{ab}
Color intensity	GCZ	8.90±0.02^{d}	9.27±0.05^{c}	9.33±0.03^{b}	9.99±0.02^{a}	9.35±0.06^{b}	9.33±0.02^{bc}	10.04±0.02^{a}
	YC	6.84±0.02^{f}	7.01±0.02^{d}	7.17±0.03^{c}	8.01±0.03^{b}	6.92±0.01^{e}	7.21±0.03^{c}	8.14±0.02^{a}
	YN	6.90±0.04^{d}	6.88±0.01^{d}	7.29±0.02^{b}	7.69±0.08^{a}	6.94±0.01^{d}	7.23±0.02^{c}	7.67±0.02^{a}
Hue	GCZ	0.85±0.00^{d}	0.90±0.00^{b}	0.86±0.00^{c}	0.90±0.00^{a}	0.80±0.00^{f}	0.80±0.00^{e}	0.75±0.00^{g}
	YC	0.82±0.00^{c}	0.86±0.00^{b}	0.86±0.00^{b}	0.90±0.01^{a}	0.82±0.01^{c}	0.83±0.01^{c}	0.78±0.00^{d}
	YN	0.81±0.00^{c}	0.82±0.00^{ab}	0.83±0.01^{a}	0.81±0.01^{bc}	0.78±0.00^{d}	0.77±0.01^{e}	0.79±0.00^{d}

Note: GCZ: Ganchengzi sub-region; YC: Yinchuan sub-region; YN: Yongning sub-region; NM: the control group without MOX treatment. B10, B20, and B30 are the oxygen doses of 10, 20, and 30 (mL/L)/month before MLF. A1, A5, and A8 are the oxygen doses of 1, 5, and 8 (mL/L)/month after MLF. Values are means±standard deviation of three independent experiments. Different letters within the same horizontal line indicate significant differences (Duncan's test, p<0.05). The same as below.

3.2 Effects of MOX on VOCs

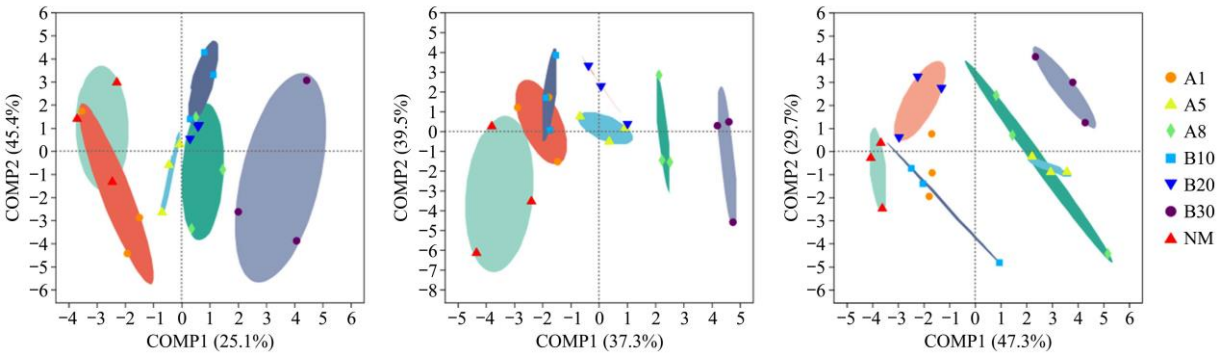
Given the large number of VOCs identified, the results were analyzed by grouping some of them with a similar chemical structure and a comparable evolutionary pattern, such as fatty alcohols, ethyl esters, isoamyl esters, and acetic esters. The rest of the VOCs were studied one by one, as stated in Table S2.

An unconstrained ordination approach (NMDS) and a supervised analysis (PLS-DA) were introduced to assess the differences in VOCs between the MOX-treated wines and the control wines (Figure S3, Table 3, and Figure 1). The NMDS stress values of three sub-region data varied from 0.050 to 0.061, and the first two principal components of PLS-DA score plots explained more than 70% of the variance, indicating that the discrimination was efficient. The dissimilarity was further investigated using the ANOSIM and PERMANOVA analysis based on Bray-Curtis distance matrices (after 9999 permutations) (Table 3). It was clear that the variation among different treatments was significantly greater than that among every three duplicates (R>0, R^2>0, p<0.05). The above results preliminarily proved that the MOX treatment did induce noticeable alterations in VOCs.

Table 3 Variances in VOCs among different treatments

Sub-regions	NMDS stress value	Statistical test	R or R^2	p-value
GCZ	0.050	Bray-Curtis ANOSIM	0.331	0.001**
		Bray-Curtis PERMANOVA	0.556	0.011*
YC	0.061	Bray-Curtis ANOSIM	0.282	0.006**
		Bray-Curtis PERMANOVA	0.551	0.005**
YN	0.059	Bray-Curtis ANOSIM	0.493	0.001**
		Bray-Curtis PERMANOVA	0.662	0.001**

Note: The NMDS stress value below 0.1 indicates good discrimination among different treatments. R or R^2>0, the variation among different treatments was greater than that among every three duplicates. * p<0.05, ** p<0.01.



a. Ganchengzi (GCZ) sub-region b. Yinchuan (YC) sub-region c. Yongning (YN) sub-region

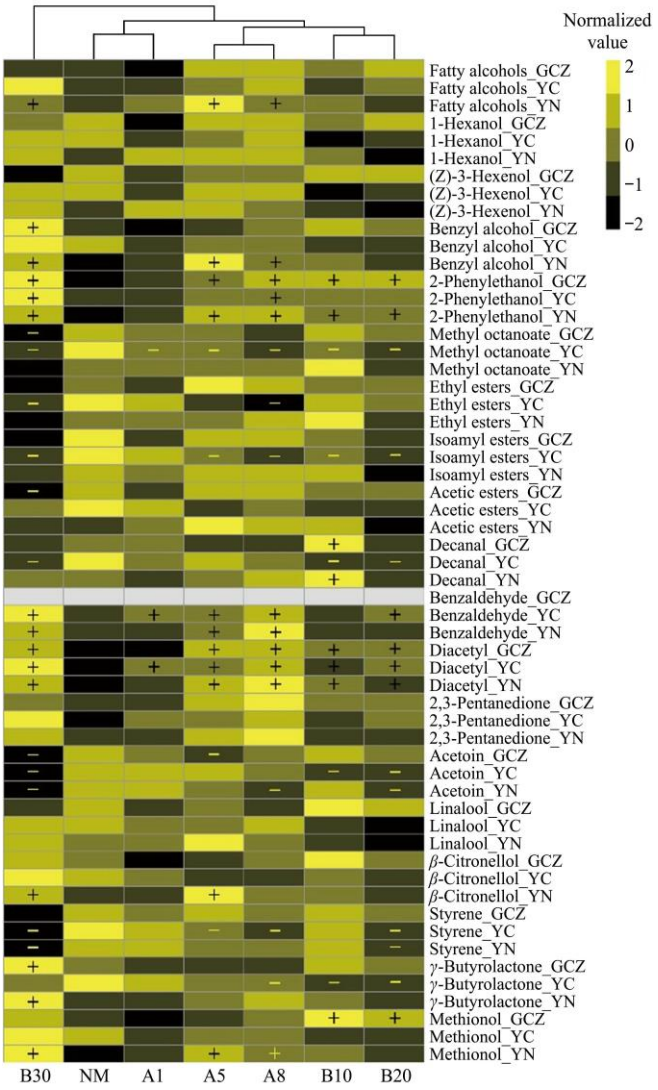
Note: NM: Control group without MOX treatment. B10, B20, and B30 are the oxygen doses of 10, 20, and 30 (mL/L)/month before MLF. A1, A5, and A8 are the oxygen doses of 1, 5, and 8 (mL/L)/month after MLF. The overall variance is explained by the first two principal components (COMP1 and COMP2). MLF: Malolactic fermentation; MOX: Micro-oxygenation. The same as below.

