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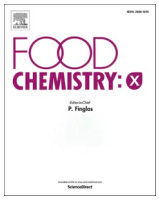

Effects of full shading of clusters from véraison to ripeness on fruit quality and volatile compounds in Cabernet Sauvignon grapes

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ABSTRACT

Sunlight exposure of grape clusters is frequently reported to influence grape aromas greatly. Among them, the effects of full shading (FS) of clusters on fruit quality and volatile compounds in grape berries has scarcely been investigated. In the present study, the effects of FS from véraison to ripeness on fruit quality and volatile compounds in Cabernet Sauvignon grapes were studied. The results showed that FS treatment reduced fruit size and berry weight, delayed fruit maturity, and decreased the contents of anthocyanins, phenols, and tannins in grape berries. In addition, volatile compounds in grape berries were analyzed, and 55 and 53 volatile compounds were detected in the control (CK) and FS groups, respectively. The results indicated that the concentrations of straight-chain fatty aldehydes, straight-chain fatty alcohols, straight-chain fatty acids, and branched-chain fatty acids, norisoprenoids, and total concentration of volatile compounds were all higher in FS group than in CK group. Specifically, FS treatment had significant promoting effects on the concentrations of β-damascenone, terpineol, 2-ethyl-1-hexanol, and 2-hexenal, and remarkably decreased the concentrations of geranial, benzeneacetaldehyde, neral, and ethyl acetate. Partial least squares-discriminant analysis (PLS-DA) revealed a clear separation between the control (CK) and FS groups, and showed that 2-hexenal and hexanal were the main characteristic aroma compounds in the FS group. Moreover, an increase in the intensity of fruity, herbaceous, floral, and mushroom aromas was recorded in FS grapes. This study provides new insights into the effects of the exclusion of sunlight exposure on volatile compound accumulation in grape berries.

Introduction

Volatile compounds in grape berries are important secondary metabolites, where they are found in a wide concentration range of nanograms to milligrams per liter (Kalua & Boss, 2009). The components and concentrations of volatile compounds have important influences on the sensory quality of wine grape and on the flavor and typicality of corresponding wines (Sánchez Palomo et al., 2006). Volatile compounds in grape berries are mainly present in lower epidermal cells of the pericarp and exist in two forms: free and glycosidically bound volatile compounds. Therein, the free volatile compounds can be directly volatilized and present various fragrances, while glycosidically bound volatile compounds are non-volatile (Vilanova et al., 2012). Free volatile compounds play an essential role in grape quality and determine the varietal characteristics thereof.

At present, more than 1000 volatile compounds such as alcohols, esters, terpenes, norisoprenoids, aldehydes, alkenes, and organic acids have been found to emanate from grape berries, and these compounds are mainly categorized into varietal aromas (Tian et al., 2023). The typical varietal aromas of grape berries contribute to the varietal typicality and overall aroma of wines (Cataldo et al., 2021). Based on their biosynthetic pathways, volatile compounds in grape berries fall into three categories: volatile compounds from fatty acid metabolism, amino acid metabolism, and isoprene metabolism (Dudareva et al., 2004) (Supplemental Fig. 1). Volatile compounds from fatty acid metabolism use fatty acids as the precursor to produce hydroperoxides by lipoxygenase pathways, then by oxidation, cleavage, and dehydrogenation, the hydroperoxides are gradually transformed into all kinds of straight-chain fatty aldehydes, straight-chain fatty alcohols, straight-chain fatty acids, straight-chain fatty esters, and straight-chain fatty ketones (Shalit

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et al., 2001). Volatile compounds from amino acid metabolism use amino acids as the precursor to generate various branched-chain alcohols, aldehydes, acids, ketones, and esters, volatile phenols, as well as a variety of aromatics (Torrea et al., 2011). Volatile compounds from isoprene metabolism use isoprenoids as the precursor to synthesize terpenes (e.g., monoterpenes and sesquiterpenes) as well as C9 and C13 norisoprenoids degraded by carotenoids (Muhlemann et al., 2014). Volatile compounds from different synthetic pathways also exhibit different aromatic characteristics, among which volatile compounds from fatty acid metabolism are found to impart herbaceous–green odor notes (Palomo et al., 2007). Volatile compounds from isoprene metabolism are a class of compounds with floral and fruity aromas with a low odor threshold, which contribute to the varietal aromas of grapes (Mendes-Pinto, 2009). Volatile compounds from amino acid metabolism mainly impart pleasant fruity aromas (Torrea et al., 2011).

The biosynthesis and accumulation of volatile compounds in grape berries are influenced by factors such as grape cultivar (Yang et al., 2009), place of origin (Mendez-Costabel et al., 2013; Xu et al., 2015), cultivation and management practices (Buesa et al., 2021; Hernandez-Orte et al., 2015), and climate (Sabon et al., 2002). In recent years, more studies investigated the effects of various light exposure treatments on volatile compounds in grape berries. Generally, sunlight exposure exerts important effect on the synthesis of volatile compounds (Bureau et al., 2000), and photolepsy has greater effect on grape clusters than on whole vines (Ji and Dami, 2008). Grape berries receiving more sunlight contained higher concentrations of terpenes (Bureau et al., 2000; Friedel et al., 2016; Sasaki et al., 2016). Besides, sunlight exposure affected norisoprenoids, and enhanced sunlight caused by leaf removal could enhance norisoprenoid concentrations (Kwasniewski et al., 2010; Feng et al., 2015; Hernandez-Orte et al., 2015; Hickey et al., 2018). However, he et al. (2020) reported that norisoprenoids were reduced after enhanced sunlight caused by leaf removal at véraison. Moreover, cluster shading from fruit setting to harvesting increased the content of free C6 volatile compounds, and the difference was mainly because of delayed fruit maturity after shading treatment (Bureau et al., 2000). In the dry-hot seasons of the Xinjiang region of China, enhanced sunlight caused by leaf removal at véraison markedly increased the concentration of C6 alcohols with no relationship with fruit maturity (He et al., 2020). Obviously, the results were not consistent. Moreover, the full-shading effect of clusters during the ripening process on volatile compounds in grape berries not been individually studied.

In the present study, light-shielding boxes were used on grape clusters in the vineyard to fully shield berries from solar radiation from véraison until harvest, and berry weight, berry diameter, ripening index, the contents of phenolics and volatile compounds were evaluated. The aim of the study was to investigate the effects of full shading of grape clusters from the onset of véraison to ripeness on the fruit quality and volatile compounds in grapes, and we hypothesized that full shading of grape clusters from véraison to ripeness affects berry ripening, phenolic accumulation, and aroma formation in grape berries. These results provide new insights into the effects of avoiding sunlight exposure on volatile compounds in grape clusters and will help viticulturists better understand the response of grape berries to sunlight exposure, enabling them to adjust shading strategies accordingly to meet the demand for preferred fruit quality.

Materials and methods

Experimental materials and design

The field experiment was performed during the 2019 growing season in a commercial vineyard in Jingyang, Shaanxi, China (34°65′N, 108°75′E). Samples used in this study were from five-year-old, own-rooted grapevines (*Vitis vinifera* L. cv. Cabernet Sauvignon) in a north–south orientation, drip irrigated, and trained to the vertical shoot positioned training system with two strong horizontal cordons at 50 cm above ground and spur-pruned with vertically trained shoots. The spacing between rows for grape vines was 0.8 m × 2.5 m. Two treatments, namely, the control check (CK) and the full shading of cluster treatment (FS), were conducted according to the following procedure. Experimental grape clusters were randomly and equally divided into two groups at the véraison stage (when color changes were visible). The CK group consisted of experimental grape clusters in which leaves that covered grape clusters were moved away to allow sunlight exposure. The FS group comprised grape clusters that were placed in light-shielding boxes, designed according to Downey et al., 2004, in which airflow was maintained, while light was excluded, thereby minimizing changes in temperature and humidity (Supplemental Fig. 2). Three biological repeats were set up for each of the two groups. In each repeat, 60 healthy grape clusters had consistent fruit size, growth height, cluster size, and growth period before harvesting and sampling.

