



Blending strategies for wine color modification I: Color improvement by blending wines of different phenolic profiles testified under extreme oxygen exposures



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ARTICLE INFO

Keywords:

Red wine
Blending
Polyphenols
Color modification
Oxygenation
Oxidation

ABSTRACT

Limited oxygenation and over-oxidation experiments were designed to compare the phenolic and chromatic characters of base wines Cabernet Franc (CF), Cabernet Sauvignon (CS), and their counterparts that blended with modifier wines Marselan (MA) and Petit Verdot (PV). In both limited oxygenation and over-oxidation conditions, all blend wines generally contained higher C*_{ab}, a* and Red%, and lower h_{ab}, b* and Yellow% than their base wine counterparts, because MA contributed flavonols (copigments) and anthocyanins, and PV contributed flavanols (anthocyanin derived pigments precursors). Chromatic changes that can be perceived by human eye (ΔE^*_{ab}) in CF based blend wines were more obvious than that of CS based blend wines, which indicate that base wine with lower phenolic concentrations and weak phenolic profiles (CF) might be more prone to be chromatically modified than base wine with higher phenolic concentrations and distinct phenolic profiles (CS). Chemical influences of different blending strategies on anthocyanin derivatives' formations were depending on phenolic profiles of the modifier wines and base wines, and also on the oxygen exposure. The results suggest that the chromatic improvement of base wines could be realized by blending modifier wines under different oxygen exposures.

1. Introduction

Wine color is usually the first organoleptic property perceived by consumers and is to a large extent determined by wine phenolic composition. Anthocyanins directly confer red wines their color, mainly due to the flavylium form (de Freitas, Fernandes, Oliveira, Teixeira, & Mateus, 2017). Other non-anthocyanin phenolics may participate in non-covalent interactions with flavylium and protect flavylium from hydration, lead to an increase in the chromatic absorption (hyperchromic effect) and/or in the visible maximum wavelength (bathochromic effect) of wines (Escribano-Bailón & Santos-Buelga, 2012). The wine color stabilization effect of non-anthocyanin phenolics, referred to as copigmentation effect, together with the color contribution directly conferred by anthocyanins, diminish during wine aging due to the influence of different reactions that phenolic compounds participate

in (de Freitas et al., 2017).

During aging, there are three major reactions anthocyanins participate in, namely, direct polymerization between anthocyanins and flavanols that produces anthocyanin-flavanol (A-T) and/or flavanol-anthocyanin (T-A) oligomers, indirect polymerization between anthocyanins and flavanols via acetaldehyde, which produces purple flavanol-ethyl-anthocyanin adducts, and the formation of pyranoanthocyanins (Li & Duan, 2018). Compared with grape-derived anthocyanins and other anthocyanin derivatives, F-ethyl-A adducts and pyranoanthocyanins are more resistant to hydration and sulfite bleaching and are less sensitive to degradation, therefore, they are recognized as very important wine pigments for aged wines (de Freitas et al., 2017; Escribano-Bailón & Santos-Buelga, 2012).

Co-fermentation and blend (*coupage*) are very common techniques in winemaking. By using more than one kind of grape varieties (co-

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fermentation) or monovarietal wines (blend), winemaker can take advantages of different varieties and make final wine products with ideal organoleptic properties (Escudero-Gilete, González-Miret, & Heredia, 2010; García-Marino et al., 2010, 2013; Monagas, Bartolomé, & Gómez-Cordovés, 2006; Monagas, Martín-Álvarez, Gómez-Cordovés, & Bartolomé, 2007). Nowadays, wine blending is still a very empirical practice based on enologists' own experiences, and winemakers so far generally focused on investigating different blending strategies for grape varieties traditionally used in their local wine industries, which makes the practical experiences and research results hard to be referenced by others.

In this research, we selected four monovarietal wines of different chromatic and phenolic characteristics, namely, Cabernet Franc (CF), Cabernet Sauvignon (CS), Marselan (MA) and Petit Verdot (PV). CF and CS exhibited statistically lower color intensity (lower C^*_{ab} values), less reddish components (lower Red% values), lighter reddish color (lower a^* values), more yellowish components (higher Yellow% values) and deeper yellowish color (higher b^* and h_{ab} values) than MA and PV, therefore, CF and CS were defined as base wines (whose color quality needed to be improved), meanwhile, MA and PV as modifier wines (used to improve color quality of base wines). We blended base wines and modifier wines with different ratios, preserved the base wines and their blend counterparts under both limited oxygenation and over-oxidation conditions. By comparing the color quality and phenolic composition of these wines periodically, and analyzing chromatic-phenolic relationship with the help of statistical tools, we aimed to evaluate the behaviors of different wines preserved under two extreme oxygen exposure conditions, to propose some phenolic chemical explanation for wine blending in color improvement, and to provide scientific guide that can be more widely accepted for making wines of good color quality in practical situation.

2. Materials and methods

2.1. Chemicals

Methanol, acetonitrile, and formic acid of high-performance liquid chromatography (HPLC) grade were purchased from Fisher (Fairlawn, N.J., U.S.A.). Ultrapure water was obtained from a Milli-Q Element water purification system (Millipore, Milford, Mass., U.S.A.). All the phenolic standards were purchased from Sigma-Aldrich (St. Louis, Mo., U.S.A.) except laricitrin, syringetin, syringetin-3-O-glucoside and syringetin-3-O-galactoside were purchased from ChromaDex (Irvine, Calif., U.S.A.),isorhamnetin-3-O-glucoside and myricetin-3-O-glucoside were purchased from Extrasynthese (Genay, France).

2.2. Wines

Monovarietal dry red wines, namely, *Vitis vinifera* L. cv Cabernet Franc (CF), Cabernet Sauvignon (CS), Marselan (MA), Petit Verdot (PV) from 2017 vintage were kindly provided by CITIC Guoan Winery (Xinjiang, China) immediately after fermentation and stabilization. The values of free sulfur dioxide, total sulfur dioxide, volatile acid, titratable acid, pH and alcohol degree of these four wines were shown in Supplementary Material 1, Table A.1, with average levels of 29.8 ± 5.2 mg/L, 46.3 ± 10.3 mg/L, 0.5 ± 0.1 g/L, 5.6 ± 0.5 g/L, 3.8 ± 0.1 and $13.9 \pm 0.6\%$, respectively. CF and CS were used as base wines because according to Table 1, CF and CS exhibited statistically lower color intensity (lower C^*_{ab} values), less reddish components (lower Red% values), lighter reddish color (lower a^* values), more yellowish components (higher Yellow% values) and deeper yellowish color (higher b^* and h_{ab} values) than MA and PV, meanwhile, MA and PV were used as modifier wines due to their statistically higher color intensity (higher C^*_{ab} values), more reddish components (higher Red% values), deeper reddish color (higher a^* values), less yellowish components (lower Yellow% values) and lighter yellowish color (lower b^*

and h_{ab} values) (Table 1).