Figure 1 PLS-DA score plots based on VOCs distinguishing among different treatments

The data of VOCs were thereinafter normalized and visualized by a clustering heatmap (Figure 2). A1 and NM were assigned to the same group, which was segregated from other treatments, suggesting that the influence of the lowest level of MOX on VOCs was limited. The clustering distance between B30 and NM, on the other hand, was remarkable, signaling that a higher amount of oxygen added before MLF was hoped to generate the most obvious changes in VOCs.

Higher alcohols are one of the main secondary metabolites of yeast during alcohol fermentation, where their concentrations below 300 mg/L impart a desirable sense of richness to grape wine, whereas concentrations greater than this level would produce some unpleasant odors[2]. In light of Figure 2, no significant difference in fatty alcohols was found between the control and the micro-oxygenated GCZ or YC wines. Although higher-level MOX treatments (B30, A5, and A8) caused a discernible increment

of fatty alcohols in YN wines, the concentrations remained less than 300 mg/L and would not impair the aroma quality (Table S2). C6 alcohols [1-hexanol and (Z)-3-hexenol] are released by the oxidation of linoleic and linolenic acids when grape berries are crushed, giving herbaceous/green notes to wine[26]. There was no significant change in the content of 1-hexanol or (Z)-3-hexenol after MOX treatment. 2-Phenylethanol (2-PE) is the most common aromatic alcohol in fermented wines, having a rose- and honey-like flavor[27]. From Figure 2, different levels of MOX applied either before or after MLF raised 2-PE concentrations in all the wine samples, with higher dosages resulting in significant increments, especially by the B30 treatment.



Note: A '+' or a '–' indicates a significant increase or a significant decrease in the VOC content, compared with the NM group (Duncan's test, $p<0.05$). The clustering of columns is based on the Euclidean distance and on the complete linkage method.

Figure 2 Difference and clustering of VOCs among different treatments

Esters are responsible for the fruity and floral scents of grape wine. Yeasts yield several esters, including methyl esters, ethyl esters, acetic esters, and isoamyl esters, via enzymatic reactions of alcohols with acyl CoA[28]. In this experiment (Figure 2), the contents of methyl, ethyl, and isoamyl esters in YC wine fell to varying degrees after MOX treatment. Only the B30 group of GCZ wine was characterized by significantly lower quantities of methyl and acetic esters than the NM group. However, MOX had no effect on the esters in YN wine.

Aldehydes are generated as a result of alcohol oxidation and the Strecker degradation of α-amino acids[29]. According to Table S2, the benzaldehyde contents of YC wines grew by 19.9%-228% after MOX treatment, while climbing by 11.5%-162% in YN wines. A higher level of benzaldehyde was produced when there was more oxygen present before or after MLF. Decanal concentration, on the other hand, remained unaltered or declined in most of the samples.

Diacetyl and acetoin are recognized as generically pleasant nutty, caramel, and buttery smells below 1 mg/L, and can be transformed mutually in wine matrix through enzymatic or non-enzymatic processes[30]. In this investigation, diacetyl content increased by 1.1%-65.2% in GCZ wine, 58.6%-206% in YC wine, and 15.6%-95.1% in YN wine, whilst acetoin decreased to various levels (Table S2). 2,3-Pentanedione shares a comparable chemical structure and odor description with diacetyl as well as a similar change pattern after MOX treatment.

Terpenes (linalool and citronellol) exist in glycoside-conjugated forms in grape berries and can be released by hydrolysis events throughout the fermentation and aging stages, endowing floral and citrus aromas to wine[31]. Figure 2 showed that different MOX treatments before or after MLF had no significant effect on the concentrations of linalool and citronellol in GCZ and YC wine samples, but B30 and A5 treatments significantly improved citronellol contents in YN wines.

3.3 Effect of physicochemical indices on MOX

Considering that the evolutionary patterns of most VOCs varied across three sub-regions, the physicochemical features of the grape and the original wine (before being treated with MOX) were examined (Table 4). Viticulturists have several metrics that can be used to determine grape maturity, including total sugar, acidity, sugar-to-acid ratio, pH, and sugar ×pH[32,33]. Table 4 revealed that the grape material from the GCZ sub-region owned the highest ripeness, followed by YC, and the lowest was YN. With regard to the initial wine indicators, the ethanol content, dry matter, total phenols, tannins, anthocyanins, and the anthocyanin-to-tannin ratio of the GCZ wine were significantly higher than those of the YC and YN wines. As a result, GCZ grape/wine was rated as having the greatest quality, while YN grape/wine was ranked as having the lowest quality.

Table 4 Physicochemical indices of the grape and the original wine

Sample type	Physicochemical indices	GCZ	YC	YN
Grape	Total sugar/g Ł$^{-1}$	252.70±0.90^a	240.10±0.90^b	226.10±0.90^c
	Total acid/g Ł$^{-1}$	5.70±0.10^c	6.50±0.10^a	6.10±0.10^b
	Total sugar/total acid	44.60±0.90^a	37.10±0.20^b	36.90±0.40^b
	Must pH	3.59±0.00^a	3.54±0.00^b	3.52±0.01^c
	Total sugar×pH	907.10±9.80^a	849.90±8.70^b	795.20±5.00^c
Original wine	Alcohol/(%, v/v)	15.40±0.10^a	14.80±0.10^b	14.10±0.00^c
	Dry matter/g Ł$^{-1}$	35.30±0.10^a	33.60±0.10^b	31.40±0.20^c
	Free SO_2/mg Ł$^{-1}$	10.10±0.40^a	11.30±0.70^a	10.30±0.40^a
	Residual sugar/g Ł$^{-1}$	1.73±0.02^a	1.77±0.02^a	1.67±0.03^b
	Titratable acid/g Ł$^{-1}$	5.50±0.10^c	6.30±0.10^a	6.00±0.10^b
	Wine pH	3.83±0.01^a	3.64±0.00^c	3.78±0.01^b
	Total phenols/g Ł$^{-1}$	2.72±0.06^a	2.51±0.02^b	2.47±0.03^b
	Tannins/g Ł$^{-1}$	2.58±0.09^a	2.19±0.07^b	2.11±0.06^b
	Anthocyanins/mg Ł$^{-1}$	617.00±6.00^a	454.00±6.00^b	418.00±4.00^c
	Anthocyanins/Tannins	0.24±0.01^a	0.21±0.01^b	0.20±0.00^b

Note: Values are means±standard deviation of three independent experiments. Different letters within the same horizontal line indicate significant differences (Duncan's test, $p<0.05$).

The Kruskal-Wallis H test was used to calculate VOC variances caused by MOX in each sub-region wine (Table 5). We noticed that there were more VOCs in YN wine that exhibited statistically significant changes after experimental treatments, indicating that the impact of MOX on VOCs was more noticeable for lower-quality wine.