Analysis of reducing sugar content and titratable acidity

The reducing sugar content and titratable acidity of grape juice were determined by Fehling reagent titration and sodium hydroxide (NaOH) titration, respectively, following the national standard method (GB/T 15038–2006 Analytical methods of wine and fruit wine). Titratable acidity was measured by titration to pH 8.2 and expressed as the tartaric acid equivalent.

Analysis of phenolics

Extraction of phenolic compounds: Phenolics in grape berries were extracted according to previous reports (Song et al., 2015). In the frozen state, the skins of 150 grape berries were immediately peeled, pulverized into powder, and freeze-dried in a freeze-drying machine for 24 h. Subsequently, 1 g dry powder was weighed in a 50-mL centrifuge tube with 20 mL hydrochloric acid (HCl)–methanol reagent (60 % methanol, 0.1 % HCl). Ultrasonic extraction was performed for 30 min at 30 °C and 40 W, the samples were centrifuged at 10000 rpm for 10 min at 4 °C, and the supernatant was collected. Another 20 mL of HCl–methanol was added to the precipitate, and the above extraction steps were repeated two times. All supernatants of the three repeats were mixed and stored in the refrigerator at − 80 °C for further use.

The total phenolic content was determined by the Folin–Ciocalteu method with some modifications (Meng et al., 2012). To a glass cuvette, 2.9 mL of distilled water, 0.1 mL of phenolic extract, and 0.5 mL of Folin–Ciocalteu reagent were added successively. After extraction for 5 min, 1.5 mL $NaCO_3$ was added. The mixture was allowed to react in the dark for 2 h at room temperature, and the absorbance was determined at 765 nm. A control was prepared by replacing the sample with methanol. Total phenolic content was expressed as milligrams of gallic acid equivalence (GAE) per berry (mg/berry).

Total tannin content was determined by the methyl cellulose method (Sarneckis et al., 2006). In a glass cuvette, 3 mL of methyl cellulose was added to 0.5 mL of phenolic extract. The solution was incubated at room temperature for 3 min, 2 mL of saturated $(NH_4)_2SO_4$ was added, and the mixture was diluted with distilled water to 10 mL and allowed to react for 10 min at room temperature. After centrifugation at 1800 *g* for 5 min, the absorbance of the supernatant was determined at 280 nm. Methyl cellulose was replaced by distilled water in the control. Total tannin content was expressed as milligrams of (+)-catechin equivalence (CE) per berry (mg/berry).

Total anthocyanin content (TAC) was determined by the pH-differential method (Stojanovic and Silva, 2007). First, 0.25 mL of extracted sample was added to two tubes, and the samples were then diluted to 5 mL with KCl buffer at pH 1 and $CH_3CO_2Na{\cdot}3H_2O$ buffer at pH 4.5, respectively. The absorbances of the two mixtures were measured at 520 and 700 nm, respectively, and calculated using the equation $A = (A_{520}–A_{700})\ pH_1 - (A_{520}–A_{700})\ pH_{4.5}$. Each phenolic extract was diluted such that the sample in the buffer at pH 1 had an

absorbance < 1. TAC was expressed as milligrams of malvidin-3-monoglucoside equivalence per berry (mg/berry) and calculated using the equation TAC = (A × MW × DF × Ve × 1000) / (ε × 1 × M), where A is the absorbance, MW is the molecular weight of malvidin-3-glucoside (493.5), DF is the dilution factor, V_e is the extraction volume, ε is the molar extinction coefficient of malvidin-3-glucoside (28,000), and M is the mass of extracted skins.

Analysis of volatile compounds

Extraction method: volatile compounds in grape berries were extracted by headspace (HS) sampling according to previous reports (Xu et al., 2015). Frozen grape samples (100 g) were de-seeded in liquid nitrogen, and the samples were well ground with 1 g of polyvinylpolypyrrolidone (PVPP). The frozen grape powder was transferred to 50-mL centrifuge tubes and kept at 4 °C overnight for cold stabilization and equilibration of volatile compound extraction. Subsequently, the thawed homogenate was centrifuged at 8000 rpm for 15 min at 4 °C, and the supernatant was collected. Thereafter, 5 mL supernatant, 1 g NaCl, and 10 mL of 4-methyl-2-pentanol (internal standard) were blended into the 15-mL sample vial, and the vial was tightly capped with a polytetrafluoroethylene–silicone septum containing a magnetic stirrer. Afterward, the vial containing the sample was equilibrated at 40 °C for 30 min while being stirred on a hot plate. Subsequently, a pretreated (conditioned at 270 °C for 1 h) solid-phase microextraction (SPME) fiber (50/30 μm DVB/CAR/PDMS; Supelco, Bellefonte, PA, USA) was inserted into the headspace for HS-SPME, and the mixture was extracted at 40 °C for 30 min with continuous heating and agitation. Volatile compounds trapped in the fiber were subsequently desorbed by gas chromatography (GC) for 8 min.

Determination method: The separation and identification of the volatile compounds were performed on an Agilent 6890 GC equipped with an Agilent 5975 mass spectrometer (MS). The column was a 60 m × 0.25 mm HP-INNOWAX capillary with 0.25 μm film thickness (J & W Scientific, Folsom, CA, USA). The carrier gas was helium (purity > 99.999 %) at a flow rate of 1 mL/min. The initial oven temperature was 50 °C, held for 1 min, raised to 220 °C at a rate of 3 °C/min, and held at 220 °C for 5 min. The mass spectrometer in the electron impact mode (MS/EI) at 70 eV was scanned in the range of m/z 30 to 350 U. The ion source temperature was 230 °C, and the MS transfer line temperature was 280 °C. The concentrations of volatile compounds and 4-methyl-2-pentanol were analyzed in the selected ion monitoring mode.

Qualitative and quantitative analyses: volatile compounds were identified by matching the retention index with the reference standards in the NIST 11 MS database and aligning the spectra with the reference standards. MSD Chemstation was used for peak integration. The quantitative analysis of volatile compounds was performed by using the calibration curves, which were prepared by using the existing standard compounds. Briefly, the calibration curves of aroma standards were established based on the mixed standard solution which was diluted to 15 levels in succession with the synthetic model matrix. The grape juice synthetic model matrix was prepared in distilled water containing 200 g/L glucose and 7 g/L tartaric acid, and pH was adjusted to 3.3 with 5 M NaOH. The internal standard was 4-methyl-2-pentanol. Volatile compounds without corresponding calibration curves were quantified based on compounds with a similar number of carbon atoms and/or the same functional group.

Data analysis

The data were analyzed by Microsoft Excel 2010 and were presented as the means of triplicate experiments. One-way analysis of variance (ANOVA) with the *t*-test using SPSS software (v.7.5, SPSS Inc., Chicago, IL, USA) was used to determine the differences in the concentrations of volatile compounds among samples at a significance level of 0.05. Heatmap analysis and partial least squares-discriminant analysis (PLS-DA) were performed using MetaboAnalyst (https://www.metaboanalyst.Ca/MetaboAnalyst/faces/home.xhtml). Volcano plot, ridgeline plot, and radar chart analyses were performed using OmicShare tools, a free online platform for data analysis (https://www.omicshare.com/tools).