2.3. Wine blending experiments

Base wines, together with their blend wine counterparts that included 20% or 40% of modifier wines, were treated with limited oxygenation and over-oxidation (Fig. 1). Wines for limited oxygenation were bottled with 750 mL wine bottles and were laid down to ensure that wines can contact with closures to decrease air ingress, these samples were bottle aged for six months and analyzed every 2 months (Fig. 1B). Glass flasks (3L) were used to preserve wines for over-oxidation experiments and each flask was equipped with PSt3 oxygen sensors (Nomacorc SA, Thimister-Clermont, Belgium) for monitoring dissolved oxygen in wines. Wines for over-oxidation (Fig. 1C) were at first put into 3 L glass flasks (1.5 L wine per flask), then enriched with oxygen to trigger oxidation reactions by stirring in air until the dissolved oxygen in each wine sample reached its maximum (which indicated that a saturation of dissolved oxygen was achieved, the dissolved oxygen saturation level in wines was 7.7 ± 0.5 mg/L according to the dissolved oxygen monitor). Air-saturated wines were then stored in glass flasks sealed with air-tight crown caps, to ensure that wines can only receive continuous oxidation from the oxygen initially dissolved and the oxygen from the headspace. These samples were stored for 12 days and were collected and analyzed every 4 days. Each time for sample collecting, flasks were opened and 80 mL wine samples were collected, and flasks' headspace was replenished with air again. After sampling, glass flasks were sealed with air-tight crown caps. During the 12-day preservation, the dissolved oxygen level in all wines was maintained at 6.41 ± 0.66 mg/L according to the dissolved oxygen monitor, which we thought it guaranteed a continuous over-oxidation condition since this dissolved oxygen level was similar to the dissolved oxygen levels of air-saturated wines (6–6.6 mg/L) reported by other researchers (Ferreira, Carrascon, Bueno, Ugliano, & Fernandez-Zurbano, 2015; Gambuti, Picariello, Rinaldi, & Moio, 2018). All samples were stored at 21 ± 0.5 °C condition.

2.4. Chromatic parameters measurements

A spectrophotometer Shimadzu UV-vis 2600 (Shimadzu, Kyoto, Japan) was used to record the wines' absorbance spectra (380–700 nm), with a 1 mm path length quartz cuvette. Ultra-pure water was set as blank reference. Each analysis was performed in triplicate. Prior to analysis, each wine sample of 1 mL was filtered using polyether sulfone membranes (0.45 µm, Membrana Co., Germany).

The color parameters of CIELAB system (L^* , a^* , b^* , C^*_{ab} , h_{ab}) were determined according to the CIELAB method (Ayala, Echávarri, & Negueruela, 1997). Color difference (ΔE^*_{ab}) between blend wines and their base wine counterparts (CFM82, CFM64, CFP82, CFP64 vs. CF; CSM82, CSM64, CSP82, CSP64 vs. CS) during each period of experiments was calculated by the following equation:

$$\Delta E^*_{ab} = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$

ΔE^*_{ab} value ≥ 3 represents wine chromatic changes that can be perceived by human eye (Martínez, Melgosa, Pérez, Hita, & Negueruela, 2001).

Recording absorbance values at 420, 520 and 620 nm, percentage of yellow (Yellow%), percentage of red (Red%) and percentage of blue (Blue%) were calculated according to the Glories method (Glories, 1984):

$$\text{Yellow\%} = 100\% \times \text{Abs}_{420}/(\text{Abs}_{420} + \text{Abs}_{520} + \text{Abs}_{620})$$

$$\text{Red\%} = 100\% \times \text{Abs}_{520}/(\text{Abs}_{420} + \text{Abs}_{520} + \text{Abs}_{620})$$

$$\text{Blue\%} = 100\% \times \text{Abs}_{620}/(\text{Abs}_{420} + \text{Abs}_{520} + \text{Abs}_{620})$$

For each wine sample (5 mL per sample), the color contribution of

Table 1
Initial chromatic comparison of four monovarietal wines.

Chromatic parameters	CF	CS	MA	PV
L*	64.85 ± 1.26a	43.16 ± 1.2c	49.52 ± 0.72b	39.15 ± 2.04d
a*	37.16 ± 1.11c	47.59 ± 2.89b	52.25 ± 0.54a	53.56 ± 1.1a
b*	7.99 ± 0.1b	9.84 ± 0.28a	5.37 ± 0.02c	2.02 ± 0.11d
C* _{ab}	38.01 ± 1.07c	48.6 ± 0.77b	52.53 ± 0.53a	53.6 ± 1.09a
h _{ab}	12.15 ± 0.49a	11.71 ± 0.01b	5.86 ± 0.08c	2.16 ± 0.16d
Yellow%	36.85 ± 0.31a	36.14 ± 0.35a	31.75 ± 0.14b	31.29 ± 0.11b
Red%	51.73 ± 0.15c	50.1 ± 0.35d	57.19 ± 0.14a	55.42 ± 0.03b
Blue%	11.42 ± 0.16b	13.75 ± 0.01a	11.06 ± 0.01b	13.29 ± 0.08a
Fr%	20.9 ± 0.63b	16.13 ± 0.34c	30.54 ± 1.5a	16.53 ± 0.23c
Co%	42.67 ± 0.70a	35.96 ± 0.33b	39.34 ± 1.44a	30.66 ± 0.29c
Po%	36.43 ± 0.07c	47.91 ± 0.01b	30.12 ± 0.07d	52.8 ± 0.06a

Notes:

^aData are mean value ± deviation (n = 3). Different superscripts in the same row indicate significant differences (*p* < 0.05).

^bFr%: the color contribution of free anthocyanins; Co%: the color contribution of copigments; Po%: the color contribution of anthocyanin derived pigments.

