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Hyperoxygenation and Bottle Storage of Chardonnay White Wines: Effects on Color-Related Phenolics, Volatile Composition, and Sensory Characteristics

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ABSTRACT: The effects of hyperoxygenation on Chardonnay white musts and the influence of subsequent storage on the corresponding wines have been evaluated. Attention was focused on the color characteristics, phenolic and volatile composition, and sensorial analysis, not previously reported in conjunction. On the one hand, the hyperoxygenation treatment provoked a significant decrease in the concentration of virtually all phenolic compounds in musts, young wines, and one-year-stored wines. In addition, a higher resistance to browning was observed in stored wines derived from hyperoxygenated musts. Different storage conditions (light and dark) produced significant differences in the 2-*S*-glutathionylcaftaric acid derivatives amounts. On the other hand, significant differences were observed in the volatile composition of wines due to the hyperoxygenation treatment, such as a decrease in the isoamylic alcohols concentration, acetaldehyde, and β-damascenone, even after storage under different conditions. Finally, Chardonnay white wines derived from hyperoxygenated musts had higher banana odor and lower herbaceous and flowery notes.

KEYWORDS: color, hyperoxygenation, Chardonnay, polyphenols, white wine, volatile compounds, sensorial analysis

INTRODUCTION

The oxidation of white wines is a well-known problem in the winemaking industry that usually develops as browning. The must and wine browning alters the color, aroma, and sensory properties, even after a short time of bottle storage. Enologists traditionally recommended must preventive protection against oxidation, generally with sulfur dioxide, with the aim of avoiding browning during bottle storage. However, strict regulations about sulfur dioxide employment exist in the food industry, due to its toxic and allergenic effects on human health. Nowadays, a widespread technique based on oxygen addition has been applied in wineries, the so-called hyperoxygenation.

Hyperoxygenation is a prefermentative technique characterized by an external oxygen addition to a nonsulfited must until saturation. After an optimum sedimentation, the conventional process of vinification is carried out. Once alcoholic fermentation is completed, a small amount of sulfur dioxide is added to avoid malolactic fermentation development.

Oxygen addition favors the enzymatic oxidation of some precursors of polyphenolic compounds present in the must, which are susceptible to oxidation, thus giving rise to their transformation into brown and oxidized polymers of high molecular weight. After precipitation of the flavonoid phenols responsible for bitterness, astringency, and browning during wine aging,[1] they might be removed from the must before alcoholic fermentation due to their high solubilization in alcohol. Although virtually all phenolic compound concentrations decrease as a consequence of oxygen addition, the hydroxycinnamic acid derivatives present in white wines were the main polyphenolic compounds that take part in browning, particularly the major caftaric acid and *p*-coutaric acid.[2,3] As a result, the hyperoxygenation technique produces white wines that are lighter, more stable in sensory parameters, and more resistant to browning than those produced by conventional technology.[4−7]

With regard to the effect of hyperoxygenation treatment on white wine aroma, it greatly depends on variety, must composition, and quantity of oxygen. On the one hand, Singleton et al.[4] and later Dubourdieu et al.[5] affirmed that wines derived from hyperoxygenated musts were characterized by a lack of varietal aroma and suffered a decrease of their aromatic intensity, contrarily to the studies of several authors in Grenache and Chardonnay wines.[8] C_6 alcohols concentrations decreased as a consequence of hyperoxygenation technique applied to Semillon musts, and no significant differences were observed in terpene compound concentrations.[5] According to Cheynier et al.,[1] oxygen addition not only preserved the aroma profile but also increased its quality in Chardonnay, Moscatel of Alejandría, and Penedés white wines (Macabeo and Parrellada). In this way, taking into account as hyperoxygenation conditions the completion of the oxygen consumption for all musts, an increase in the concentration of acetates of large-chain alcohols, fatty acids and their esters, and free terpenes was observed by Artajona et al.[9] in Parrellada, Muscat, and Chardonnay white wines. The same results were obtained by Schneider[10] in other geographic zones (France and Germany) and varieties (Faberrebe). From a sensorial point of

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view, an increase of lemon aroma and a diminution of peach aroma were observed in Riesling wines derived from hyperoxygenated musts.[11] Moreover, the vegetal aroma increased in French white wines,[12] contrary to the findings of Nicolini[13] in Sauvignon blanc wines.