Table 5 Variations in VOCs caused by MOX

Compounds	*p* value of the Kruskal-Wallis H test		
	GCZ	YC	YN
Fatty alcohols	0.123	0.079	**0.015***
1-Hexanol	0.575	0.381	0.119
(Z)-3-Hexenol	0.282	0.172	0.171
Benzyl alcohol	**0.022***	0.194	**0.023***
2-Phenylethanol	**0.019***	**0.025***	**0.018***
Methyl octanoate	0.528	**0.030***	0.095
Ethyl esters	0.660	0.183	0.062
Isoamyl esters	0.742	0.082	0.216
Acetic esters	0.519	0.809	0.109
Decanal	0.187	**0.033***	0.061
Benzaldehyde	n.d.	**0.004****	**0.005****
Diacetyl	**0.005****	**0.004****	**0.004****
2,3-Pentanedione	**0.004****	**0.004****	**0.004****
Acetoin	**0.038***	**0.023***	**0.018***
Linalool	0.162	0.128	0.099
β-Citronellol	0.185	0.090	0.087
Styrene	0.466	**0.011***	**0.028***
γ-Butyrolactone	0.057	0.182	0.070
Methionol	**0.019***	0.289	**0.025***
Total VOCs	0.150	0.663	**0.033***

Note: * $p<0.05$, ** $p<0.01$, n.d.: not detected.

3.4 Effects of MOX on aroma profiles

Ten aroma descriptors were scored using the QDA method (Figure 3). Green, fresh fruit, dried fruit, floral, and spicy MF values were higher than other descriptors, suggesting that they are the typic olfactory characteristics of Cabernet Sauvignon dry red wines in Ningxia. Compared to the control group (NM), the green intensities of all the MOX-treated wines were downregulated. MOX also reduced the fresh fruit flavor of YC wine due to a decrease in ester content (Figure 2), but the GCZ or YN wine did not show the same changing pattern as the YC wine. The dried fruit intensities of all the micro-oxidized wines were enhanced, with the largest increments observed in the A8 treatments. The MF values of floral aroma increased to varying degrees, with the most apparent improvements registered in the B30 and the A8 treatments. Previously, Hernández-Orte et al.[15] demonstrated

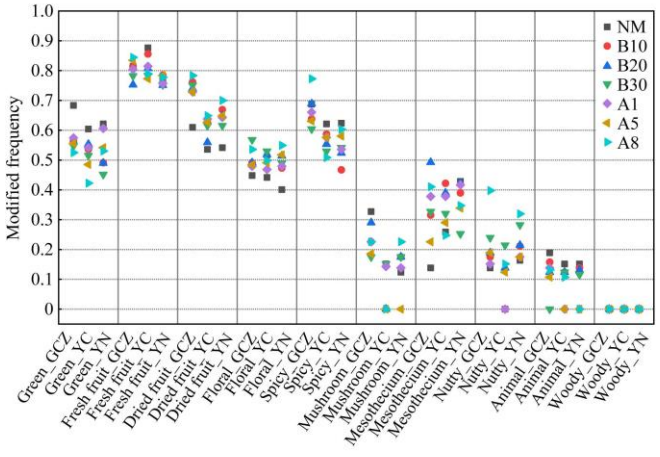


Figure 3 Modified frequencies (MFs) of aroma descriptors

that MOX either improved or decreased the spicy flavor of Cabernet Sauvignon dry red wine. Our study reached a consistent conclusion with them (Figure 3).

Although the MF values for the other five descriptors (mushroom, mesothecium, nutty, animal, and woody) were lower, they contributed to the complexity of the wine aroma. Figure 3 illustrated that the odor of mushroom, or mesothecium, did not shift in a consistent way among the three sub-region wines. The nutty flavor of MOX-treated wines was enhanced (particularly in the B30 and A8 treatments), and the animal scent was reduced. Panelists did not detect the woody flavor since oak products were not added to the wine samples throughout the trial.

3.5 Correlation analysis between VOCs and aroma descriptors

The Mantel test was introduced to assess the connection between the two data sets, VOC concentrations, and the MF values of aroma descriptors[34]. The VOC data for YC wine was significantly connected with the sensory data, regardless of whether the computation strategy used was Euclidean distance or Manhattan distance (Table 6). The Manhattan algorithm-based Mantel test revealed that the VOC data for YN wine was significantly linked with the sensory data. The correlation in GCZ wine, on the other hand, was not statistically significant. Therefore, additional investigation into the one-to-one correlation between VOC and aroma descriptor was necessary.

Table 6 Relationship between VOCs and aroma descriptors

Sub-regions	Statistical test	Mantel coefficient	*p*-value
GCZ	Euclidean Mantel test	0.164	0.270
	Manhattan Mantel test	0.188	0.264
YC	Euclidean Mantel test	0.579	**0.012***
	Manhattan Mantel test	0.631	**0.013***
YN	Euclidean Mantel test	0.363	0.090
	Manhattan Mantel test	0.487	**0.034***

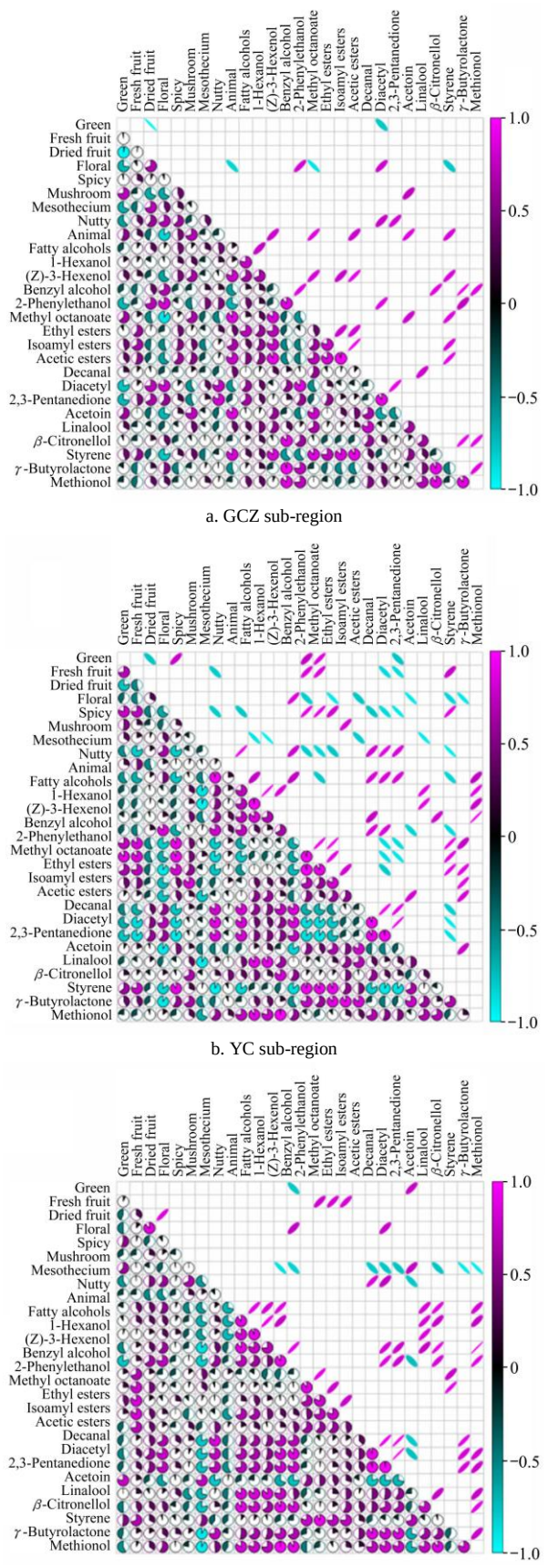
Note: * $p<0.05$.