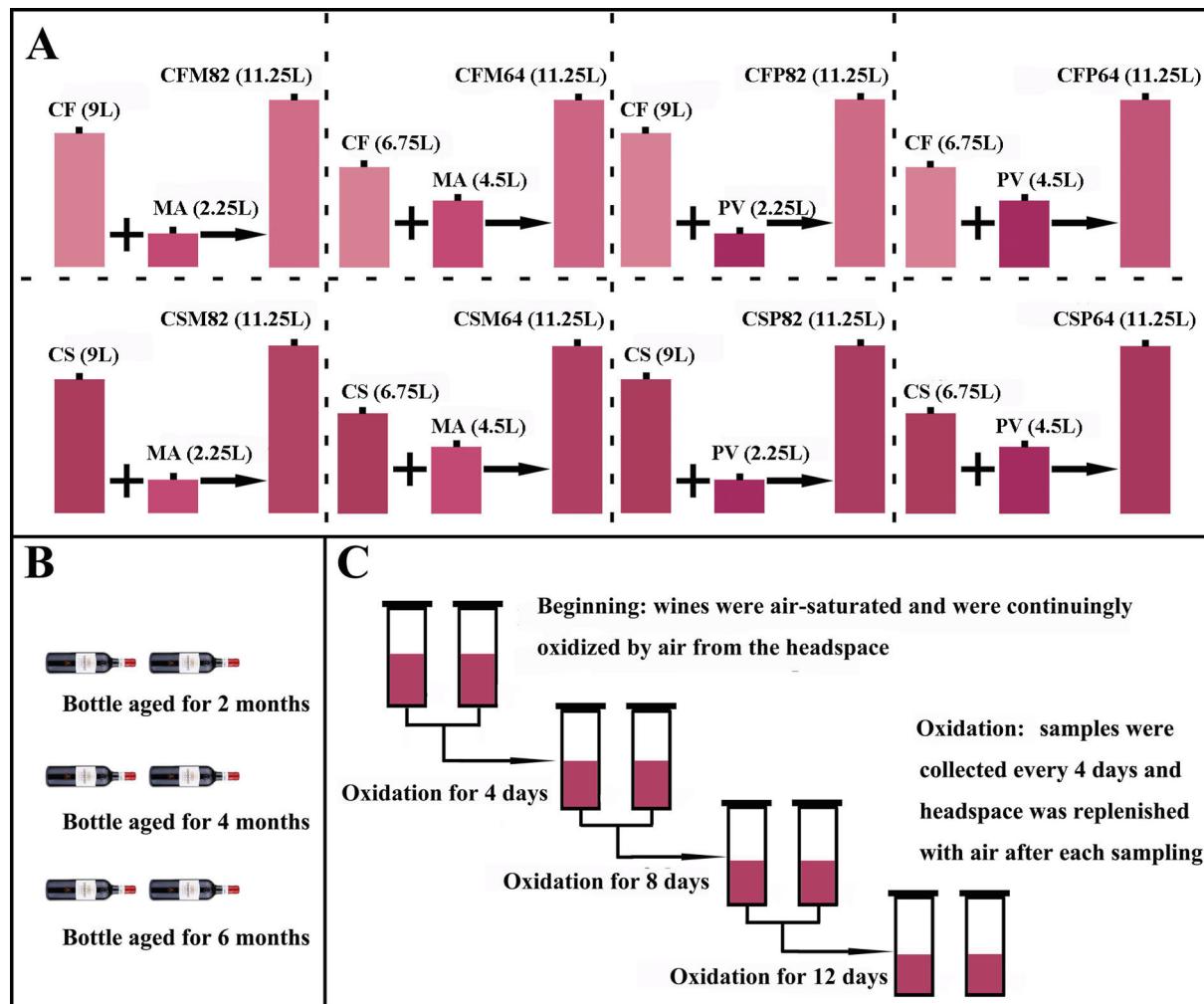


Fig. 1. A: Wine blending experiment; B: Limited oxygenation treatment; C: Over-oxidation treatment. CFM82: blend wine of 80% CF and 20% MA; CFM64: blend wine of 60% CF and 40% MA; CFP82: blend wine of 80% CF and 20% PV; CFP64: blend wine of 60% CF and 40% PV; CSM82: blend wine of 80% CS and 20% MA; CSM64: blend wine of 60% CS and 40% MA; CSP82: blend wine of 80% CS and 20% PV; CSP64: blend wine of 60% CS and 40% PV.

free anthocyanins (Fr%), copigments (Co%) and anthocyanin derived pigments (Po%) were determined according to the method reported by Boulton (Boulton, 1996). In brief, each wine sample was adjusted to pH 3.6 and divided into three sub-sample groups (group1, group2, group3). For group1 (2 mL per sub-sample), 20 µL of 10% (v/v) acetaldehyde solution was added to each wine sub-sample and incubated for 45 min at room temperature. For group2 (2 mL per sub-sample), 160 µL of 6%

potassium metabisulfite was added to each wine sub-sample. For group3 (250 µL per sub-sample), each wine sub-sample was diluted 20 times with model wine solution (12% ethanol, 5 g/L tartaric acid, 0.2 mol/L NaCl, pH 3.6). Afterwards, all sub-samples were filtered through cellulose filters (0.45 µm, Membrana Co., Germany), and the absorbance values of sub-samples from group1, group2 and group3 at 520 nm were recorded and labeled as Abs^{acet}, Abs^{SO2} and Abs²⁰,

respectively. Fr %, Co % and Po % of each wine sample were calculated according to the following equations:

$$\text{Fr\%} = 100\% \times (20 \times \text{Abs}^{20} - \text{Abs}^{\text{SO}_2})/\text{Abs}^{\text{acet}}$$

$$\text{Co\%} = 100\% \times (\text{Abs}^{\text{acet}} - 20 \times \text{Abs}^{20})/\text{Abs}^{\text{acet}}$$

$$\text{Po\%} = 100\% \times \text{Abs}^{\text{SO}_2}/\text{Abs}^{\text{acet}}$$

2.5. Phenolic compounds analysis

Phenolic compounds were identified and quantified using high performance liquid chromatography triple-quadrupole tandem mass spectrometry (HPLC-QQQ-MS/MS). The HPLC-QQQ-MS/MS methods were shown to be reliable for detecting and quantifying commercial standards for 45 non-anthocyanin and five grape derived anthocyanin compounds (delphinidin-3-O-glucoside, cyanidin-3-O-glucoside, petunidin-3-O-glucoside, peonidin-3-O-glucoside, malvidin-3-O-glucoside), and the acylated (acetyl/coumaroyl) forms of these five anthocyanins could also be detected based on their mass information and chromatographic peak features (elution order and disturbing peaks exclusion) and their quantification was on the basis of the calibration curve of malvidin-3-O-glucoside (Li et al., 2016, 2017).

For anthocyanin derivatives, given the diversity of F-ethyl-A adducts and pyranoanthocyanins in wines, we only detected malvidin-based F-ethyl-A adducts and pyranoanthocyanins, namely, (epi)catechin-ethyl-malvidin-3-O-(acetyl/coumaroyl)-glucoside (F-ethyl-A adducts), vinylformic acid adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (vitisin A), acetaldehyde adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (vitisin B), 4-vinylcatechol adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (hydroxyphenyl-pyranoanthocyanins), 4-vinylphenol adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (hydroxyphenyl-pyranoanthocyanins), 4-vinylguaiacol adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (hydroxyphenyl-pyranoanthocyanins) and vinyl-(epi)catechin adducted malvidin-3-O-(acetyl/coumaroyl)-glucoside (flavanyl-pyranoanthocyanins). Their identification was based on their mass information and chromatographic peak features (elution order and disturbing peaks exclusion) and their quantification was on the basis of the calibration curve of malvidin-3-O-glucoside (Supplementary Material 2, Fig. A.1–Fig. A.7) (Li et al., 2016).

HPLC analysis was performed using an Agilent series 1200 instrument (Agilent Technologies, Santa Clara, Calif., USA) fitted with a Poroshell 120 EC-C18 column (150 × 2.1 mm, 2.7 µm, Agilent Technologies). Water was used as mobile phase A and acetonitrile/methanol (50:50, v/v) as mobile phase B, both with 0.1% formic acid added, and the elution gradient was from 10% to 46% B for 28 min, from 46% to 10% B for 1 min, with a flow rate at 0.4 mL/min. Column temperature was set at 55 °C. An Agilent 6410 QQQ instrument equipped with an electrospray ionization source (ESI) was used, with a spray voltage of 4 kV in negative mode and positive mode for non-

anthocyanin phenolics and anthocyanin compounds, respectively. The source temperature was kept at 150 °C, while the gas temperature was 350 °C. The gas flow rate was 12 L/h with the nebulizer pressure being 35 psi. The [M-H]⁻ and [M + H]⁺ ions were selected as precursor ions of non-anthocyanin phenolics and anthocyanin compounds, respectively. Multiple reaction monitoring (MRM) mode was selected for both identification and quantification. All wine samples of 1 mL were filtered using polyether sulfone membranes (0.45 µm, Membrana Co., Germany) prior to analysis and 1 µL being injected directly for analysis. Each wine sample was analyzed in triplicate. Phenolic identification and quantification analysis was performed by Agilent Mass Hunter workstation software version B.04.00.

2.6. Statistical analysis

Significant differences were determined by one-way analysis of variance (ANOVA) and Turkey's honestly significant difference test using SPSS program (SPSS Inc., Chicago, Ill., USA). Principal component analysis (PCA) was performed using SIMCA 14.1 program (Umetrics, Umeå, Sweden). Adobe Photoshop 7.0 (San Jose, CA, USA) was used to create artwork.

3. Results and discussion

In total, there were 21 non-anthocyanin phenolic compounds successfully identified and quantified, including 7 flavanols, 9 flavonols, 3 hydroxybenzoic acids and 2 hydroxycinnamic acids, other non-anthocyanin compounds were excluded due to their trace amounts or absence in wines. As for anthocyanin compounds, 15 grape derived anthocyanins and 33 anthocyanin derivatives were detected and quantified, and the anthocyanin data was simplified into 5 grape derived anthocyanin families and 7 anthocyanin derivative families regardless of their acylation, for the purpose of minimizing the variables and reducing the complexity in the following statistical analysis (Supplementary Material 3, Table A.2). Initial phenolic compositions of the four monovarietal wines (CF, CS, MA, PV) are listed in Table 2. When comparing the concentration of each phenolic group in CF with those of other three wines, it can be seen that all of its phenolic groups had the lowest or second lowest concentrations, except for hydroxyphenyl-pyranoanthocyanins (especially, second lowest concentrations of anthocyanins, flavonols and flavanols). Therefore, its relatively lower color intensity (lower C*_{ab} values), less reddish components (lower Red% values), lighter reddish color (lower a* values), more yellowish components (higher Yellow% values) and deeper yellowish color (higher b* and h_{ab} values) might be caused by insufficient quantity of pigments (anthocyanins), copigments (flavonols) and anthocyanin derivative precursors (flavanols). CS, on the contrary, had the highest or second highest concentrations in each phenolic group, its relatively lower color intensity (lower C*_{ab} values), less reddish components (lower Red% values), lighter reddish color (lower a* values), more yellowish

Table 2
Initial groups of phenolics in four monovarietal wines.