Several changes in wine color and aroma profile were produced during bottle storage in white wines. On the one hand, with regard to the phenolic compounds, significant losses (35−50%) in flavan-3-ol concentration after a year of storage in Albillo white wines were reported by Pérez-Magariño et al.[14] Different opinions about the evolution after storage in hydroxycinnamic acid derivatives have been reported. A decrease in the concentration of the esters of tartaric acid (caftaric, *p*-coutaric, and fertaric acids) and an increase in their respective acids were observed in several white wines,[15] contrary to the results obtained by Mayén et al.[16] in Pedro Ximenez and Baladi white wines. Moreover, the glycoside flavonols were more unstable than aglycon flavonols, which increased during storage,[17] probably due to the chemical hydrolysis of glycosylated flavonol. With regard to the color changes, the storage period produced an increase of the chroma (C^*_{ab}) and a decrease of the hue (h_{ab}), according to Recamales et al.[15] and Hernanz et al.[18] in Zalema and Colombard white wines. Moreover, a year-stored white wine changed its color from pale yellow to yellow-brown, due to the sharp increase of the a^* and b^* values. On the other hand, the storage conditions determined the phenolic content and color of final wines.

In addition, changes in the aroma profile can occur due to the appearance of some volatile compounds that could produce the decline in the aroma quality,[19] which could be accelerated by light and temperature.[20] In this way, Cheynier et al.[21] affirmed that illumination and temperature produce a degradation of phenolic compounds, according to Recamales et al.,[15] who observed significantly lower concentrations of tyrosol and caftaric acid in Zalema white wines under light exposure storage.

The aim of this research was based on the effects of white must hyperoxygenation on color and phenolic compounds, volatile composition, and sensory characteristics of the resulting wines. The study was performed on a Chardonnay white wine, and our interest was focused on several perspectives not previously considered in conjunction. Also, to reflect commercial storage conditions, we have studied the influence of storage in light and dark conditions on white wines derived from hyperoxygenated musts, which was not previously reported in hyperoxygenation studies.

MATERIALS AND METHODS

Winemaking. Grapes from *Vitis vinifera* var. Chardonnay cultivated in Ciudad Real (region of Castilla-La Mancha, Spain), were harvested at their optimal ripening stage and in good sanitary conditions (pH, 3.19; titratable acidity, 7.60 g/L; °Brix, 23.5). After the grapes were destemmed and crushed in a bladder press (yield of 55 L of must per 100 kg of grapes), the must was homogenized in a stainless steel tank of 1000 L capacity and later was distributed in four stainless steel tanks of 250 L. Two tanks were submitted to hyperoxygenation treatment, and the other two tanks contained untreated, control wine.

Control tanks were submitted to SO_2 addition (100 mg/L, as $K_2S_2O_7$) to avoid possible must oxidation. A silicon diffuser was connected to an oxygen cylinder (purity > 99.9%) and was introduced to the must destined for hyperoxygenation treatment, and later oxygen was pumped from the bottom to the top of the tank. After 4.5 h, oxygen introduced into the Chardonnay must was 50 mg/L, using an oxymeter for flow control (Laffort, Spain). Later, both musts were cold-settled at 4 °C for 48 h, and clean fractions were racked and inoculated with *Saccharomyces cerevisiae* selected yeasts (UCLM S377, Found-Springer, France) for alcoholic fermentation development. The fermentation was controlled by monitoring of density and enzymatic methods of residual sugar (Boehring Mannheim, Germany). All fermentations were conducted with a temperature adjustment of 18 °C. After filtration, both control wine and wine derived from hyperoxygenated must were supplied with 60 mg/L of SO_2 to prevent malolactic fermentation. All fermentations were performed in the experimental winery of Castilla-La Mancha University (Ciudad Real, Spain) and were carried out in duplicate.

After bottling, control wine and wine derived from hyperoxygenated must were stored for 1 year at 15 °C under separate dark and light conditions.

Samples were collected and analyzed from each tank at the start (must), at the end of alcoholic fermentation, and after 1 year of bottle storage, in dark and light conditions.

Wine conventional analytical data were analyzed by OIV International Oenological Codex.[22]

Analysis of Wine Polyphenolic Compounds and Color Parameters. For the analysis of the main phenolic types by spectrophotometry, a Hewlett-Packard 8452A apparatus was used. Total polyphenolic compounds, hydroxycinnamic acid derivatives, and flavonols[23] and flavan-3-ol families[24] were measured. Also, the CIELAB chromatic coordinates (illuminant D65 and 10° observer) (L^*, C^*_{ab}, h_{ab}, a^*, and b^*) were calculated.[25]

Prior to the HPLC method, phenolic compound extracts were obtained by solid-phase extraction (SPE) on reverse-phase cartridges (Sep-pack, 500 mg of adsorbent; Waters). Two milliliters of white wine was passed through the C18 cartridges, previously conditioned with 4 mL of methanol and 4 mL of water. After washing with 2 mL of water to remove soluble compounds (sugars and other low molecular weight polar compounds), the phenolic compounds were eluted with 10 mL of methanol. The eluate was dried in a rotatory evaporator (40 °C) and resolved in 2 mL of the phase A used in the HPLC separation.