Figure 4 depicts a graphical representation of a correlation matrix. Despite the notion that C6 alcohols can impart green smells to wine, the change in green MF value was unrelated to 1-hexanol or (Z)-3-hexenol. Esters contributed to the fruity flavor of wine, as proved in Figure 4. Although MOX improved dried fruit intensity (Figure 3), this flavor was not closely related to any VOC. According to Figure 4, the enhancement of floral flavor was significantly and positively correlated with the increase of 2-PE in three sub-region wines. The improvement of nutty fragrance also had a significant and positive association with diacetyl and benzaldehyde.

4 Discussion

Given that the aroma complexity and typicality of Cabernet Sauvignon red wine in Ningxia deteriorate rapidly during the aging process, this study conducted different levels of MOX treatments prior to wine aging, intending to examine the practicability of this technique.

It was found that the application of MOX did not affect basic oenological parameters of wine, but reduced the concentrations of total phenols, tannins, and free anthocyanins. The reason may be that MOX has promoted the reactions of phenolic molecules[35,36]. For example, oxygen stimulates the synthesis of acetaldehyde, which is involved in the polymerization of monomeric anthocyanins with tannins or organic acids[37-39]. Polymeric anthocyanins can modify wine color because they are resistant to



a. GCZ sub-region

b. YC sub-region

c. YN sub-region

Note: The pie graphs in the lower-left matrix represent the positive (magenta) and negative (cyan) correlation coefficients. The ellipses in the upper-right matrix represent significant correlations (Pearson test, $p<0.05$), specifically a magenta ellipse shows a significantly positive correlation, while the cyan ellipse shows a significantly negative correlation.

Figure 4 Correlation matrices between VOCs and aroma descriptors

discoloration by SO_2 and are responsible for better color stability at wine pH[4,5]. In addition, MOX was helpful to improve the color intensity of the wine in this study, which was consistent with previous works[9,40]. Likewise, combined tannins and other polymeric phenolic compositions are usually related to reduced astringency, less bitterness, or enhanced fullness of red grape wines[41,42].

VOCs were analyzed after six months of aging. It is necessary to highlight that MOX has the potential to increase the concentrations of 2-phenylethanol, benzaldehyde, diacetyl, and 2,3-pentanedione in all the wine samples. Although the MOX-induced increment of 2-phenylethanol was also proposed in another study[43], it seems strange because alcohols tend to be oxidized in the presence of oxygen. A possible explanation could be that there were some residual viable yeasts presented in the unfiltered wines, and the addition of oxygen stimulated their growth, resulting in a further generation of 2-phenylethanol and other volatile compounds[10,44]. Benzaldehyde is produced via the enzymatic activities of yeasts during AF or formed by the non-enzymatic oxidation of phenylalanine[45], and it was proved to be either upregulated[46] or downregulated[16] under micro-oxygenated conditions. The vicinal diketones diacetyl (2,3-butanedione) and 2,3-pentanedione arise from microbial metabolism of yeasts and bacteria, or chemical non-enzymatic oxidative decarboxylation of α-acetolactate and α-acetohydroxybutyrate[47]. Lasik-Kurdyś[48] and Moreira et al.[49] described an increased pattern of diacetyl caused by MOX effects during MLF and bottle aging, but an earlier study came to the opposite results[15]. Although the change of 2,3-pentanedione was rarely stated in MOX traits of dry red wines, it continued to accumulate during oxidative aging of Pedro Ximenez sweet wines[50].

Other VOCs behaved differently in micro-oxygenated wines across three sub-regions, thus we could not conclude the effects of MOX on them in Cabernet Sauvignon wines of Ningxia. In a Spanish study, the MOX treatment produced a decrease in some esters, alcohols, and benzenic compounds and increased a few terpenes and C13-norisoprenoids of Merlot wines[13]. Regarding Tempranillo, Cabernet Sauvignon, and Tinta del Pa ś wines in Spain, however, the concentrations of higher alcohols tended to be raised after micro-oxidation treatment[15,43], which was consistent with our results in the YN sub-region. Based on two-dimensional gas chromatography, Schmarr et al.[51] demonstrated that a set of volatiles, such as 2-phenylethanol, methionol, decanal, and some esters, could be considered chemical markers for the MOX-treated German Cabernet Sauvignon wines. Additionally, MOX could promote the extraction of guaiacol and its derivatives from wood staves in a Portuguese wine spirit[11], but induced a lower amount of 4-ethyl guaiacol in an Italian Sangiovese red wine[52]. Aroma compounds in white wine can also be influenced by MOX treatment, with elevated concentrations of higher alcohols, esters, and fatty acids in Pinot Blanc and Pinot Gris wines[53].

Although one of the objectives of MOX is to remove the green smells from wine, no research established that the decline of green notes is related to the decrease of C6 alcohols in MOX-treated wines[11,13,16,54]. Our results indicated that the green odor was downregulated by MOX treatment, but there was no significant change in the content of 1-hexanol or (Z)-3-hexenol (Figure 2). The correlation matrices (Figure 4) also showed a weak concordance between the green odor and C6 alcohols. Therefore, the cutback in green odor should be related to other components.

A partial least squares regression (PLSR) model was introduced to investigate the compounds potentially correlated with the decrease of green smells in MOX-treated wines (Table S3). The results revealed that isoamyl esters in all the sub-region wines were positively related to the green odor with a VIP value higher than 1, indicating a considerable contribution to the green characteristic of wine. However, this has not yet been proven experimentally.

Some studies suggested that the micro-oxidizing environment was not conducive to the formation of esters, presumably due to the direct attack by hydroxyl radicals or by ester interaction with *o*-quinones[45]. Others believed MOX treatment would not alter ester concentrations[15,54]. The data of this study lend support to the former point of view. Linalool and citronellol are terpene odorants found in grape wines that provide floral and citrus aromas. This study discovered that MOX did not affect linalool concentrations, but B30 and A5 treatments increased citronellol concentrations in YN wine. Some researchers hypothesized that oxygen could enhance the hydrolysis of citronellol precursors, but the process varied depending on the type of wine[15,54].

Wine is a complex matrix with numerous components. Due to the matrix effect of non-volatile organic compounds and the addition/masking phenomenon among different VOCs, it is not reasonable to infer the evolution of wine aroma profiles only through the change of VOCs[55,56]. Sensory analysis is the most reliable criterion. The aroma properties of wine were evaluated through a panel of trained judges using quantitative descriptive analysis[25]. The result showed that the green and animal scents were reduced, while the dried fruit, floral, and nutty flavors were enhanced, no matter whether the MOX treatments were performed at any stage or dosage. The subsequent correlation analysis illustrated that the intensification in flower aroma correlated with the increment of 2-phenylethanol, and the enrichment in nutty flavor was related to the accumulation of benzaldehyde and diacetyl. Other aroma profiles, such as fresh fruit and spicy scents, did not change consistently across the three sub-region wines. Therefore, it is not realistic to assume that oxidative reactions always reduce the fruitiness or pungency of grape wine[12,14,15].

Interestingly, the quality of the grape material and the initial wine determined the MOX outcomes. The influences of MOX treatments on VOCs were obvious in wine samples with poor grape maturity, lower phenolic content, and lower anthocyanin-to-tannin ratio. Future studies will be required to assess the matrix effect of wine on MOX efficiency.