Phenolic concentrations	CF	CS	MA	PV
Flavanols (mg/L)	216.77 ± 2.16c	238.96 ± 5.9b	172.09 ± 1.88d	289.98 ± 3.42a
Flavonols (mg/L)	56.3 ± 3.75c	63.63 ± 3.35b	80.09 ± 0.31a	58.68 ± 1.91bc
Hydroxybenzoic acids (mg/L)	14.74 ± 0.04c	22.72 ± 1.9a	5.95 ± 0.19d	16.34 ± 0.4b
Hydroxycinnamic acids (mg/L)	0.82 ± 0.02c	51.36 ± 6.07a	Tr	21.02 ± 0.39b
Anthocyanins (mg/L)	359.86 ± 1.42c	371.81 ± 1.19b	384.91 ± 2.92a	280.48 ± 4.42d
Flavanol-ethyl-anthocyanin adducts (mg/L)	0.99 ± 0.08d	4.33 ± 0.09b	1.57 ± 0.01c	6.66 ± 0.08a
Flavanyl-pyranoanthocyanins (mg/L)	0.13 ± 0.01c	0.62 ± 0.01b	0.13 ± 0.01c	0.9 ± 0.01a
Hydroxyphenyl-pyranoanthocyanins (mg/L)	1.91 ± 0.05a	2.25 ± 0.01a	1.12 ± 0.03b	0.45 ± 0.01c
Vitisins (mg/L)	2.45 ± 0.03d	6.68 ± 0.06a	3.22 ± 0.06c	6.21 ± 0.15b

Notes:

^aData are mean value ± deviation (n = 3). Different superscripts in the same row indicate significant differences (p < 0.05).

^bTr: trace.

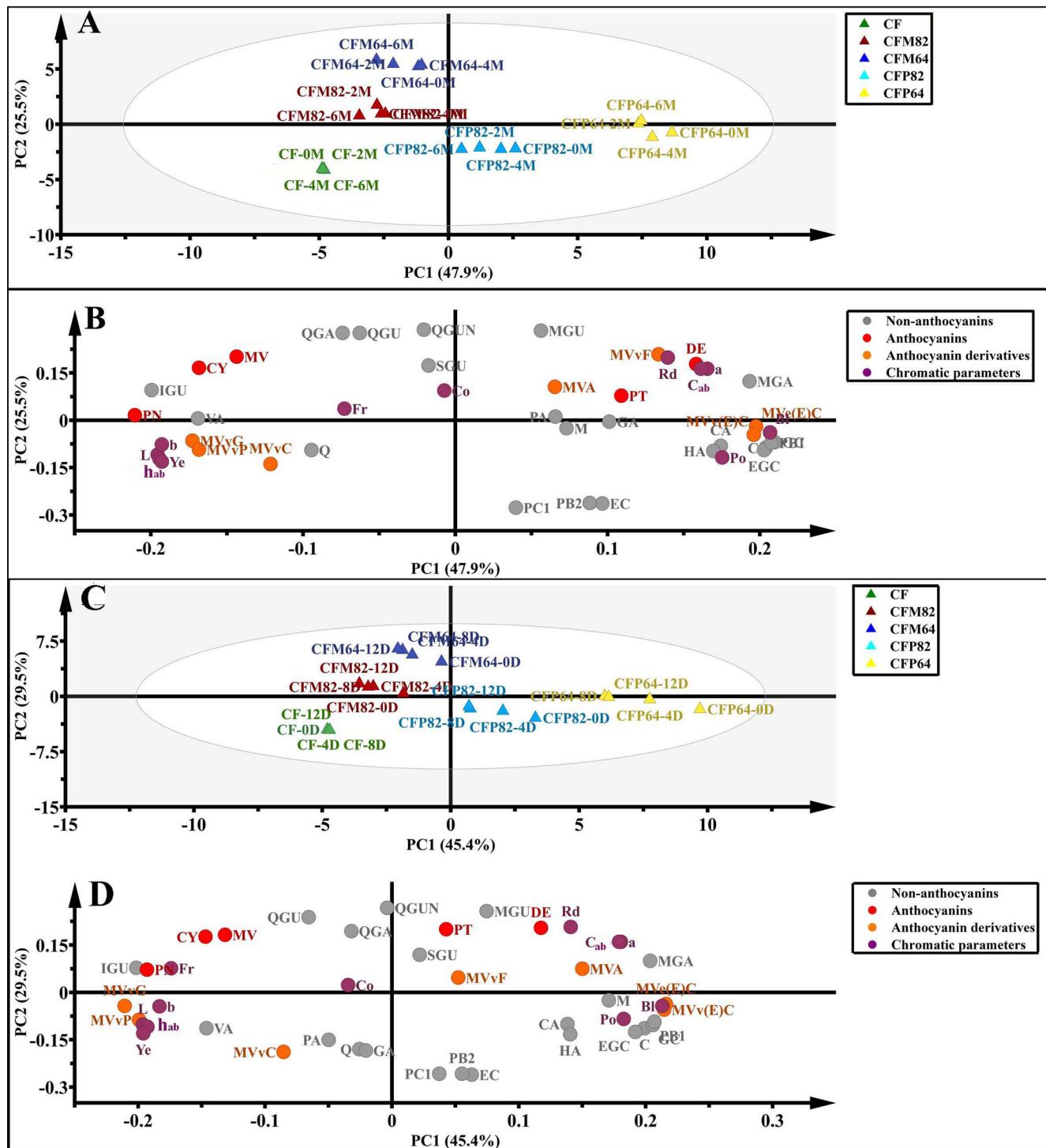


Fig. 2. Principal component analysis of CF base wine and its blend counterparts treated with limited oxygenation (A, B) and over-oxidation (C, D). Abbreviations: Flavanols: (+)-catechin (C), (-)-epicatechin (EC), (+)-gallocatechin (GC), (-)-epigallocatechin (EGC), procyanidin B1 (PB1), procyanidin B2 (PB2), procyanidin C1 (PC1); Flavonols: syringetin-3-O-glucoside (SGU), isorhamnetin-3-O-glucoside (IGU), myricetin (M), myricetin-3-O-galactoside (MGA), myricetin-3-O-glucoside (MGU), quercetin (Q), quercetin-3-O-galactoside (QGA), quercetin-3-O-glucoside (QGU), quercetin-3-O-glucuronide (QGUN); Hydroxybenzoic acids: gallic acid (GA), vanillic acid (VA), protocatechuic acid (PA); Hydroxycinnamic acids: caffeoic acid (CA), 3-hydroxycinnamic acid (HA); Anthocyanins: malvidin-3-O-(acetyl/coumaroyl)-glucoside (MA), petunidin-3-O-(acetyl/coumaroyl)-glucoside (PT), peonidin-3-O-(acetyl/coumaroyl)-glucoside (PN), delphinidin-3-O-(acetyl/coumaroyl)-glucoside (DE), cyanidin-3-O-(acetyl/coumaroyl)-glucoside (CY); Flavanol-ethyl-anthocyanin adducts: malvidin-3-O-(acetyl/coumaroyl)-glucoside-ethyl-(epi)catechin [MV_e(E)C]; Vitisins: vitisin A (MVvF), vitisin B (MVA); Hydroxyphenyl-pyrananthocyanins: malvidin-3-O-(acetyl/coumaroyl)-glucoside-vinylguaiacol (MVvG), malvidin-3-O-(acetyl/coumaroyl)-glucoside-vinylcatechol (MVvC), malvidin-3-O-(acetyl/coumaroyl)-glucoside-vinylphenol (MVvP); Flavanyl-pyrananthocyanins: malvidin-3-O-(acetyl/coumaroyl)-glucoside-ethyl-(epi)catechin [MV_v(E)C]; Chromatic parameters: L* (L), a* (a), b* (b), C*_{ab} (C_{ab}), h_{ab} (h_{ab}), Yellow% (Ye), Red% (Rd), Blue% (Bl), Fr% (Fr), Co% (Co), Po% (Po).