HPLC separation, identification, and quantification of phenolic compounds were performed on an Agilent 1100 series system (Agilent, Waldbronn, Germany), equipped with a DAD photodiode detector (G1315B) and a LC-MSD Trap VL (G2445C VL) electrospray ionization mass spectrometry (ESI/MS^n) system, both coupled to an Agilent Chem Station (version B.01.03) for data processing. The samples, after filtration (0.20 μm, polyester membrane, Chromafil PET 20/25, Machery-Nagel, Düren, Germany), were injected (50 μL) in duplicate on a reversed-phase column Zorbax Eclipse XDB-C18 (4.6 × 250 mm; 5 μm particle; Agilent), thermostated at 40 °C. We used the chromatographic method developed by Castillo-Muñoz et al.[26]

Quantification was made using the DAD chromatograms obtained at 320 nm for the hydroxycinnamic acid derivatives, at 280 nm for the flavan-3-ols compounds and benzoic acids, and at 360 nm for flavonols. For identification, ESI-MS^n was used in positive mode for flavan-3-ols, whereas both positive and negative modes were used for flavonols, benzoic acids, and hydroxycinnamic acid derivatives.[26,27]

Analysis of Wine Volatile Compounds. For major volatile compounds, the samples were centrifuged for 30 min at 12000 rpm and 4 °C, then passed through glass wool, spiked with 2-pentanol as internal standard (1 g/L), and directly injected (on split mode) in a Hewlett-Packard 5890 series II gas chromatograph coupled to a flame ionization detector.

Minor volatile compounds of wines were extracted in duplicate by using SPE technique, according to the method proposed by Sánchez-Palomo et al.[28] The extracts were concentrated to 200 μL under a gentle stream of nitrogen and were stored in a freezer (−20 °C) until chromatographic analysis in scan rate. A volume of 1 μL of extracts was injected in splitless mode into an Agilent Technology 6890 N Network GC System equipped with an Agilent Technology 5973 *inert*

Table 1. Conventional Analysis of Chardonnay White Wines (CW, Control Wine; HW, Wine Derived from Hypeoxygenated Must)[a]

	CW	HW
total acidity (g/L)	7.99 c ± 0.11	7.54 b ± 0.02
volatile acidity (g/L)	0.22 ± 0.06	0.23 ± 0.02
pH	3.17 b ± 0.01	3.25 c ± 0.01
relative density	0.9945 ± 0.0074	0.9963 ± 0.0042
free SO_2 (mg/L)	9 ± 0	9 ± 0
total SO_2 (mg/L)	41 ± 3	35 ± 4
alcoholic strength (% vol)	12.7 ± 0.23	13.0 ± 0.05
reducing sugars (g/L)	1.53 ± 0.07	1.62 ± 0.16
fructose (g/L)	<1.20	<1.20
glycerol (g/L)	6.19 c ± 0.18	5.39 b ± 0.17
glucose (g/L)	1.05 ± 0.07	1.25 ± 0.21
tartaric acid (g/L)	4.64 ± 0.06	4.52 ± 0.02
L-malic acid (g/L)	1.51 ± 0.01	1.56 ± 0.01
L-lactic acid (g/L)	0.55 ± 0.07	0.53 ± 0.28

[a] Different letters in the same row denote significant differences according to Student's *t* test ($p < 0.05$) between CW and HW.

Mass Selective Detector. The chromatographic conditions were followed according to the method proposed by Sánchez-Palomo et al.[28]

The identification was based on comparison of the mass spectra with those provided for authentic standards and by the NBS75K and Wiley A libraries. The response factor of each volatile compound was calculated by injection of a commercial standard. For compounds for which commercial standards were not available, the response factors of compounds with similar chemical structures were used. All of the samples were injected in duplicate.

Descriptive Sensorial Analysis. Chardonnay control wine and wine derived from hyperoxygenated must were tested by a panel of assessors (between 12 and 15) with experience in sensorial analysis, between 25 and 45 years old. Discriminative tests afforded assessors training in descriptive sensorial analysis during 15 sessions. Reference standards were used for the descriptors evaluation. Assessment took place in a standard sensory analysis chamber,[29] equipped with separate booths and wine-testing glasses[30] covered with a watch-glass to minimize the escape of volatile compounds. Wines were sniffed and tasted. Then judges generated sensory terms individually. Finally, six olfactive (fresh, green apple, floral, fruity, tropical fruit, and banana), three olfactive−gustative (green apple, fruity, and tropical fruit), and six in-mouth feel attributes (acidity, bitterness, herbaceous, body, intensity, and quality of persistence) were selected by consensus. Also, global impression was valued for each tester.