5 Conclusions

This study proved that micro-oxygenation can increase the aroma quality of Cabernet Sauvignon dry red wine in Ningxia, China, with the modified wine characterized by diminished green and animal off-smells and enhanced pleasant flavors of flowers, nuts, and dried fruits. 2-Phenylethanol, benzaldehyde, diacetyl, and 2,3-pentanedione are typical volatile indicators for identifying micro-oxygenated wines. An oxygen dose of 30 (mL/L)/month is recommended to be supplied to the wine prior to malolactic fermentation.

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Appendix

Table S1 VOCs measured in the HS-SPME-GC-MS method, their retention indices (RIs), calculated retention indices (CRIs), and selected ion monitoring (SIM) qualifying ions

Compound name		RI	CRI	SIM ions
Fatty alcohols	1-Propanol	1038	1041	59/42/60
	1-Butanol	1148	1150	56/41/43
	1-Pentanol	1256	1256	55/42/70
	1-Octanol	1559	1559	56/70/84
	1-Nonanol	1665	1666	56/69/70
	1-Decanol	1767	1769	83/70/55
	2-Heptanol	1321	1326	45/55/83
	2-Nonanol	1521	1522	45/69/98
	2-Methyl-1-propanol	1094	1096	43/42/74
	3-Methyl-1-butanol	1217	1218	70/55/42
	3-Methyl-1-pentanol	1343	1337	56/69/84
	4-Methyl-1-pentanol	1316	1319	56/69/41
C_6 alcohols	1-Hexanol	1359	1359	56/69/101
	(Z)-3-Hexenol	1386	1386	67/41/82
Aromatic alcohols	Benzyl alcohol	1866	1867	79/107/108
	2-Phenylethanol	1899	1899	91/92/122
Methyl esters	Methyl octanoate	1386	1388	74/87/127
Ethyl esters	Ethyl butyrate	1031	1034	71/88/116
	Ethyl valerate	1136	1134	88/85/101
	Ethyl hexanoate	1236	1236	88/99/144
	Ethyl heptanoate	1336	1335	88/101/158
	Ethyl octanoate	1440	1438	88/101/127
	Ethyl nonanoate	1535	1533	88/101/141
	Ethyl decanoate	1638	1641	88/101/200
	Ethyl undecanoate	1739	1741	88/101/169
	Ethyl dodecanoate	1850	1844	88/101/228
	Ethyl lactate	1340	1343	45/43/75
	Ethyl 2-phenylacetate	1780	1775	91/65/164
	Ethyl 2-methylbutyrate	1050	1051	102/57/130
	Ethyl 3-methylbutyrate	1067	1066	88/85/130
	Ethyl 9-decenoate	1688	1688	88/110/152
	Diethyl succinate	1675	1675	101/129/174
Isoamyl esters	Isoamyl butyrate	1259	1267	71/43/70
	Isoamyl hexanoate	1450	1456	70/99/117
	Isoamyl octanoate	1657	1659	70/127/145
	Isoamyl lactate	1570	1564	45/55/70
Acetic esters	3-Methylbutyl acetate	1121	1121	70/55/87
	Hexyl acetate	1275	1273	56/61/84
	2-Phenylethyl acetate	1801	1799	104/43/91
Aldehydes	Decanal	1494	1492	57/43/70
	Benzaldehyde	1508	1507	106/105/77
Ketones	Diacetyl	968	968	43/86
	2,3-Pentanedione	1054	1057	43/57/100
	Acetoin	1278	1278	45/43/88
Terpenes	Linalool	1548	1548	93/71/136
	β-Citronellol	1767	1772	69/81/156
Miscellaneous	Styrene	1250	1251	104/78/103
	γ-Butyrolactone	1601	1603	42/86/41
	Methionol	1710	1709	106/61/58

Note: The RIs on DB-Wax column were obtained from the NIST Chemistry WebBook (https://webbook.nist.gov/). The CRIs were calculated according to the retention times of C8-C20 n-alkanes and each VOC.

Table S2 Concentrations of VOCs in three sub-region wines treated or untreated with MOX