Table 3Visual differences (ΔE^*_{ab}) of CF based blend wines compared with CF during every period of limited oxygenation and over-oxidation treatments.

Sampling point	CFM82	CFM64	CFP82	CFP64
0	6.33 ± 1.67c	9.67 ± 2.29b	8.05 ± 1.74bc	16.23 ± 1.48a
2 M	9.41 ± 0.64c	10.57 ± 0.19b	7.92 ± 0.39d	17 ± 1.04a
4 M	5.22 ± 0.6d	10.42 ± 1.53b	8.18 ± 0.21c	14.93 ± 1.03a
6 M	4.99 ± 0.95c	8.07 ± 2.19b	7.29 ± 2.49bc	16.17 ± 1.75a
4D	6.44 ± 1.27d	11.04 ± 0.96b	9.43 ± 0.47c	18.82 ± 0.33a
8D	6.77 ± 0.59c	11.15 ± 0.02b	6 ± 0.34c	17.95 ± 0.46a
12D	6.23 ± 1.1d	10.05 ± 0.19b	8.55 ± 0.05c	17.22 ± 0.68a

Notes:

^aData are means ± deviation (n = 3). Different superscripts in the same row indicate significant differences (p < 0.05);^bSampling points: initial point (0), two-month limited oxygenation (2 M), four-month limited oxygenation (4 M), six-month limited oxygenation (6 M), four-day over-oxidation (4D), eight-day over-oxidation (8D), twelve-day over-oxidation (12D).

components (higher Yellow% values) and deeper yellowish color (higher b* and h_{ab} values) might be due to an unbalanced proportion of anthocyanins to some pyranoanthocyanin precursors, which lead to an abundant concentration of yellow-orange pyranoanthocyanin pigments (CS had the second highest concentration of flavanyl-pyranoanthocyanins, and highest concentrations of hydroxyphenyl-pyranoanthocyanins and vitisins). MA had the highest concentrations of flavonols and anthocyanins, which could explain its relatively higher color intensity (higher C*_{ab} values), more reddish components (higher Red% values), deeper reddish color (higher a* values), less yellowish components (lower Yellow% values) and lighter yellowish color (lower b* and h_{ab} values) and also its high Co% and Fr% (Table 1), since flavonols were reported as good copigments (Escribano-Bailón & Santos-Buelga, 2012), and anthocyanins confer wines red hue directly. PV had the highest concentrations of flavonols, flavanol-ethyl-anthocyanin adducts and flavanyl-pyranoanthocyanins, and the lowest concentration of anthocyanins. The relatively high flavanol-anthocyanin ratio (ratio = 1, toward ratios from 0.4 to 0.6 in the other wines) could benefit the formation of stable pigments such as flavanol-ethyl-anthocyanin adducts and flavanyl-pyranoanthocyanins, which may be the reason of its relatively higher color intensity (higher C*_{ab} values), more reddish components (higher Red% values), deeper reddish color (higher a* values), less yellowish components (lower Yellow% values) and lighter yellowish color (lower b* and h_{ab} values), and highest Po% (Table 1).

3.1. Chromatic and phenolic modifications on CF base wine

Principal component analysis (PCA) was used to find out the chromatic and phenolic modification on CF and CS treated with different blending strategies. Before analysis, all phenolic and chromatic data were normalized first, and values of each chromatic parameter and phenolic compound of wine samples from a specific period (for example, all wines bottle aged for 4 months: CF-4M, CFM82-4M, CFM84-4M, CFP82-4M, CFP84-4M) divided by the values of their chromatic and phenolic counterparts in CF of the same period (CF-4M). In this way, values of each chromatic parameter and phenolic compound of all CF base wines (CF-0M, CF-2M, CF-4M, CF-6M, CF-0D, CF-4D, CF-8D, CF-12D) were standardized to 1, and chromatic and phenolic values of other blend wines were normalized according to the division of chromatic and phenolic values of CF counterparts of the same period. Such data normalization would effectively diminish time factor's impact on chromatic and phenolic differences of wine samples and was useful to compare blend wines with their base wine counterparts, thus help summarizing chromatic and phenolic modification effects of different blending strategies.

From Fig. 2, we can see that PCs (PC1 and PC2) explained 73.4% and 74.9% of total variance under limited oxygenation and over-oxidation, respectively. Wine samples made from a same blending strategy gathered closely, regardless of their sampling time background.

Compared with CF samples, blend wine samples CFM and CFP were more prone to have higher scores on PC2 and PC1 as the proportion of modifier wines (MA and PV) increase, respectively.

Fig. 2B and D show the loading plots of different variables under limited oxygenation and over-oxidation, respectively. From both Fig. 2B and D, we can see that the blending of PV into CF effectively increased wines' phenolic concentrations, including some anthocyanins (petunidin and delphinidin based anthocyanins) and anthocyanin derivatives (vitisins and polymerized pigments such as anthocyanin-ethyl-flavanol adducts and flavanyl-pyranoanthocyanins), all flavonols and some other non-anthocyanin phenolics (some myricetin based flavonols and phenolic acids), and also enhanced wines' chromatic values of a*, C*_{ab}, Red%, Blue% and Po%. On the other hand, variables that positively related with PC2 indicate that the blending of MA into CF effectively increased wines' phenolic concentrations such as all anthocyanins and vitisins, and most flavonols, and also enhanced wines' chromatic values of a*, C*_{ab}, Red%, Co% and Fr%.

The results in Fig. 2 indicate that for CF wine, its blend wine counterparts exhibited almost the same chromatic modifications under both limited oxygenation and over-oxidation conditions when the same modifier wine (MA or PV) was used, namely, the use of both MA and PV effectively improved CF wine's color quality because all blend wines had higher a*, C*_{ab} and Red% values than CF. All blend wines showed visual differences compared with their base wine counterparts CF, and PV blending generally led to more obvious visual differences than MA blending under both limited oxygenation and over-oxidation conditions (Table 3).

3.2. Chromatic and phenolic modifications on CS base wine

Fig. 3 illustrates the wine blending effects on CS, we can see that PCs (PC1 and PC2) explained 65% and 73.6% of total variance under limited oxygenation and over-oxidation, respectively. Samples made from a same blending strategy gathered closely, regardless of their sampling time background.

Fig. 3B shows the loading plots of different variables under limited oxygenation. We can see that the blending of PV into CS effectively increased wine phenolic compounds that positively related with PC1, including some anthocyanins (petunidin and delphinidin based anthocyanins) and anthocyanin derivatives (especially polymerized pigments such as anthocyanin-ethyl-flavanol adducts and flavanyl-pyranoanthocyanins), all flavonols and some other non-anthocyanin phenolics (myricetin-3-O-galactoside and hydroxybenzoic acids), and also enhanced wines' chromatic values of a*, C*_{ab}, Red%, Blue% and Po%. CSM samples got very higher scores on PC2 than CS, therefore, the blending of MA into CS effectively increased phenolic compounds that positively related with PC2, such as all anthocyanins and most flavonols, some hydroxyphenyl-pyranoanthocyanins and hydroxycinnamic acids, and also enhanced wines' chromatic values of a*, C*_{ab}, Red%, Fr% and Co%.