The panelists used a 10 cm unstructured scale to rate the intensity of each attribute. The left extreme of the scale indicated a null intensity of the descriptor and the right extreme the maximum value. All wine samples were evaluated in duplicate.

Statistical Analysis. Statistical analyses were carried out by using the SPSS version 15.0 for Windows statistical package. Student's *t* test and Student−Newman−Keuls test were applied to discriminate the means of chemical data. Furthermore, a principal component analysis (PCA) was carried out with the aim of highlighting the main contributors to the variance among samples.

RESULTS AND DISCUSSION

General Parameters. Table 1 shows the general composition of Chardonnay wines, control (CW) and derived from hyperoxygenated musts (HW), corresponding to the 2006 vintage.

Table 2. Mean Values of Concentration (Milligrams per Liter) and Standard Deviations ($n = 2$) of Several Types of Polyphenolic Compounds Belonging to Different Chemical Families (Hydroxycinnamic Acid Derivatives (HCAD), Benzoic Acids, Flavan-3-ols, and Flavonols), Identified by HPLC-MS^n, Global Types of Phenolic Families, and Chromatic Characteristics by Spectrophotometric Measures, in Control (CM) and Hyperoxygenated (HM) Chardonnay White Musts[a]

	CM	HM
HCAD		
t-GRP	20.0 c ± 0.62	9.38 b ± 1.03
c-GRP	6.64 ± 0.03	5.33 ± 0.20
t-caftaric acid	4.47 c ± 0.29	1.99 b ± 0.07
t-coutaric acid	1.61 c ± 0.25	1.09 b ± 0.04
c-coutaric acid	2.54 ± 0.51	0.55 ± 0.58
t-fertaric acid	4.45 c ± 0.03	3.41 b ± 0.08
c-fertaric acid	1.08 ± 0.08	0.77 ± 0.16
benzoic acids		
gallic acid	4.35 c ± 0.10	3.33 b ± 0.10
flavan-3-ols		
(+)-catechin	8.16 c ± 0.24	5.73 b ± 0.36
(−)-epicatechin	2.28 c ± 0.08	0.57 b ± 0.24
flavonols		
quercetin-3-glucuronide	3.95 c ± 0.05	2.23 b ± 0.03
quercetin-3-glucoside	3.02 c ± 0.07	1.32 b ± 0.17
kaempferol-3-galactoside	0.67 c ± 0.01	0.44 b ± 0.02
kaempferol-3-glucoside	1.24 c ± 0.01	0.77 b ± 0.02
isorhamnetin-3-glucoside	0.10 ± 0.02	0.07 ± 0.00
quercetin	0.35 c ± 0.04	0.12 b ± 0.04
kaempferol	0.27 c ± 0.02	0.08 b ± 0.04
global families		
total polyphenols	337 c ± 16.1	248 b ± 16.4
HCAD	95.3 c ± 3.64	61.4 b ± 3.34
flavonols	84.7 c ± 4.39	56.8 b ± 4.25
flavan-3-ols	19.9 ± 6.73	20.1 ± 6.27
chromatic characteristics		
L^*	37.7 ± 1.38	31.8 ± 9.69
C^*_{ab}	44.1 ± 1.65	38.9 ± 7.75
h_{ab}	70.8 ± 0.09	68.2 ± 5.56
a^*	14.5 ± 0.48	14.0 ± 0.64
b^*	41.7 ± 1.59	36.1 ± 8.57

[a] Different letters in the same row denote significant differences according to Student's *t* test ($p < 0.05$) between CM and HM. GRP, grape reaction product, 2-*S*-glutathionylcaftaric acid.