VOCs	Sub-regions	Concentrations/μg L^{-1}						
		NM	B10	B20	B30	A1	A5	A8
Fatty alcohols	GCZ	31415.5±828.1^{ab}	32937.0±744.7^{a}	34141.0±537.4^{a}	30895.6±1432.2^{ab}	29533.4±3820.6^{b}	33217.0±1583.6^{a}	34039.8±2172.3^{a}
	YC	26859.2±1844.4^{bc}	26167.1±348.2^{c}	27687.0±1141.4^{abc}	29236.7±927.9^{ab}	26148.8±1148.4^{c}	27760.1±695.4^{abc}	28865.8±1470.3^{ab}
	YN	24343.4±333.0^{d}	25774.3±777.9^{bcd}	24718.5±598.6^{cd}	26826.5±1140.8^{b}	26294.0±446.9^{bcd}	28820.3±1500.3^{a}	26607.9±1680.7^{bc}
1-Hexanol	GCZ	1291.7±62.4^{a}	1268.4±49.0^{a}	1287.0±12.4^{a}	1251.4±101.7^{a}	1184.5±167.5^{a}	1280.9±42.4^{a}	1282.6±90.1^{a}
	YC	1050.1±84.8^{a}	979.0±37.7^{a}	997.6±30.9^{a}	1062.5±62.2^{a}	998.1±54.8^{a}	1035.2±16.0^{a}	1058.1±55.9^{a}
	YN	921.8±25.4^{ab}	949.9±47.5^{ab}	901.4±14.9^{b}	982.4±46.3^{ab}	972.7±26.1^{ab}	991.4±67.0^{a}	974.0±73.7^{ab}
(Z)-3-Hexenol	GCZ	24.2±1.1^{a}	23.7±0.8^{a}	23.8±0.3^{a}	20.9±1.3^{a}	21.9±2.7^{a}	23.5±1.1^{a}	23.5±1.6^{a}
	YC	21.1±1.6^{ab}	19.2±0.5^{b}	19.9±0.8^{ab}	21.2±1.0^{a}	20.0±1.1^{ab}	21.0±0.5^{ab}	21.6±1.1^{a}
	YN	24.7±0.3^{a}	24.7±1.5^{a}	24.2±0.5^{a}	26.7±0.7^{a}	26.7±0.3^{a}	26.9±2.6^{a}	26.0±2.3^{a}
Benzyl alcohol	GCZ	198.3±19.0^{bc}	230.9±9.0^{ab}	223.0±1.5^{ab}	251.0±43.0^{a}	182.5±9.9^{c}	195.5±14.5^{bc}	210.5±15.0^{bc}
	YC	184.1±20.4^{ab}	165.9±15.6^{b}	165.2±14.8^{b}	206.4±24.1^{a}	162.1±10.7^{b}	177.4±15.5^{ab}	181.0±20.8^{ab}
	YN	177.0±9.0^{c}	206.6±34.3^{bc}	199.7±11.9^{bc}	252.1±13.9^{a}	202.1±10.0^{bc}	253.6±21.8^{a}	224.2±26.8^{ab}
2-Phenylethanol	GCZ	32838.1±2440.4^{d}	45102.3±4310.2^{ab}	45486.4±393.9^{ab}	50794.2±8415.4^{a}	34279.5±2396.5^{cd}	40933.3±2909.1^{bc}	44840.1±2839.4^{ab}
	YC	34901.2±3347.9^{c}	40594.5±3179.9^{bc}	41073.7±2847.7^{bc}	49441.2±5674.4^{a}	34969.3±1991.7^{bc}	39044.6±2264.5^{bc}	41327.0±4154.5^{b}
	YN	26630.0±1054.6^{d}	36349.5±5094.7^{abc}	34060.7±835.9^{bc}	40242.6±2208.1^{a}	31702.2±1013.3^{cd}	38955.6±2958.3^{ab}	37930.5±4654.6^{ab}
Methyl octanoate	GCZ	217.0±19.9^{a}	213.1±11.0^{ab}	204.0±2.6^{ab}	177.1±30.0^{b}	197.1±33.0^{ab}	204.3±3.5^{ab}	192.3±17.0^{ab}
	YC	198.3±20.6^{a}	166.5±13.5^{b}	147.5±2.1^{b}	143.9±21.1^{b}	165.4±12.4^{b}	154.3±8.5^{b}	140.8±9.3^{b}
	YN	240.9±19.6^{ab}	268.3±15.2^{a}	222.4±13.2^{b}	211.2±10.8^{b}	231.8±13.5^{b}	228.2±4.0^{b}	239.2±29.4^{ab}
Ethyl esters	GCZ	50405.4±3505.8^{a}	50110.9±2490.1^{a}	50328.5±571.7^{a}	47513.0±8005.2^{a}	48460.3±7350.8^{a}	53017.2±1263.6^{a}	52099.5±4517.4^{a}
	YC	47712.6±4732.8^{a}	44676.3±3361.7^{ab}	42513.8±385.7^{ab}	41250.4±4343.6^{b}	44937.3±2971.1^{ab}	41867.8±748.9^{ab}	39511.0±3518.8^{b}
	YN	48045.6±2920.2^{abc}	53396.3±3181.1^{a}	46070.3±2764.2^{bc}	43968.7±2258.3^{c}	49324.5±2507.1^{abc}	49175.8±1350.4^{abc}	51341.3±5972.2^{ab}
Isoamyl esters	GCZ	1355.2±113.1^{a}	1279.5±69.4^{a}	1266.2±10.4^{a}	1217.6±218.7^{a}	1254.1±190.4^{a}	1328.3±40.4^{a}	1329.4±119.9^{a}
	YC	1361.7±154.4^{a}	1148.4±101.0^{b}	1067.8±10.5^{b}	1079.0±120.4^{b}	1235.7±92.1^{ab}	1124.2±34.8^{b}	1076.6±99.3^{b}
	YN	1351.3±87.9^{ab}	1373.7±108.0^{a}	1203.5±56.4^{b}	1260.0±73.4^{ab}	1323.0±67.3^{ab}	1349.4±18.5^{ab}	1383.2±167.7^{a}
Acetic esters	GCZ	5675.4±290.7^{a}	5374.2±176.0^{ab}	5373.2±20.1^{ab}	4865.3±499.8^{b}	5197.8±746.4^{ab}	5541.3±205.4^{ab}	5585.4±364.0^{ab}
	YC	6382.1±509.4^{a}	5999.2±320.0^{a}	5908.5±151.4^{a}	6145.0±499.2^{a}	6207.1±335.0^{a}	5975.6±61.2^{a}	6074.0±366.0^{a}
	YN	5243.3±191.8^{abc}	5571.5±273.4^{ab}	5025.0±163.7^{c}	5172.8±238.1^{bc}	5453.9±191.6^{abc}	5737.3±266.3^{a}	5496.0±501.8^{abc}
Decanal	GCZ	49.4±11.9^{b}	63.5±3.3^{a}	43.1±1.3b	42.8±3.6^{b}	44.8±5.5^{b}	42.9±3.3^{b}	42.0±7.5^{b}
	YC	44.7±6.4^{a}	31.8±4.6^{c}	32.6±2.6^{c}	34.1±0.4^{c}	37.6±3.4a^{bc}	41.3±4.9^{ab}	37.9±2.7^{abc}
	YN	54.9±2.1^{bc}	66.4±10.0^{a}	51.0±4.8^{c}	58.2±3.1^{abc}	51.1±3.2^{bc}	56.4±3.4^{abc}	62.5±9.0^{ab}
Benzaldehyde	GCZ	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	YC	39.7±3.3^{e}	47.6±3.4^{de}	57.3±3.2^{cd}	130.2±14.1^{a}	60.0±3.3^{c}	66.7±1.6^{c}	90.4±7.8^{b}
	YN	49.5±0.7^{d}	55.2±1.7^{d}	58.9±5.4^{d}	111.2±3.7^{b}	56.1±0.9^{d}	88.5±5.5^{c}	129.7±16.3^{a}
Diacetyl	GCZ	130.0±2.6^{d}	175.1±2.1^{c}	179.3±4.3^{c}	204.3±15.1^{ab}	131.4±14.5^{d}	189.8±5.1^{bc}	214.8±16.5^{a}
	YC	69.6±7.1^{f}	110.4±11.1^{e}	133.5±7.4^{d}	212.8±8.3^{a}	119.4±10.5d^{e}	152.8±6.1^{c}	169.9±4.7^{b}
	YN	79.9±1.4^{e}	109.3±5.8^{c}	100.6±2.6^{cd}	137.4±7.3^{b}	92.4±3.8d^{e}	137.8±8.0^{b}	155.9±14.2^{a}
2,3-Pentanedione	GCZ	21.0±0.5^{e}	28.3±0.5^{cd}	26.6±1.0^{d}	29.9±0.9^{c}	21.6±2.4^{e}	34.3±2.6^{b}	39.0±2.5^{a}
	YC	10.8±1.0^{f}	16.9±0.3^{e}	22.4±0.9^{d}	38.0±0.7^{a}	22.8±1.4^{d}	26.8±0.8^{c}	35.0±1.9^{b}
	YN	9.6±0.2^{d}	12.6±0.3^{c}	11.8±0.3^{c}	19.6±1.0^{b}	11.6±0.2^{c}	20.2±1.1^{b}	24.0±1.2^{a}
Acetoin	GCZ	75.2±3.6^{a}	74.8±7.3^{a}	69.1±1.1^{ab}	59.2±8.2^{c}	69.7±6.3^{ab}	61.5±3.9^{bc}	65.9±4.7^{abc}
	YC	73.9±8.3^{a}	58.0±5.6^{bc}	57.6±2.0^{bc}	54.3±7.0^{c}	71.7±4.0^{a}	71.3±2.7^{a}	66.5±6.4^{ab}
	YN	45.1±0.7^{ab}	42.5±4.9^{ab}	37.4±1.0^{cd}	33.8±2.4^{d}	44.6±0.7^{a}	39.4±0.4^{bc}	36.3±4.1^{cd}
Linalool	GCZ	20.3±1.4^{ab}	21.7±1.1^{a}	20.4±0.7^{ab}	18.9±2.5^{b}	18.2±1.9^{b}	19.1±1.7^{ab}	18.6±1.2^{b}
	YC	20.4±1.5^{ab}	18.4±1.3^{ab}	18.0±0.6^{b}	20.0±2.0^{ab}	19.3±1.5^{ab}	19.8±0.4^{ab}	20.6±1.7^{a}
	YN	24.1±1.1^{abc}	23.5±2.5^{bc}	22.5±1.1^{c}	25.8±0.3^{ab}	24.4±0.5^{ab}c	26.5±0.9^{a}	24.3±2.3^{abc}
β-Citronellol	GCZ	21.4±1.8^{ab}	23.7±1.2^{a}	21.4±0.7^{ab}	23.1±3.8^{ab}	19.6±1.5^{b}	20.2±0.6^{ab}	21.1±2.0^{ab}
	YC	22.8±0.2^{ab}	21.7±2.0^{b}	20.4±0.8^{b}	25.2±3.3^{a}	21.1±1.9^{b}	20.3±0.8^{b}	21.0±0.9^{b}
	YN	19.0±0.2^{c}	21.3±2.7^{abc}	19.4±0.7^{c}	22.1±0.3^{ab}	20.0±0.7^{bc}	22.8±1.3^{a}	20.6±2.2^{abc}
Styrene	GCZ	341.6±22.5^{a}	335.5±10.8^{ab}	321.1±1.1^{ab}	296.4±37.6^{ab}	321.5±42.4^{ab}	340.6±17.9^{a}	328.4±15.2^{ab}
	YC	299.0±21.2^{a}	268.3±19.6^{ab}	235.2±2.2^{cd}	216.8±22.4^{d}	276.2±12.1^{ab}	253.7±3.6^{bc}	230.9±21.4^{d}
	YN	327.3±9.7^{a}	324.3±26.4^{a}	269.7±14.3^{bc}	237.6±11.0^{c}	312.3±14.2^{a}	306.4±3.6^{a}	301.6±34.5^{ab}
γ-Butyrolactone	GCZ	161.1±15.8^{bc}	186.8±25.2^{ab}	174.1±8.7^{abc}	198.1±36.3^{a}	144.3±4.8^{c}	156.9±7.4^{bc}	154.9±12.9^{bc}
	YC	225.1±52.5^{a}	161.1±21.3^{b}	154.4±10.7^{b}	174.7±21.5^{ab}	194.2±15.3^{ab}	184.0±13.1^{ab}	174.7±39.9^{b}
	YN	123.5±11.3^{bc}	125.1±14.0^{bc}	114.6±10.7^{c}	150.2±11.6^{a}	117.9±5.5^{c}	133.1±13.0^{abc}	143.8±14.0^{ab}
Methionol	GCZ	274.3±20.2^{cd}	343.6±32.1^{a}	321.2±2.2^{ab}	317.8±37.0a^{bc}	248.4±13.7^{d}	269.9±22.4^{d}	281.8±18.0^{bcd}
	YC	255.3±35.5^{ab}	231.8±17.5^{ab}	231.2±22.6^{ab}	275.0±29.8^{a}	225.5±17.0^{b}	252.1±17.7^{ab}	252.2±31.7^{ab}
	YN	195.2±11.9^{c}	224.1±30.0^{bc}	218.0±22.3^{bc}	282.3±27.9^{a}	223.4±7.1^{bc}	270.8±13.7^{a}	245.3±27.7^{ab}