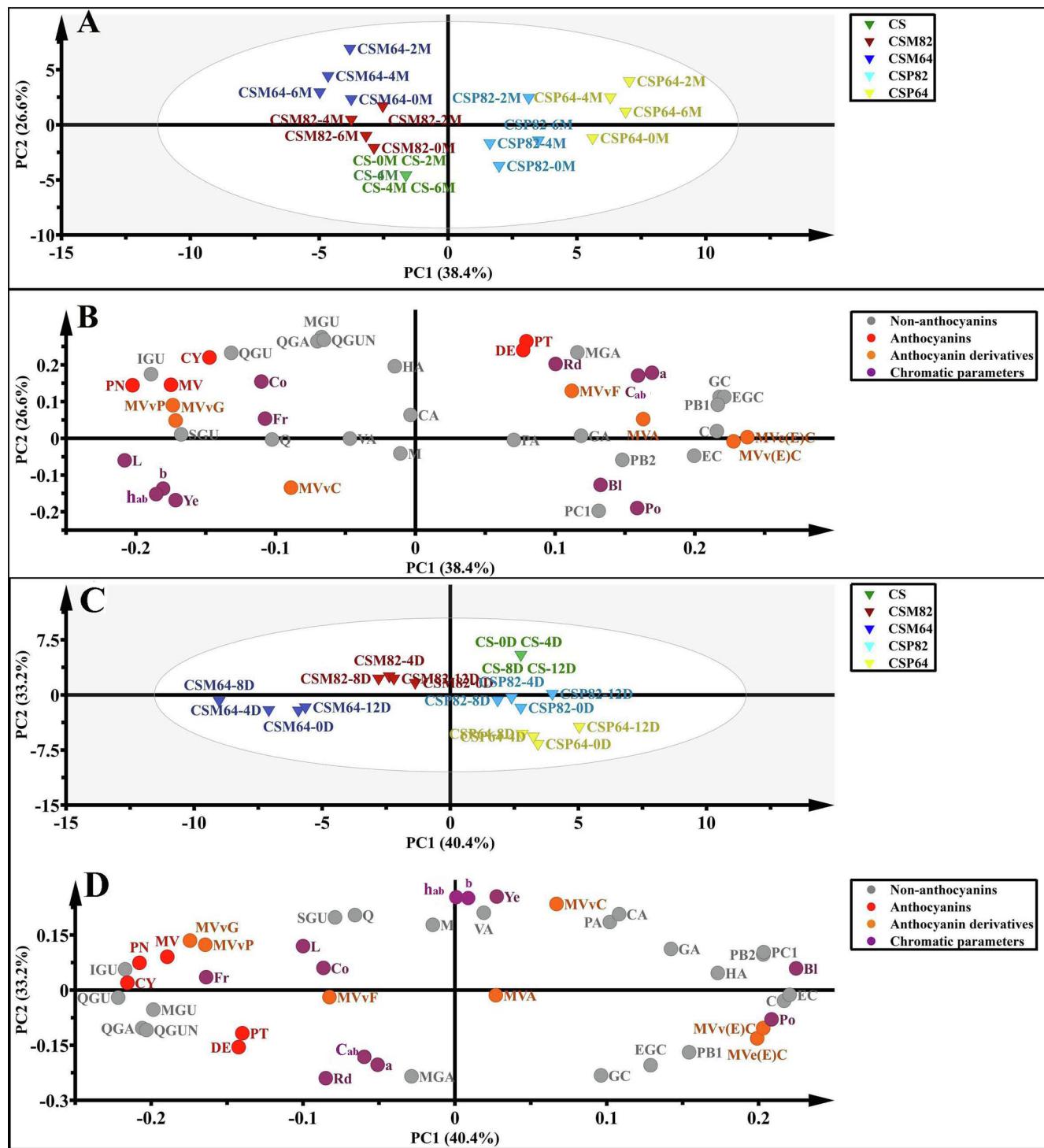


Fig. 3. Principal component analysis of CS base wine and its blend counterparts treated with limited oxygenation (A, B) and over-oxidation (C, D).

Fig. 3D shows the loading plots of different variables under over-oxidation. We can see that CS samples mainly had higher hydroxyphenyl-pyrananthocyanins and phenolic acids, and exhibited more yellowish components (higher Yellow% values) and deeper yellowish color (higher b^* and h_{ab} values) than their blend wine counterparts CSM and CSP. The results in Fig. 3 indicate that for CS samples, their blend wine counterparts exhibited color improvements in containing more a^* , C^*_{ab} and Red%, and less b^* , h_{ab} and Yellow%, than CS under limited oxygenation and over-oxidation conditions, respectively. Comparing the blending strategies on CF and CS, CF based blend wines were obvious more prone to exhibit visual differences than their CS

counterparts and to achieve better chromatic modification, since CS based blend wines always had lower ΔE^*_{ab} values and some visual differences could not be perceived by human eye (Table 3, Table 4). Given limited oxygenation and over-oxidation represent two extremes of oxygen exposures in wines, the effects of blending on color improvements and the variations of visual differences indicate that in practical situation, the blend of base wines and modifier wines should be carefully considered, mainly depending on phenolic profiles of different wines and maturation, aging and preservation plans made by winemakers (such as oxygen control and preservation time).

Table 4

Visual differences (ΔE^*_{ab}) of CS based blend wines compared with CS during every period of limited oxygenation and over-oxidation treatments.

Sampling point	CSM82	CSM64	CSP82	CSP64
0	2.34 ± 0.21d	5.1 ± 0.3b	4.1 ± 0.56c	7.6 ± 0.62a
2 M	1.86 ± 0.01d	3.27 ± 0.46c	6.43 ± 0.29b	7.84 ± 0.87a
4 M	2.7 ± 0.77b	2.84 ± 0.13b	2.74 ± 0.02b	6.88 ± 0.77a
6 M	1.4 ± 0.27d	2.3 ± 0.39c	5.07 ± 0.09b	8.07 ± 0.75a
4D	3.83 ± 0.03b	3.87 ± 0.33ab	2.78 ± 0.6c	4.52 ± 0.33a
8D	3.07 ± 0.1b	4.32 ± 0.4a	1.33 ± 0.09c	4.06 ± 0.17a
12D	1.77 ± 0.25c	4.46 ± 0.25a	2.89 ± 0.09b	4.52 ± 0.64a

Notes:

^aData are means ± deviation (n = 3). Different superscripts in the same row indicate significant differences (p < 0.05).

3.3. Chemical influences of different blending strategies on anthocyanin derivatives' formation

All blend wines showed better color quality compared with their base wine counterparts, namely, higher color intensity (higher C^*_{ab} values), more reddish components (higher Red% values), deeper reddish color (higher a^* values), less yellowish components (lower Yellow% values) and lighter yellowish color (lower b^* and h_{ab} values). MA mainly contributed flavonols and anthocyanins to the CF and CS wines, which should be the main reason for MA blend samples' color improvement compared with their base wine counterparts because flavonols are good copigments, and anthocyanins confer wines reddish hue directly. Although PV had the lowest anthocyanin concentration among four monovarietal wines, it had the highest flavanol concentration (Table 2). Blending of PV might help increase tannin-anthocyanin ratio, thus promoted the formation of stable pigments (Picariello, Gambuti, Picariello, & Moio, 2017). This result is similar to the work reported by García-Marino (García-Marino, Escudero-Gilete, Heredia, Escrivano-Bailón, & Rivas-Gonzalo, 2013) and Monagas (Monagas et al., 2007). In their work on phenolic and chromatic investigations for base wine and different blending wines, wines of higher Red%, a^* , C^*_{ab} values and lower Yellow%, b^* and h_{ab} values contained relatively more flavonols, whereas their contents of anthocyanins or flavonols were not always very high. Besides, other researchers found that during both forced oxidation and bottling aging conditions, wines with higher tannin-anthocyanin ratios benefited the increase of Abs520 and/or Abs620 (Gambuti et al., 2017; Picariello, Gambuti, Petracca, Rinaldi, & Moio, 2018).