Results

Effects of FS treatment on physicochemical parameters of grape berries

Cabernet Sauvignon grape berries in the CK and FS groups were harvested on the same day. The reducing sugar content of grape berries in the FS group was 177.57 g/L, significantly lower than 203.50 g/L in the CK group ($P < 0.05$) (Table 1). The titratable acid content of grape berries in the FS group was higher than that in the CK group, while the difference was not significant. Correspondingly, the sugar–acid ratio of grape berries in the FS group was 17.82 % lower than that in the CK group, showing a significant difference ($P < 0.05$). Berry weight and fruit size were also lower in the FS group than in the CK group, with a significant difference in fruit size. Moreover, FS treatment significantly decreased the contents of total phenols, tannins, and anthocyanins in grape berries. This indicates that FS could significantly reduce fruit size, fruit maturity, and phenolics in grape berries.

Analysis of the effect of FS treatment on volatile compounds

Qualitative and quantitative analyses were performed by GC–MS to analyze volatile compounds in grape samples. As shown in Table 2, a total of 57 volatile compounds were detected, and these volatile compounds were divided into eight categories: straight-chain fatty esters, straight-chain fatty aldehydes, straight-chain fatty alcohols, straight-chain fatty acids, terpenes, norisoprenoids, aromatics, and branched-chain fatty acids. Among these, 55 and 53 volatile compounds were identified from grape berries in the CK and FS groups, respectively, and the total concentration of volatile compounds in grape berries in the CK and FS groups was 13,114.92 and 18,199.5 μg/L, respectively, which showed a significant difference. Thus, FS treatment from véraison to ripeness increased the total concentration of volatile compounds in grape berries.

Among all volatile compounds, the concentration of straight-chain fatty esters in grape berries accounted for 10.67 % and 6.18 % of all volatile compounds in the CK and FS groups, respectively (Table 2). Specifically, the straight-chain fatty esters were dominated by ethyl acetate, and its concentration in the CK group was significantly higher than that in the FS group. Moreover, the small amounts of ethyl decanoate, ethyl octanoate, and ethyl dodecanoate showed no significant difference between the CK and FS groups. Therefore, the total concentration of straight-chain fatty esters in the CK group (1399.15 μg/L) was significantly higher than that in the FS group (1124.96 μg/L). Thus, FS treatment could significantly reduce the concentration of straight-chain fatty esters in grape berries (Fig. 1-a).

Table 1
Effect of full-shading treatment on physicochemical parameters of grape berries.

Samples	Berry Weight (g)	Berry diameter (mm)	Reducing sugar (g/L)	Tiratable acid (g/L)	Sugar/TA ratio (g/L)	Total phenolics (mg/berry)	Total tannins (mg/berry)	Total anthocyanins (mg/berry)
CK	1.35 ± 0.01a	14.21 ± 0.09a	203.50 ± 3.87a	5.75 ± 0.36a	36.13 ± 2.45a	2.00 ± 0.11a	1.59 ± 0.01a	0.39 ± 0.00a
FS	1.25 ± 0.07a	12.55 ± 0.08b	177.57 ± 2.10b	5.98 ± 0.34a	29.69 ± 0.90b	1.10 ± 0.04b	0.95 ± 0.06b	0.07 ± 0.00b

Table 2
Effect of full-shading treatment on the concentrations of volatile compounds of grape berries (μg/L). nd: not detected; trace: smaller value can be detected but not quantified; a and b indicate significant differences at $P < 0.05$.

NO.	Volatile Compounds (ug/L)	Retention time (min)	Quantitative standards	Calibration cruves	Aroma descriptor	Treatments	
						CK	FS
	Straight chain fatty esters						
A1	Ethyl acetate	6.263	Ethyl acetate	Y = 1896.30 + 0.00	Fruity	1364.59 ± 11.32a	1090.16 ± 54.02b
A2	Ethyl hexoate	16.181	Ethyl hexoate	Y = 164X + 2.2774	Fruity (Banana, green apple)	4.78 ± 0.15a	4.73 ± 0.06a
A3	Ethyl octanoate	24.500	Ethyl octanoate	Y = 70.328X + 9.5333	Fruity	9.84 ± 0.07a	9.93 ± 0.06a
A4	Isopentyl hexanoate	25.294	Isopentyl hexanoate	Y = 41.43X + 2.3972	Fruity (green apple, pineapple)	2.84 ± 0.01a	2.96 ± 0.16a
A5	Ethyl decanoate	32.350	Ethyl decanoate	Y = 59.057X + 10.595	Fruity	10.67 ± 0.08a	10.75 ± 0.10a
A6	Ethyl dodecanoate	40.295	Ethyl dodecanoate	Y = 82.695X + 6.3497	Fruity	6.43 ± 0.05a	6.44 ± 0.04a
	Total of straight chain fatty esters					1399.15 ± 11.17a	1124.96 ± 54.19b
	Proportion (%)					10.67 %	6.18 %
	Straight chain fatty aldehydes						
A7	Hexanal	10.761	Hexanal	Y = 4099.8X-588.41	Herbaceous, Green	5623.99 ± 80.69b	6609.41 ± 598.57a
A8	2-Hexenal	15.776	2-Hexenal	Y = 7487.2X-1462	Fruity (green apple)	4950.21 ± 142.63b	9052.46 ± 295.17a
A9	Octanal	18.623	Octanal	Y = 307.32X + 0.1127	Fruity	1.47 ± 0.11b	2.23 ± 0.03a
A10	(Z)-2-heptenal	20.090	Octanal	Y = 307.32 + 0.1127	Fruity	0.97 ± 0.04b	1.50 ± 0.48a
A11	nonanal	22.910	nonanal	Y = 111.4X-0.5154	Herbaceous, Green	0.04 ± 0.00	trace
A12	(E,E)-2,4-Hexadienal	23.476	2-Hexenal	Y = 7487.2X-1462	Herbaceous, Green	trace	trace
A13	Decanal	27.250	Decanal	Y = 192.93X + 1.0234	Floral	2.25 ± 0.03b	2.85 ± 0.35a
A14	(E,E)-2,6-nonadienal	30.920	nonanal	Y = 111.4X-0.5154	Floral	6.47 ± 0.26b	9.50 ± 0.90a
	Total of Straight chain fatty aldehydes					10585.4 ± 222.17b	15678 ± 353.02a
	Proportion (%)					80.71 %	86.14 %
	Straight chain fatty alcohols						
A15	1-pentanol	16.650	1-pentanol	Y = 484.82X + 5.8745	Herbaceous, Green	10.72 ± 0.26a	8.95 ± 0.90b
A16	2-heptanol	19.580	2-heptanol	Y = 398.58X-0.7589	Fruity	5.24 ± 0.41a	5.45 ± 0.71a
A17	1-hexanol	20.969	1-hexanol	Y = 1920.7X-11.009	Herbaceous, Green	408.56 ± 13.92b	535.03 ± 25.56a
A18	(E)-3-hexen-1-ol	21.480	E)-3-hexen-1-ol	Y = 4696X-0.6423	Herbaceous, Green	23.39 ± 1.10a	24.37 ± 1.30a
A19	(Z)-3-hexen-1-ol	22.385	(Z)-3-hexen-1-ol	Y = 7514.4x-18.471	Herbaceous, Green	49.00 ± 1.95b	84.63 ± 6.50a
A20	(E)-2-hexen-1-ol	23.282	(E)-2-hexen-1-ol	y = 2286.2x − 362.31	Herbaceous, Green	106.15 ± 18.79b	170.90 ± 20.33a
A21	(Z)-2-hexen-1-ol	23.670	(Z)-2-hexen-1-ol	Y = 2642.1X + 0.7991	Herbaceous, Green	12.19 ± 0.24a	11.75 ± 1.23a
A22	1-octen-3-ol	25.067	1-octen-3-ol	Y = 158.51X-0.0758	Mushroom	0.92 ± 0.08b	1.15 ± 0.09a
A23	1-Heptanol	25.245	1-Heptanol	Y = 568.33X-2.1779	Oily	0.69 ± 0.16b	1.39 ± 0.50a
A24	2-Nonanol	27.822	2-Nonanol	Y = 35.187X + 0.1629	Floral	0.31 ± 0.04a	0.34 ± 0.05a
A25	1-octanol	29.450	1-octanol	Y = 19.885X-0.1515	Floral	trace	trace
A26	(E)-2-octen-1-ol	31.820	(E)-2-octen-1-ol	Y = 904.43x + 6.21	Mushroom	7.38 ± 0.10b	7.85 ± 0.23a
A27	1-decanol	37.782	1-decanol	Y = 706.34X-0.03	Floral	1.05 ± 0.04b	2.64 ± 0.88a
A28	1-Dodecanol	44.940	1-Dodecanol	Y = 51.536X-0.2945	Floral	trace	trace
	Total of straight chain fatty alcohols					625.6 ± 35.22b	854.45 ± 54.22a
	Proportion (%)					4.77 %	4.69 %
	Straight chain fatty acids						
A29	hexanoic acid	40.647	Hexanoic acid	Y = 5776.2X + 37.535	Fatty, Cheese	120.85 ± 3.51b	143.39 ± 18.89a
	Total straight chain fatty acids					120.85 ± 3.51b	143.39 ± 18.89a