Note: Values are means±standard deviation of three independent experiments. n.d., not detected. Different letters within the same horizontal line indicate significant differences (Duncan's test, p<0.05).

Table S3 Coefficients and Variable Importance (VIP) values of partial least squares regression (PLSR) between VOCs and green odor

VOCs	Green odor in GCZ wines		Green odor in YC wines		Green odor in YN wines	
	Coefficient	VIP value	Coefficient	VIP value	Coefficient	VIP value
Fatty alcohols	−0.063	0.822	−0.069	1.154	0.041	0.812
1-Hexanol	0.028	0.359	−0.039	0.654	0.102	0.907
(Z)-3-Hexenol	0.066	0.854	−0.050	0.846	0.211	0.124
Benzyl alcohol	−0.060	0.783	−0.019	0.314	−0.122	0.987
2-Phenylethanol	−0.127	1.649	−0.044	0.738	−0.305	1.406
Methyl octanoate	0.111	1.447	0.088	1.483	−0.114	0.843
Ethyl esters	−0.016	0.205	0.103	1.720	−0.020	0.695
Isoamyl esters	**0.079**	**1.034**	**0.079**	**1.324**	**0.210**	**1.060**
Acetic esters	0.069	0.897	0.052	0.870	0.100	0.892
Decanal	0.044	0.570	0.012	0.200	−0.264	1.023
Benzaldehyde	n.d.	n.d.	−0.062	1.048	0.013	0.903
Diacetyl	−0.129	1.674	−0.079	1.326	−0.096	0.930
2,3-Pentanedione	−0.116	1.506	−0.090	1.510	0.005	0.866
Acetoin	0.100	1.309	0.010	0.161	0.225	1.358
Linalool	0.057	0.747	−0.040	0.666	0.189	1.058
β-Citronellol	−0.012	0.150	0.026	0.442	−0.178	0.984
Styrene	0.076	0.992	0.074	1.239	0.113	1.009
γ-Butyrolactone	−0.029	0.375	0.048	0.804	−0.024	0.864
Methionol	−0.043	0.559	−0.026	0.434	−0.122	1.008

Note: VIP values larger than 1 indicate 'important' X-variables, and values lower than 0.5 indicate 'unimportant' X variables. The interval between 1 and 0.5 is a gray area, where the importance level depends on the size of the data set. n.d., not detected.