To observe the chemical influences of different blending strategies, chromatic and phenolic data of base wines (CF, CS), modifier wines (MA, PV) and blend wines with higher proportions of modifier wines (CFM64, CFP64, CSM64, CSP64) were collected at the end of limited oxygenation (6 M) and over-oxidation (12D), and blend wines' actual and theoretic anthocyanin derivatives concentrations were compared. The theoretic concentrations were calculated as the sum of 60% base

Table 5

Chemical influences of high proportion blending (40%) on wines' anthocyanin derivatives formations.

Attributes (limited oxygenation)	CFM64	CFP64	CSM64	CSP64
Flavanol-ethyl-anthocyanin adducts	None	Positive	Negative	Negative
Flavanyl-pyranoanthocyanins	Negative	Negative	Negative	Negative
Hydroxyphenyl-pyranoanthocyanins	None	Negative	Positive	Negative
Vitisins	Negative	Negative	Negative	Negative
Attributes (over-oxidation)	CFM64	CFP64	CSM64	CSP64
Flavanol-ethyl-anthocyanin adducts	None	Positive	None	Negative
Flavanyl-pyranoanthocyanins	None	Positive	Negative	Negative
Hydroxyphenyl-pyranoanthocyanins	Negative	Positive	Negative	Negative
Vitisins	Positive	Positive	Negative	Negative

wine anthocyanin derivatives concentrations and 40% modifier wine anthocyanin derivatives concentrations. As can be seen in Fig. 4 and Table 5, actual anthocyanin derivatives concentrations in blend wines that were lower or higher than the theoretic anthocyanin derivatives concentrations corresponds to negative or positive chemical influences, respectively.

Under limited oxygenation, although the blend of MA into CF increased anthocyanins concentration, concentrations of other precursors (such as flavanols) of anthocyanin derivatives decreased. Besides, the increase of flavonols enhanced copigmentation effect, which might protect anthocyanins from participating in other reactions (Escribano-Bailón & Santos-Buelga, 2012). Therefore, the overall formations of anthocyanin derivatives were generally counteracted or depressed in CFM64 wines. As for the blending of PV into CF, both flavanols concentration and flavanols-anthocyanins ratio increased, which benefited the formation of flavanol-ethyl-anthocyanin adducts. Since flavanol-ethyl-anthocyanin adducts was reported to form very quickly due to the reactivity of acetaldehyde (Li & Duan, 2018), and anthocyanins concentration decreased due to the blending of PV into CF, formations of other anthocyanin derivatives might be depressed under the dominance of Pathway1 (Fig. 5).

Under limited oxygenation, the blend of MA into CS decreased both the flavanols concentration and flavanols-anthocyanins ratio, which may be disadvantageous for the anthocyanin derivatives' formations that flavanols involved in (Pathway1, Pathway2 and Pathway3 in Fig. 5) (Li & Duan, 2018). Besides, the increase of flavonols could also reduce free anthocyanins that participate in various reactions. The only exception was the formation of hydroxyphenyl-pyranoanthocyanins, since CS contained sufficient concentration of hydroxycinnamic acids (Table 1), which could increase continually due to hydrolysis of their tartaric esters during the following preservation, thus benefited Pathway4 (Fig. 5) (Quagliari, Jourdes, Waffo-Teguo, & Teissedre, 2017). The chemical reactions of all anthocyanin derivatives were depressed due to the blending of PV into CS, and it was not common since the ratios of different anthocyanin derivatives' precursors to anthocyanins increased (anthocyanins decreased by blending PV into CS,

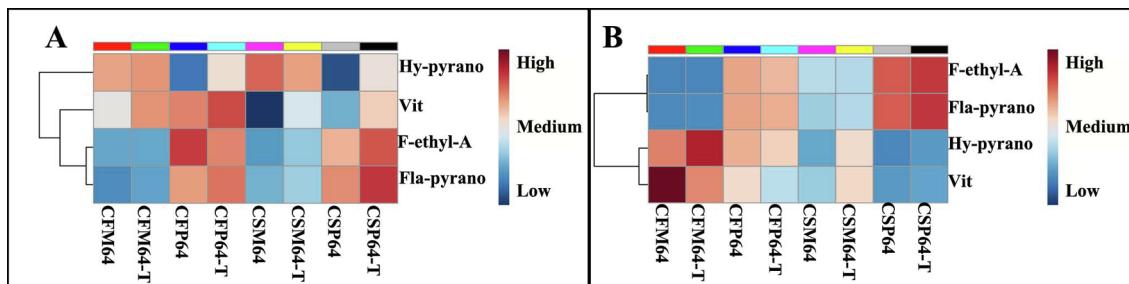


Fig. 4. Comparisons between actual and theoretic (-T) anthocyanin derivatives concentrations of wines made from high proportional blending (40%). F-ethyl-A: flavanol-ethyl-anthocyanin adducts; Fla-pyranosides: flavanyl-pyranoanthocyanins; Hy-pyranosides: hydroxyphenyl-pyranoanthocyanins; Vit: vitisins. Note: samples were collected at the end of limited oxygenation (A) and over-oxidation (B).