(*continued on next page*)

Table 2 (*continued*)

NO.	Volatile Compounds (ug/L)	Retention time (min)	Quantitative standards	Calibration cruves	Aroma descriptor	**Treatments**	
						CK	**FS**
	Proportion (%)					0.92 %	0.79 %
	Terpenes						
A30	limonene	14.374	limonene	Y = 68.34X + 0.3588	Fruity (lemon)	1.48 ± 0.05a	1.54 ± 0.04a
A31	eucalyptol	15.010	Terpinolene	Y = 97.938X + 1.5606	Oily (eucalyptus oil)	44.95 ± 1.49b	55.26 ± 1.88a
A32	p-Cymene	17.774	p-Cymene	Y = 22.722X + 1.5742	–	1.66 ± 0.01b	1.70 ± 0.02a
A33	*trans*-furan linalool oxide	25.594	*trans*-furan linalool oxide	Y = 4194.4X-0.3178	Floral	15.11 ± 0.13a	17.27 ± 2.69a
A34	linalool	28.827	linalool	Y = 34.877X + 0.2682	Floral	0.34 ± 0.02a	0.38 ± 0.03a
A35	4-terpinenol	31.480	4-terpinenol	Y = 49.475x + 0.3501	Floral (cloves)	nd	0.44 ± 0.02
A36	neral	34.502	neral	Y = 1355.1X + 9.2243	Fruity	10.19 ± 0.16	nd
A37	geranial	36.743	geranial	Y = 1355.1X + 9.2243	Fruity	9.86 ± 0.08	nd
A38	β-citronellol	37.787	β-citronellol	Y = 104.14X + 0.4498	Floral (lemon)	0.61 ± 0.01a	0.74 ± 0.08a
A39	Geraniol	40.462	Geraniol	Y = 183.99X + 8.5313	Floral (rose)	8.71 ± 0.06a	8.83 ± 0.11a
	Total terpenes					92.91 ± 1.35a	86.16 ± 4.69b
	Proportion (%)					0.71 %	0.47 %
	Norisoprenoids						
A40	6-methyl-5-hepten-2-one	20.572	6-methyl-5-hepten-2-one	Y = 365.4x-2.1	Fruity	trace	trace
A41	β-Damascenone	39.787	β-Damascenone	Y = 841.29X + 2.8361	Floral	3.58 ± 0.04b	10.39 ± 0.21a
A42	β-ionone	43.841	β-ionone	Y = 5.2266X + 0.3881	Fruity	0.38 ± 0.01a	0.39 ± 0.00a
	Total norisoprenoids					3.96 ± 0.04b	10.78 ± 0.21a
	Proportion (%)					0.03 %	0.06 %
	Aromatics						
A43	toluene	9.516	styrene	Y = 78.645X + 6.1639	–	29.86 ± 1.39a	28.62 ± 1.14a
A44	styrene	17.414	styrene	Y = 78.645X + 6.1639	–	6.41 ± 0.02b	6.47 ± 0.02a
A45	Benzaldehyde	28.620	Benzaldehyde	Y = 612.08X-4.86	Fruity	trace	trace
A46	Benzeneacetaldehyde	33.471	Benzeneacetaldehyde	Y = 2409.4X + 8.8236	Fruity	11.72 ± 0.16	nd
A47	Methyl salicylate	38.369	Methyl salicylate	Y = 81.403X + 8.1582	Oily (wintergreen oil)	8.21 ± 0.24a	8.27 ± 0.04a
A48	2-phenylethyl acetate	39.682	2-phenylethyl acetate	Y = 31.268X + 8.2684	Floral	trace	nd
A49	Benzyl alcohol	41.822	Benzyl alcohol	Y = 10331X-17.246	Floral	20.80 ± 1.51b	32.34 ± 5.48a
A50	Phenylethyl alcohol	43.021	Phenylethyl	Y = 3080.3X + 165.29	Floral (rose)	180.52 ± 3.35a	179.80 ± 2.09a
A51	Phenol	46.306	Phenol	Y = 1922X + 0.095	–	0.75 ± 0.03b	1.03 ± 0.23a
A52	phenol, 2,4-bis(1,1-dimethylethyl)-	55.470	phenol, 2,4-bis(1,1-dimethylethyl)-	—	–	trace	trace
	Total of aromatics					258.27 ± 5.19a	256.53 ± 8.77a
	Proportion (%)					1.97 %	1.41 %
	Branch chain fatty acids						
A53	2,6-dimethyl-4-heptanone	13.855	2,6-dimethyl-4-heptanone	Y = 35.187X + 0.1629	Oily, Fruity	5.91 ± 0.40a	6.13 ± 0.93a
A54	Methyl-1-pentanol	19.093	Methyl-1-Pentanol	Y = 2468.9X + 0.0321	–	11.17 ± 0.32b	17.42 ± 1.76a
A55	2,6-dimethyl-4-heptanol	20.365	2,6-dimethyl-4-heptanol	Y = 35.187X + 0.1629	Floral	0.32 ± 0.01a	0.34 ± 0.01a
A56	2-ethyl-1-hexanol	26.700	2-ethyl-1-hexanol	Y = 50.58X-0.824	Fruity	trace	0.40 ± 0.05
A57	(S)-3-Ethyl-4-methylpentanol	27.530	Methyl-1-Pentanol	Y = 2468.9X + 0.0321	Floral	11.38 ± 2.97b	20.94 ± 1.46a
	Total of branch chain fatty acids					28.78 ± 3.69b	45.23 ± 2.95a
	Proportion (%)					0.22 %	0.25 %
	Total of all aroma compounds					13114.92 ± 259.77b	18199.5 ± 228.74a