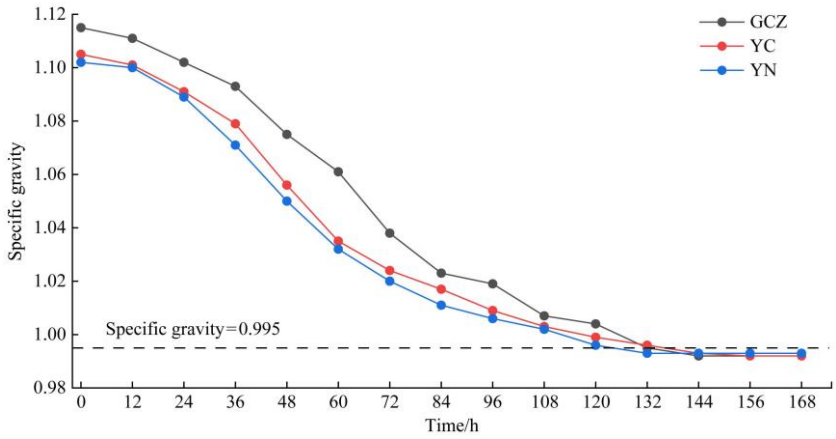
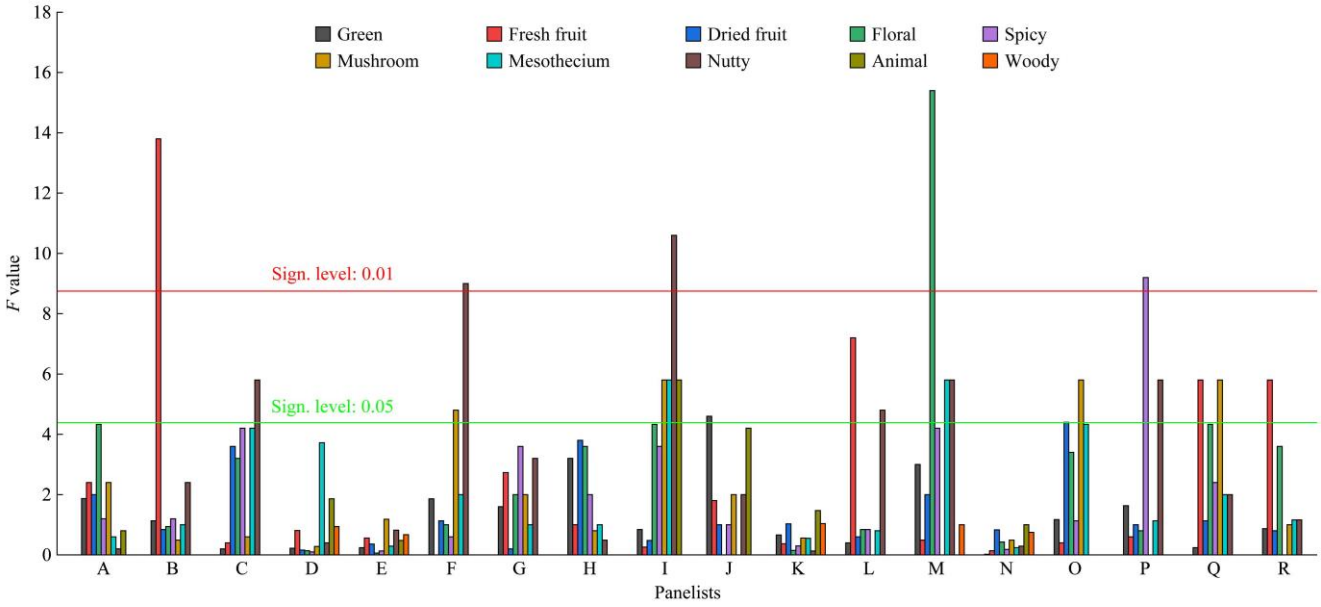
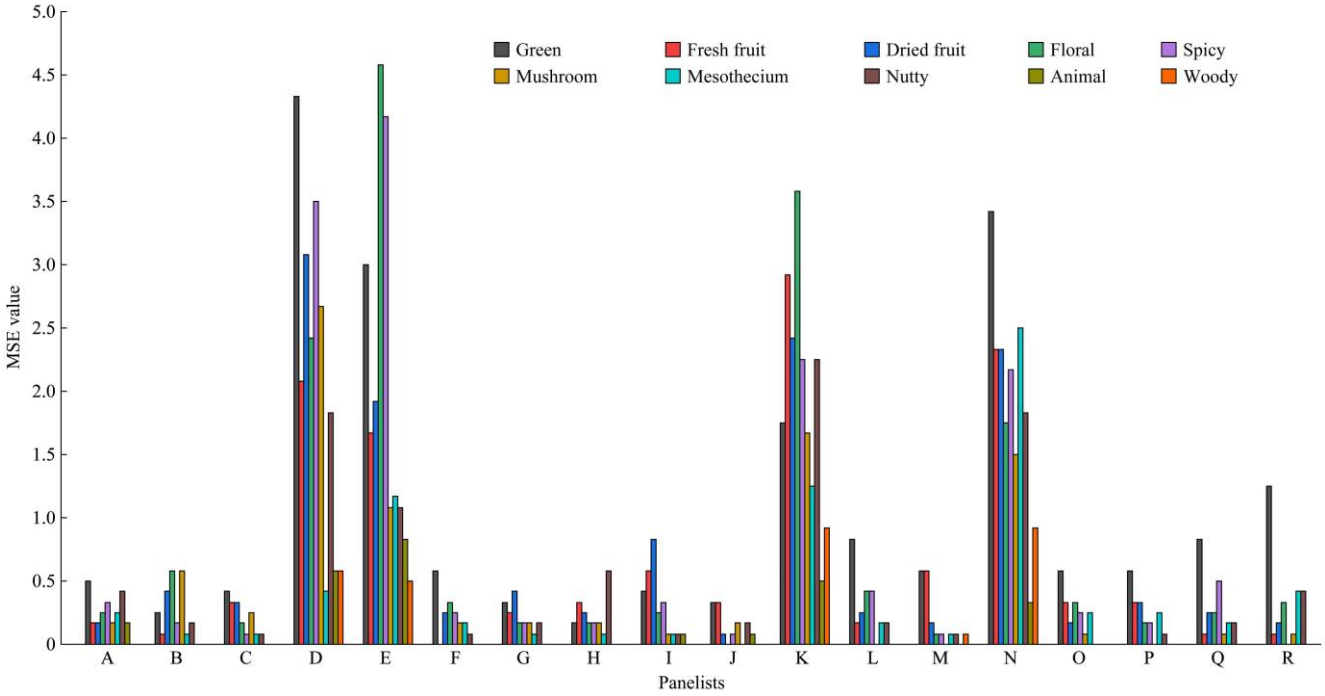


Figure S1 Alcohol fermentation of Cabernet Sauvignon grapes obtained from three sub-regions in Ningxia, China

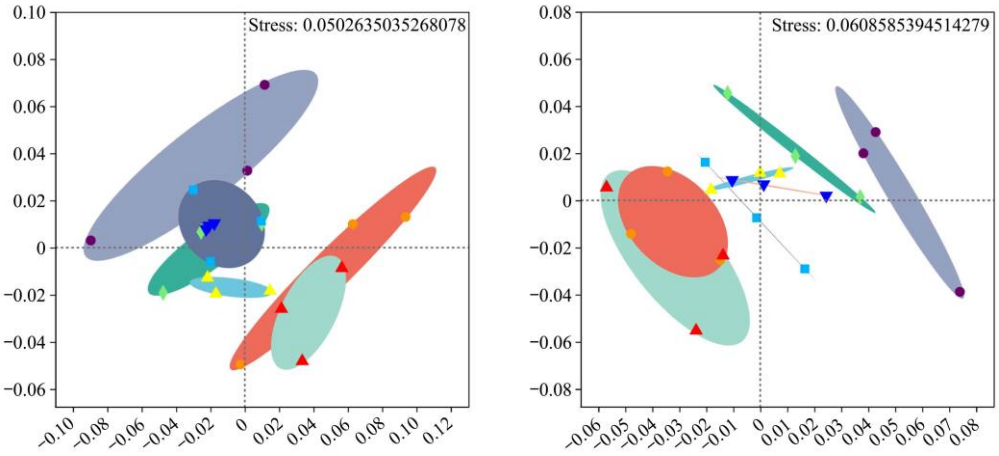


a. The *F* value is used to assess the accuracy of each panelist's evaluation for each aroma descriptor, where a higher value indicates a better accuracy. Among the 18 panelists, 3 of them (E#, K#, and N#) could not accurately recognize all the aroma categories

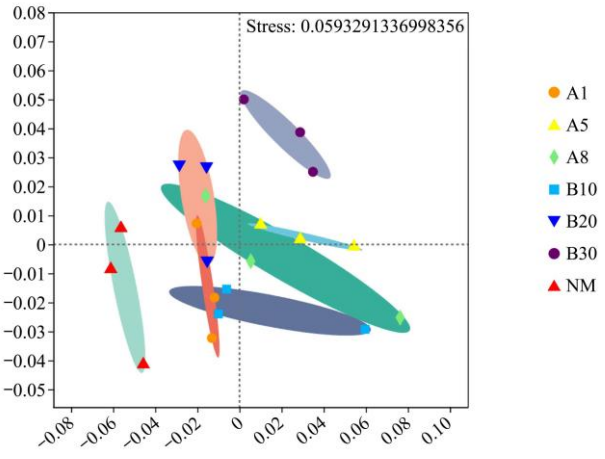


b. The MSE value is utilized to evaluate the repeatability of each panelist during the two tasting sessions, where a lower value illustrates better repeatability. Among the 18 panelists, 4 of them (D#, E#, K#, and N#) had weak reproducibility in two consecutive tastings. Therefore, 14 panelists (A#, B#, C#, F#, G#, H#, I#, J#, L#, M#, O#, P#, Q#, and R#) were chosen to participate in the formal sensory evaluation for the MOX-treated wines.

Figure S2 Accuracy and repeatability of 18 panelists



a. GCZ sub-region b. YC sub-region



c. YN sub-region

Figure S3 NMDS plots of wines with different MOX treatments in three sub-regions