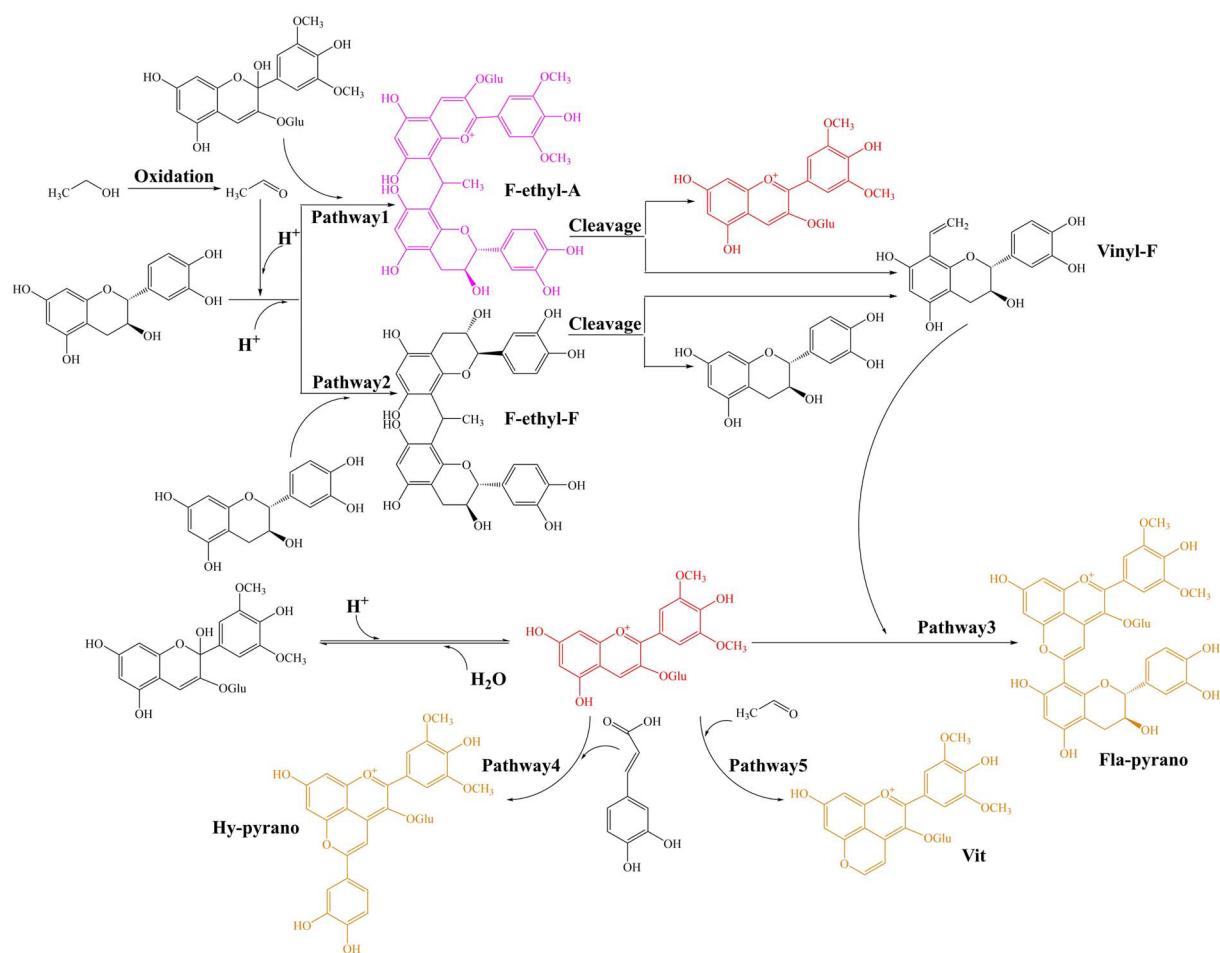


Fig. 5. Examples of pathways for the formation of anthocyanin derived pigments. F-ethyl-A: flavanol-ethyl-anthocyanin adducts; Vinyl-F: vinylflavanol unit(s); Fla-pyranoside: flavanyl-pyrananthocyanins; Hy-pyranoside: hydroxyphenyl-pyrananthocyanins; Vit: vitisins.

flavanols increased from the contribution of PV, and hydroxycinnamic acids would increase due to hydrolysis during the following period). Theoretically, the increased flavanol-anthocyanin ratio could facilitate Pathway 1, Pathway 2 and Pathway 3 in Fig. 5, thus benefit the accumulation of SO₂-resistant polymeric pigments such as flavanol-ethyl-anthocyanin adducts and flavanyl-pyrananthocyanins (Bindon, McCarthy, & Smith, 2014; Fulcrand, Dueñas, Salas, & Cheynier, 2006), meanwhile the increased ratio of hydroxycinnamic acid-anthocyanin ratio should be good for the formation of hydroxyphenyl-pyrananthocyanins (Pathway 4, Fig. 5) (Rentsch, Schwarz, Winterhalter, & Hermosín-Gutiérrez, 2007). However, the comparison of CSP64 wines' actual and theoretic anthocyanin derivatives concentrations showed opposite results that the blend of PV into CS had negative chemical influences on anthocyanin derivatives' formation. Although there are no scientific reports investigating such phenomenon so far, we suppose that the result was a compromise caused by competitions of different anthocyanin derivatives' formation mechanisms, namely: when different groups of anthocyanin derivatives coexist in wines, competitions among different anthocyanin derivatives' formation mechanisms would be intensified with the increased ratios of their precursors to anthocyanins. With very limited anthocyanins and abundant precursors of different anthocyanin derivatives in wines, formation of each anthocyanin derivative might contrary to what we usually expect, and depressed due to interferences of formation mechanisms of other anthocyanin derivatives.

Under over-oxidation condition, for the blend of MA into CF, the reaction potentials for anthocyanin derivatives' formations might be generally depressed due to the decrease of anthocyanin derivatives'

precursors (either by dilution or over-oxidation), and due to the increase of copigmented anthocyanins, except the formation of vitisins (Pathway 5 in Fig. 5). The reason might be acetaldehyde that produced from ethanol oxidation could benefit the formation of vitisin B, which were observed in some wines treated with micro-oxygenation (Cejudo-Bastante, Hermosín-Gutiérrez, & Pérez-Coello, 2011; Pérez-Magariño, Sánchez-Iglesias, Ortega-Heras, González-Huerta, & González-Sanjosé, 2007). Besides, it was reported that the conversion of oxygen into suitable oxidants could promote the formation of vitisin A, and for dry red wines, the content of vitisin A increased during the first 6 months of maturation in air-tight bottles and declined after 12-month maturation, which might also be explained by the shortage of dissolved oxygen and suitable oxidants (Asenstorfer, Markides, Iland, & Jones, 2003). The over-oxidation condition could provide sufficient suitable oxidants to promote the formation of vitisin A. On the contrary, the blend of PV into CF contributed flavanols and hydroxycinnamic acids, proper ratios between these anthocyanin derivatives' precursors and anthocyanins and the continuous supply of suitable oxidants and acetaldehyde produced under over-oxidation condition may benefit the progress of different pathways in Fig. 5. Different from CF, CS had highest or second highest concentrations in each phenolic group (Table 2), namely, it had abundant phenolic reservoir that could supply successive phenolic reactions during preservation. The blend of MA or PV and over-oxidation treatment might break the equilibrium of different phenolic reactions, due to the increase of precursors of different anthocyanin derivatives, which increased the reaction potential of different pathways in Fig. 5 and intensified their competitions for anthocyanins, thus let different anthocyanin derivatives formed slower than expected.

The phenolic compositional differences of four monovarietal wines may not only determine the chemical influences on anthocyanin derivatives' formation, but also explain the chromatic modification phenomenon that CF based blend wines exhibited more obvious chromatic changes (higher ΔE^*_{ab} values) than their CS counterparts (Table 3, Table 4). Since CF and CS had weak and distinct phenolic profiles, respectively, the former might be more prone to be chromatically modified than the latter when blended with modifier wines.

4. Conclusions

Blending of modifier wines (MA, PV) could improve the color quality of base wines (CF, CS) under different oxygen exposures. MA contributed anthocyanins and flavonols and conferred CF and CS reddish hues of free anthocyanins and copigments, while PV contributed flavanols and conferred CF and CS reddish hues of anthocyanin derived pigments. The chemical influences of different blending strategies on anthocyanin derivatives' formations and chromatic changes perceived by human eye were depending on phenolic profiles of the modifier wines and base wines, and also on the oxygen exposures of wine aging conditions. Considering the oxygen in limited oxygenation and over-oxidation conditions were two extremes for practical situation, color improvement effects could be expected based on this blending strategy in winemaking practice. Our next study will focus on comparing the color evolution over time conferred by different blending strategies.

CRediT authorship contribution statement

Si-Yu Li: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft. **Pei-Ru Zhao:** Validation, Formal analysis, Investigation. **Meng-Qi Ling:** Validation, Investigation. **Meng-Yao Qi:** Validation, Investigation. **García-Estévez Ignacio:** Writing - review & editing. **Escribano-Bailón María Teresa:** Writing - review & editing. **Xin-Jun Chen:** Resources. **Ying Shi:** Supervision, Project administration. **Chang-Qing Duan:** Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declared that there is no conflict of interest.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (Grant No. 31471723), China Agriculture Research System (Grant No. CARS-29 to Chang-Qing Duan), Xinjiang the 13th Five-Year Major Project 'Xinjiang Characteristic Winey Winemaking Technology Integration and Product Development' (Grant No. 2017A01001-3).

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodres.2019.108885>.

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