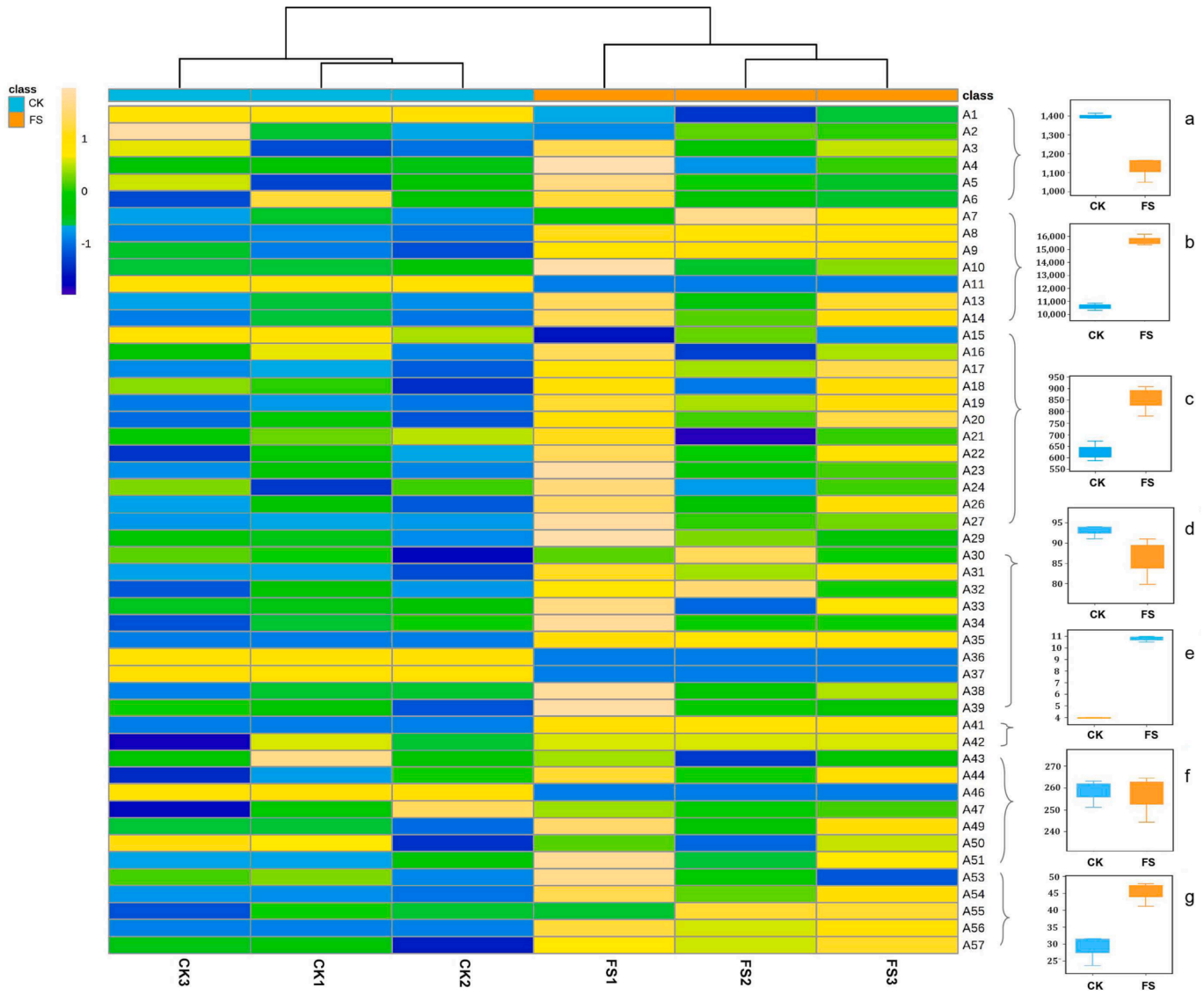

Fig. 1. The heatmap and boxplots of the concentrations of volatile compounds between CK and FS treatments. Fig. a-g are the boxplots for straight-chain fatty esters (a), straight-chain fatty aldehydes (b), straight-chain fatty alcohols (c), terpenes(d), norisoprenoids (e), aromatics (f), branch chain fatty acids (g), respectively.

Specifically, analysis of samples showed that straight-chain fatty aldehydes had the highest concentration and accounted for 80.71 % and 86.14 % of all volatile compounds in the CK and FS groups, respectively. Therein, hexanal and 2-hexenal had the largest concentrations, and both exhibited significantly higher concentrations in the FS group (Table 2). Other straight-chain fatty aldehydes, such as (E, E)-2,6-nonadienal, decanal, octanal, and (Z)-2-heptanal, which have lower concentrations in grape berries, were also higher in the FS group than in the CK group (Table 2 and Fig. 1). Thus, FS could significantly improve the concentration of straight-chain fatty aldehydes in grape berries (Fig. 1-b).

Moreover, the concentration of straight-chain fatty alcohols in grape berries was only second to that of straight-chain fatty esters, and the proportions of straight-chain fatty alcohols were 4.77 % and 4.69 % of all volatile compounds in the CK and FS groups, respectively, and their concentrations were 625.60 μg/L and 854.45 μg/L, showing a significant difference. 1-Hexanol and (E)-2-hexen-1-ol were the main straight-chain fatty alcohols detected in the samples, and their concentrations in the FS group were significantly higher than those in the CK group. Small amounts of (Z)-3-hexen-1-ol, 1-octen-3-ol, 1-heptanol, (E)-2-hexen-1-ol, and 1-decanol were also detected in samples, and their concentrations were higher in the FS group than in the CK group (Table 2), which suggested that FS promoted the concentration of straight-chain fatty alcohols in grape berries (Fig. 1-c).

On account of their typical characteristics of floral and fruity aromas, terpenes are usually used as an index to distinguish between different grape varieties. In this study, nine and eight terpenes were identified from grape berries in the CK and FS groups, respectively. Among these, eucalyptol had the largest concentration in both groups, and its concentration was higher in the FS group than in the CK group. However, higher concentrations of neral (10.19 μg/L) and geranial (9.86 μg/L) were detected in the CK group, but not detected in the FS group. Therefore, the total terpene concentration in grape berries was larger in the CK group (92.92 μg/L) than in the FS group (86.16 μg/L) (Fig. 1-d). Three norisoprenoids, namely, 6-methyl-5-hepten-2-one, β-damascenone, and β-ionone were detected in the two groups. The β-damascenone concentration was 3.58 μg/L and 10.39 μg/L in the CK and FS groups, respectively, showing a significant difference. Besides, only trace amounts of 6-methyl-5-hepten-2-one were detected, and β-ionone was not significantly different between the CK and FS groups. Thus, FS treatment significantly increased the norisoprenoid concentration in grape berries (Fig. 1-e).

For aromatics, ten and eight volatile compounds were identified from grape berries in the CK and FS groups, respectively. Benzeneacetaldehyde and 2-phenylethyl acetate were not detected in the FS group, and phenylethyl alcohol accounted for the largest proportion, but its concentrations showed no significant difference. Compared with the CK group, the FS group showed significantly increased concentrations of benzyl alcohol, styrene, and phenol, but the total aromatic concentration in the CK and FS groups was 258.27 μg/L and 256.53 μg/L, respectively, with no significant difference (Fig. 1-f). One straight-chain fatty acid (hexanoic acid) and five branched-chain fatty acids were detected from grape berries in both groups. Among these, the concentrations of hexanoic acid, (S)-3-thyl-4-methylpentanol, methyl-1-pentanol, and 2-ethyl-1-hexanol in the FS group were significantly higher than those in the CK group. In particular, the concentration of (S)-3-thyl-4-methylpentanol with the highest proportion in branched-chain fatty acids was 57.43 % higher in the FS group than in the CK group. Thus, FS treatment may significantly increase the concentrations of straight- and branched-chain fatty acids in grape berries (Table 2 and Fig. 1-g).

Fold-change (FC) analysis of volatile compounds

To prove the effect of FS treatment on the single volatile compound and volatile compound categories, FC values of single volatile compound and volatile compound categories were calculated between the FS and CK groups (Fig. 2). The FC distributions of different single volatile compound are shown in the volcano plot (Fig. 2-a), where each point represents a detected volatile compound. The abscissa of the volcano plot indicates the logarithm to base 2 of the fold-change ($\log_2$(FC) values of single volatile compound in the FS group compared with the CK group. FC value greater than 1 (the positive axis of the abscissa) suggest that FS treatment exerted a positive effect. In contrast, FC value is greater than 0 and less than 1 (the negative axis of the abscissa) suggest that FS treatment had a negative effect. The ordinate of the volcano plot represents the negative logarithm to base 10 of *p*-value ($-\log_{10}(p)$), and the larger the $-\log10(p)$ values, the greater the differences in the concentrations of the corresponding volatile compounds between the two groups. As shown in Fig. 2-a, 40 of 50 volatile compounds were found on the positive axis of the abscissa, suggesting that FS increased the concentrations of these volatile compounds. Moreover, 14 of 40 volatile compounds were in the upper-right-hand corner (where FC is larger than 1 and the *p*-value is less than 0.05), indicating that FS treatment significantly increased the concentrations of these 14 volatile compounds. Among these, four volatile compounds, namely, β-damascenone (A41), terpineol (A35), 2-ethyl-1-hexanol (A56), and 2-hexenal (A8), showed the largest FC values and the most significant differences under FS treatment. Whereas, 10 volatile compounds were on the negative axis of the abscissa, suggesting that they were negatively influenced by FS, and 4 of 10 volatile compounds, namely, geranial (A37), benzeneacetaldehyde (A46), neral (A36), and ethyl acetate (A1), were found in the upper-left-hand corner (where FC value is greater than 0 and less than 1, the *p*-value is less than 0.05). This indicates that these four volatile compounds were subjected to significant negative effects under FS treatment.

In the ridgeline plot (Fig. 2-b), the abscissa represents the FC of volatile compound categories between the FS and CK groups, while the ordinate indicates the different categories of volatile compounds. Except for straight-chain fatty esters, terpenes, and aromatics, for straight-chain fatty acids, straight-chain fatty alcohols, branched-chain fatty acids, straight-chain fatty aldehydes, and norisoprenoids, the FC values between the FS and CK groups were all greater than 1. Among these, norisoprenoids had the largest FC values. Thus, combined with the results in Table 2, FS treatment exerted the most significant effects on norisoprenoids mainly by increasing the β-damascenone concentration.

PLS-DA of volatile compounds

The contribution of single volatile compound in grape berries to the different experimental groups was established by PLS-DA (Fig. 3). As shown in the scores plot (Fig. 3-a), the first principal component explains 95.5 % of total variation, while the second principal component explains 4.5 % of total variation. The samples of the FS group were located on the positive axis of the abscissa, while those of the CK group were found on the negative axis. Each repeat in the corresponding CK or FS group could be clustered into one class and separated from other experimental groups, indicating that volatile compounds in the FS and CK groups had visible and obvious differences. To determine volatile compounds that contribute to the separation results of the model, a variable importance in projection (VIP) score plot of the PLS-DA model was drawn (Fig. 3-b), and the VIP score represented the contribution of each volatile compound to each experimental group. The larger the VIP score, the greater the contribution of the volatile compound to sample

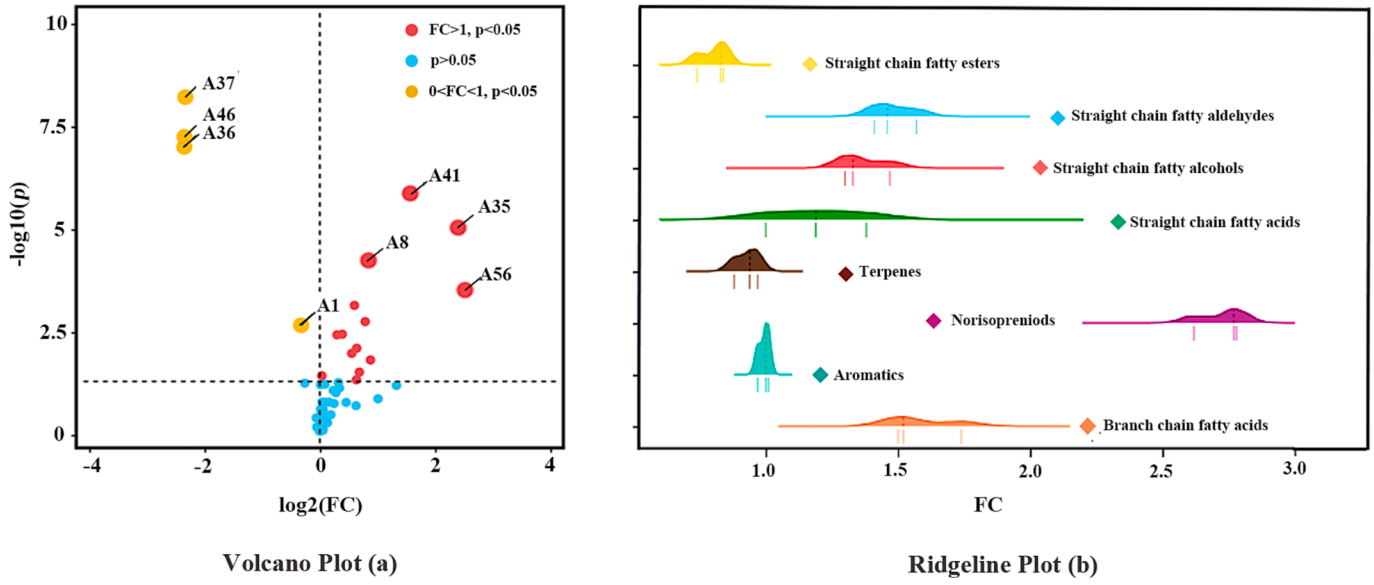

Fig. 2. Analysis of FC values of single volatile compound (a: the volcano plot) and different volatile compound categories (b: the ridgeline plot) between CK and FS treatments.

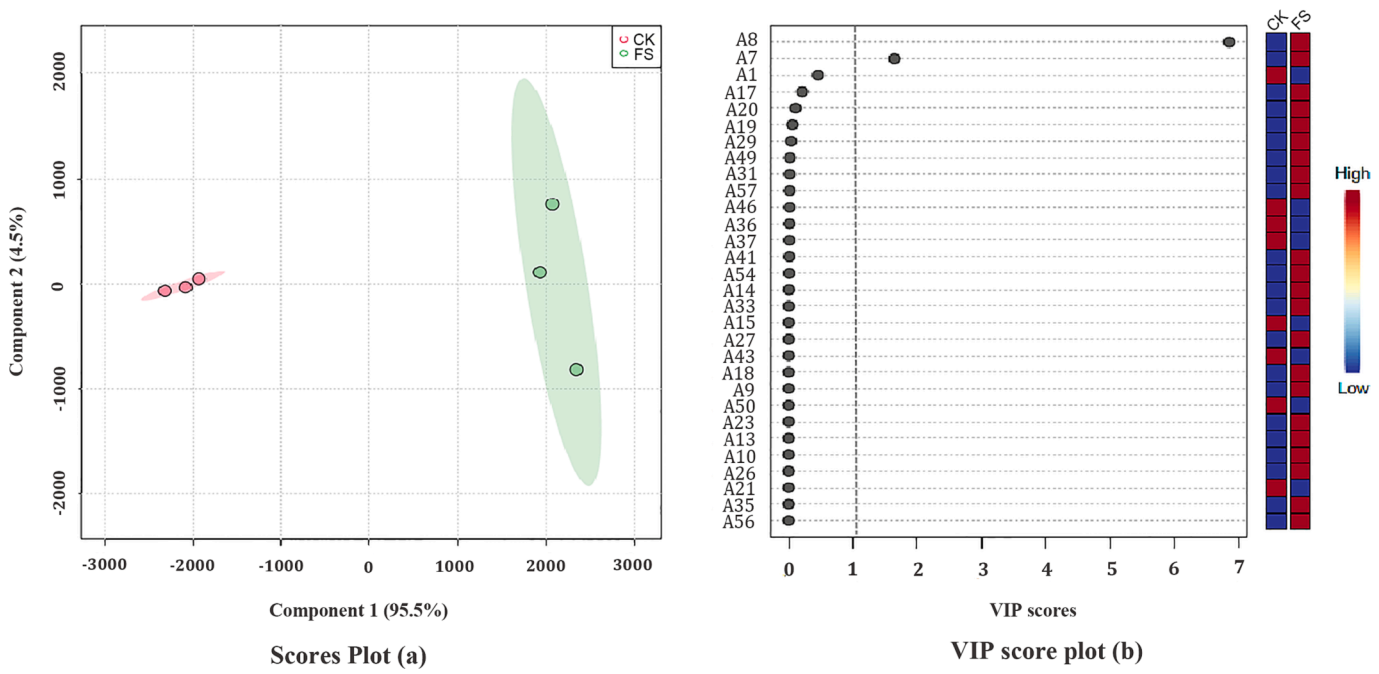

Fig. 3. The PLS-DA analysis of different volatile compounds between CK and FS treatments. (a): the scores plot; (b): the VIP score plot.

classification. In this PLS-DA model, the variables with VIP > 1 were selected as markers, and two volatile compounds were identified, namely, 2-hexenal (A8) and hexanal (A7), which greatly contributed to FS treatment. Thus, 2-hexenal and hexanal, were the most important characteristic aroma compounds in the FS group, effectively enabling discrimination between the FS and CK groups.

Effects of FS treatments on aromatic characteristics in grape berries

Volatile compounds from different synthetic pathways show different aromatic characteristics, and the aromatic characteristics identified from the samples of the two groups were divided into five categories: fruity, floral, herbaceous–green, oily–fatty, and mushroom

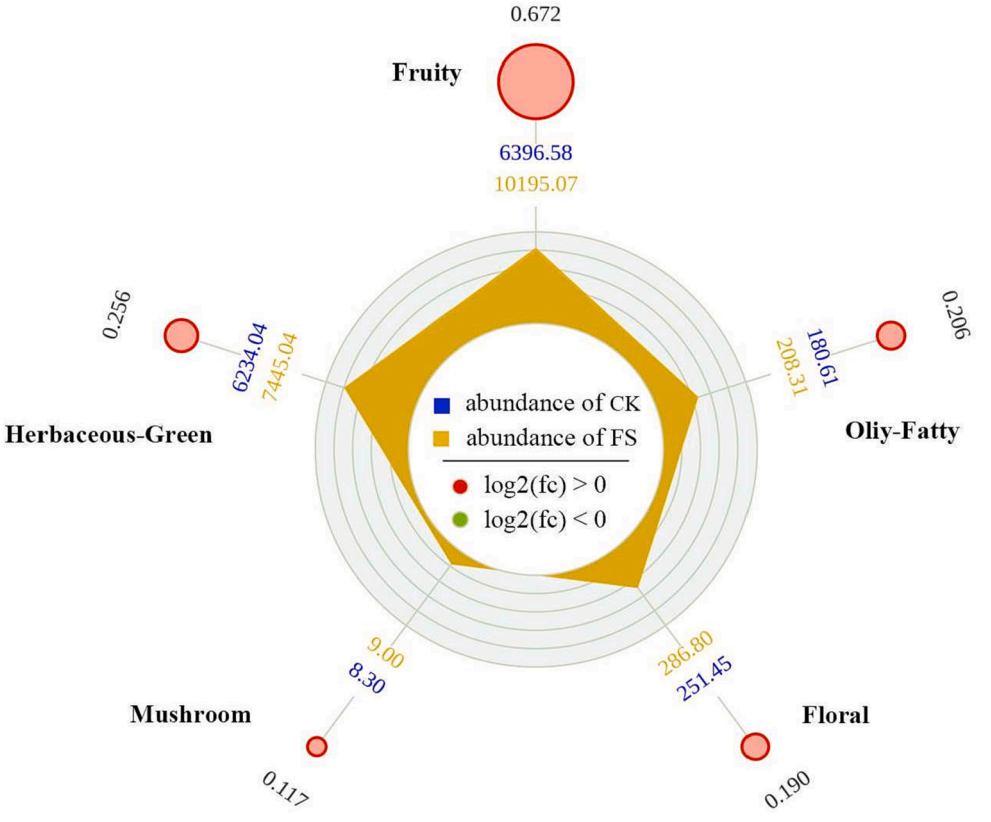

Fig. 4. Comparative analysis of the aromatic characteristics between CK and FS treatments.

aromas (Table 2). Specifically, some volatile compounds, including various straight-chain fatty esters, 2-hexenal, octanal, and (Z)-2-heptanal in straight-chain fatty aldehydes, 2-heptanol in straight-chain fatty alcohols, limonene, neral, and geranial in terpenes, 6-methyl-5-heptene-2-ketone and β-ionone in norisoprenoids, and benzaldehyde and benzeneacetaldehyde in aromatics, mainly release fruity aroma. Volatile compounds including decanal and (E,E)-2,6-nonadienal in straight-chain fatty aldehydes, nonanol, 1-octanol, 1-decanol, and 1-dodecanol in straight-chain fatty alcohols, *trans*-furan linalool oxide, terpineol, linalool, β-citronellol, and geranial in terpenes, β-damascenone in norisoprenoids, and benzyl and phenethyl alcohols and 2-phenylethyl acetate in aromatics mainly impart floral aroma. Hexanal, nonanal, and (E,E)-2,4-hexadienal in straight-chain fatty aldehydes and 1-hexanol, 3-hexen-1-ol, and 2-hexen-1-ol in straight-chain fatty alcohols release herbaceous–green aroma. Moreover, 1-octen-3-ol and (E)-2-octen-1-ol in straight-chain fatty alcohols mainly contribute to mushroom aroma. Although FS reduced the concentration of straight-chain fatty esters with fruity aroma, it apparently improved the concentrations of 2-hexenal with fruity aroma; hexanal, 1-hexanol, (Z)-3-hexen-1-ol, and (E)-2-hexen-1-ol with herbaceous–green aroma; and β-damascenone, ethyl decanoate, (E,E)-2,6-nonadienal, benzyl alcohol, and (S)-3-ethyl-4-methyl-1-pentanol with floral aroma; and 1-octen-3-ol with mushroom aroma in grape berries (Table 2 and Fig. 1). Therefore, FS treatment could impart stronger intensities of fruity, herbaceous–green, floral, and mushroom aromas to grape berries (Fig. 4).

Discussion

Although recent studies have reported the influence of artificial shading (e.g., cluster bagging) and natural shading (e.g., clusters shaded by leaves) on fruit development, sugar and acid metabolism, and the accumulation of flavonoids and volatile compounds in grape berries, the effects of FS treatment from véraison to ripeness on volatile compounds in grape berries have not yet been investigated in detail. In the present study, light-shielding boxes were used to prevent sunlight on grape clusters from véraison to ripeness, and the effects of FS treatment on fruit quality and volatile compounds were investigated in grape berries. Consistent with our study, Guérios et al. (2021) reported that FS had slight influence on cluster zone temperature, but significantly reduced sugar accumulation and organic acid metabolism, and delayed fruit maturation. In grape berries, anthocyanidins are synthesized from the véraison stage, and low-light intensity limits anthocyanidin accumulation (Ristic et al., 2007). In this study, FS treatment from initial véraison to ripeness decreased the contents of total phenols, tannins, and anthocyanins, which was in agreement with a previous study (Riesterer-Loper et al., 2019), and this was possibly associated with clusters not receiving sunlight in the FS group.

Volatile compounds, are important flavor compounds to measure the quality of table and wine grapes. Terpenes, such as linalool, terpinenol, citronellol, and geranial, significantly contributed to fruity and floral aromas with a low odor threshold, and are usually used as characteristic compounds for distinguishing grape varieties and varietal aromas of wines. Grape berries receiving more sunlight contained higher concentrations of terpenes including geranial, linalool, and neral (Bureau et al., 2000; Skinkis et al., 2010), and leaf removal promoted terpenoid accumulation because of increased sunlight exposure (Hernandez-Orte et al., 2015; Young et al., 2016). Besides, terpenoid biosynthesis, especially linalool, was dependent on light intensity and quality (Sasaki et al., 2016). Sunlight exclusion could significantly reduce the expression levels of genes involved in terpenoid metabolism, which could be elevated by re-exposure (Friedel et al., 2016). In this study, sunlight intensity on grape berries was lower in the FS group than in the CK group; thus, the concentration of terpenes, such as neral and geranial, in grape berries was lower in the FS group than in the CK group. As terpene accumulation depended on light exposure, and FS can inhibit accumulation of terpenes in grape berries.

Norisoprenoids, the main components with floral and fruity aromas, are one of the main contributors to aromas of non-aromatic grape varieties, such as Cabernet Sauvignon, Merlot, Syrah, and Chardonnay. In these varieties, the norisoprenoid content is higher than the threshold. In this study, FS treatment from véraison to ripeness can significantly improve the β-damascenone concentration compared with CK, and the FC is obviously large, which reflected the promotion of light exposure is go against norisoprenoid biosynthesis, including β-damascenone accumulation. Carotenoids are considered precursors of C13 norisoprenoids, and carotenoid synthesis starts during the initial stages of berry development and continues until véraison, and the total carotenoid content decreases during véraison and ripening (Young et al., 2012). Grapes exposed to sunlight during ripening showed a significant decrease in carotenoids compared with grapes under shade conditions (Bureau et al., 2000; Razungles et al., 1996). Thus, the higher level of β-damascenone under FS treatment in our study may be related to the availability of more carotenoids resulting from post-véraison cluster sunlight exposure that accelerates carotenoid degradation. In agreement with the current results, some studies have reported a decrease in β-damascenone content in response to cluster exposure. Kwasniewski et al. (2010) found that leaf removal at 68 days after berry set decreased the β-damascenone concentration. Lee et al. (2007) reported that norisoprenoid accumulation, especially β-damascenone, was enhanced when no leaf removal occurred. In addition, He et al. (2020) reported that increased sunlight exposure of fruits by leaf removal and leaf moving resulted in a decline the β-damascenone content. However, there are also some studies reported that exclusion of sunlight from grape clusters at the berry set or prior to flowering significantly decreased the β-damascenone content of grape berries (Bureau et al., 2000; Ristic et al., 2007), and other critical variables besides light (e.g., temperature) might be involved in the regulation of β-damascenone biosynthesis (Wang et al., 2020). In this study, the exclusion of sunlight exposure by shading boxes did not cause a detectable change in cluster zone temperature, aggravated sunshine on berry cluster during véraison to ripening could be the main factor to affect β-damascenone concentration, thus the effect of shading treatment on norisoprenoid concentration was highly dependent on timing and severity of cluster shading. Considering these points, the underlying mechanism of the influence of full shading treatment on β-damascenone biosynthesis and whether it has a relationship with carotenoids require further investigation.

C6 alcohols and C6 aldehydes are important metabolites generated by fatty acid metabolism and crucial flavor compounds in Cabernet Sauvignon, Cabernet Franc, and Merlot grapes (Kalua & Boss, 2009), and changes in C6 volatile compounds can be used a basis for determining grape maturity. In this study, C6 alcohols in grape berries mainly included 1-hexanol and (E)-2-hexen-1-ol, while C6 aldehydes included hexanal and 2-hexenal, also called ‘green leaf volatiles’ (GLVs), which produce the characteristic ‘green’ aroma. Shading treatment of clusters from fruit setting to harvesting increased the content of free C6 volatile compounds, such as hexanal and *trans*-2-hexenal, and differences were mainly because of the lower fruit maturity of fruits after shading treatment (Bureau et al., 2010). In this study, the concentrations of C6 aldehydes and alcohols such as hexanal, 2-hexenal, 1-hexanol, (E)-2-hexen-1-ol, and (Z)-3-hexen-1-ol were higher in the FS group than in the CK group, indicating that full shading of clusters improved aldehyde and alcohol accumulation in grape berries. Thus, our results agreed with previous results, and higher concentrations of C6 aldehydes and alcohols might be the consequence of lower maturity of FS grapes. In plants, aldehydes react with alcohol dehydrogenase to produce corresponding alcohols, which then transfer acyl radicals in acyl-CoA to the substrate of alcohols under the action of alcohol acyltransferase (AAT), thus forming esters. Therefore, AAT is a key enzyme for regulating ester biosynthesis (Qian et al., 2019). In our study, the concentrations of straight-chain fatty alcohols and aldehydes were significantly increased under FS treatment, while the concentration of straight-chain fatty esters was reduced; thus, FS possibly inhibited the transformation of straight-chain

fatty alcohols or acids to straight-chain fatty esters. However, whether the effect of FS on the decreased ester concentration in grape berries is related to the inhibition of AAT activity warrants further research.

In this study, the total concentrations of volatile compounds and different categories of volatile compounds in grape berries were significantly higher in the FS group compared to the CK group. Moreover, two straight-chain fatty aldehydes—2-hexenal and hexanal— were the most important characteristic aroma compounds in the FS group. These two compounds mainly impart grape berries with apple-like fruity aroma and herbaceous–green aroma, respectively. Besides, grape berries subjected to FS treatment from véraison to ripeness contained significantly higher concentrations of β-damascenone, ethyl decanoate, and (E, E)-2,6-nonadienal with floral aroma, and hexanoic acid with fatty aroma. Our results revealed the effect of full shading of grape clusters on the synthesis of volatile compounds in grape berries, while also providing grape growers with valuable information to properly optimize vineyard management strategies and produce high-quality grape berries. In hot or rainy regions, growers often use shading or bagging techniques to protect grapes from adverse weather conditions, thereby enhancing their quality. In this case, with the demand for aroma quality, full-shading of grape clusters from véraison to ripeness can be considered.

Conclusions

Full shading of clusters from véraison to ripeness decreased fruit size, reduced sugar content, delayed fruit maturity, and inhibited the accumulation of anthocyanins, phenols, and tannins in grape berries. The concentrations of straight-chain fatty esters, straight-chain fatty alcohols, straight- and branched-chain fatty acids, straight-chain fatty aldehydes, terpenes, aromatics, and norisoprenoids were all increased by full shading treatment. Specifically, β-damascenone, terpineol, 2-ethyl-1-hexanol, and 2-hexenal with floral and fruity aromas were significantly increased by full shading treatment, whereas geranial, benzeneacetaldehyde, neral, and ethyl acetate were significantly decreased by full shading treatment. Moreover, 2-hexenal with apple-like fruity aroma and hexanal with herbaceous–green aroma were the main characteristic compounds that could be used to distinguish the full shading grapes from the control group. In conclusion, full shading of clusters was conducive to improving the concentrations of some volatile compounds, and imparted stronger intensities of fruity, herbaceous–green, and floral aromas in grape berries. The future research will investigate the molecular mechanism underlying the effects of the exclusion of sunlight exposure on volatile compound biosynthesis, especially focusing on β-damascenone in grapes.

CRediT authorship contribution statement

Meiying Liu: Data curation, Formal analysis, Investigation, Writing – original draft, Methodology. **Hongliang Ji:** Data curation, Formal analysis, Software. **Qianqian Jiang:** Methodology, Writing – review & editing. **Tongyu Liu:** Data curation, Formal analysis, Writing – original draft. **Hui Cao:** Funding acquisition, Supervision, Writing – review & editing. **Zhenwen Zhang:** Resources, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2024.101232.

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