Alcoholic fermentation developed correctly as indicated the low values of reducing sugars, fructose and glucose, thus being considered as dry white wines (reducing sugars < 5 g/L). Both CW and HW wines showed optimal pH and volatile acidity values, and the latter value was below the limit established by CEE[31] (1.08 g/L). According to Student's *t* test, the content of glycerol was significantly higher in CW than in HW (Table 1), being positive from a sensorial point of view. That fact is quite logical because the SO_2 added to the control must has been likely combined with acetaldehyde, producing an imbalance in redox equilibrium of the yeast that provokes an increased deviation through the glycerol pyruvic pathway.[32]

Table 3. Mean Values of Concentration (Milligrams per Liter) and Standard Deviations ($n = 2$) of Several Types of Polyphenolic Compounds Belonging to Different Chemical Families (Hydroxycinnamic Acid Derivatives (HCAD), Benzoic Acids, Flavan-3-ols, and Flavonols), Identified by HPLC-MS, Global Types of Phenolic Families, and Chromatic Characteristics by Spectrophotometric Measures in Control Wines (CW) and Wines Derived from Hyperoxygenated (HW) Chardonnay White Musts and after 1 Year of Bottle Storage (-1) under Light and Dark Conditions[a]

	CW	HW	CW-1 light	HW-1 light	CW-1 dark	HW-1 dark
HCAD						
t-GRP	12.6 c ± 0.09	8.59 b ± 0.38	12.4 c ± 0.03	8.67 b ± 0.40	12.0 c ± 0.10	8.60 b ± 0.36
c-GRP	nd b	nd b	5.42 d ± 0.12	4.48 c ± 0.15	5.46 d ± 0.18	4.41 c ± 0.04
t-caftaric acid	8.36 c ± 0.14	2.51 b ± 0.38	10.9 d ± 0.61	3.71 b ± 0.84	10.8 d ± 0.69	3.53 b ± 0.58
t-coutaric acid	4.85 c ± 0.85	1.35 b ± 0.13	5.21 c ± 0.29	2.17 b ± 0.25	4.65 c ± 0.22	1.39 b ± 0.16
c-coutaric acid	3.79 d ± 0.34	1.78 bc ± 0.34	2.11 c ± 0.07	1.23 b ± 0.07	2.35 c ± 0.05	1.30 b ± 0.07
t-fertaric acid	1.20 ± b 0.13	0.87 b ± 0.07	3.88 d ± 0.17	2.93 c ± 0.05	3.55 d ± 0.35	2.90 c ± 0.09
c-fertaric acid	3.91 d ± 0.29	3.10 c ± 0.08	1.28 b ± 0.07	1.11 b ± 0.09	1.57 b ± 0.16	1.16 b ± 0.04
caffeic acid	2.75 b ± 0.05	2.62 b ± 0.02	3.23 c ± 0.03	2.70 b ± 0.04	3.18 c ± 0.05	2.68 b ± 0.04
p-coumaric acid	0.81 d ± 0.01	0.46 b ± 0.04	1.17 e ± 0.02	0.56 c ± 0.03	1.25 f ± 0.01	0.60 c ± 0.02
ferulic acid	1.00 d ± 0.05	0.89 ± 0.01	1.25 e ± 0.02	0.67 b ± 0.04	1.21 e ± 0.02	0.72 b ± 0.05
t-GSCf	nd b	nd b	10.0 d ± 0.05	5.25 c ± 0.37	10.0 d ± 0.15	5.08 c ± 0.04
t-GRP-Et-1	nd b	nd b	6.59 d ± 0.05	4.39 c ± 0.06	6.06 ± 0.65	4.44 c ± 0.11
c-GRP-Et-1	nd b	nd b	3.75 c ± 0.07	4.35 d ± 0.09	4.31 ± 0.09	4.34 ± 0.10
t-GRP-Et-2	nd b	nd b	5.79 d ± 0.04	4.64 cd ± 0.17	4.92 cd ± 1.05	4.24 c ± 0.03
t-GRP-Et-3	nd b	nd b	5.48 e ± 0.01	4.17 cd ± 0.16	4.81 de ± 0.82	3.61 c ± 0.01
t-glu-cys-GRP	nd	nd	nq	nq	nq	nq
benzoic acids						
gallic acid	1.18 ± 0.01	0.83 ± 0.01	1.32 ± 0.07	0.86 ± 0.03	1.12 ± 0.36	0.91 ± 0.02
flavan-3-ols						
(+)-catechin	3.11 d ± 0.21	1.69 c ± 0.02	nq b	nq b	nq b	nq b
(−)-epicatechin	8.71 c ± 0.74	7.92 c ± 0.38	5.90 b ± 0.39	5.27 b ± 0.15	6.15 b ± 0.11	4.96 b ± 0.05
(−)-epicatechin gallate ester	nq	nq	nq	nq	nq	nq
flavonols						
quercetin-3-glucuronide	1.48 c ± 0.16	1.60 c ± 0.16	nd b	nd b	nd b	nd b
kaempferol-3-glucoside	0.42 d ± 0.01	0.30 c ± 0.01	nd b	nd b	nd b	nd b
quercetin	3.24 c ± 0.00	2.00 b ± 0.30	1.87 b ± 0.08	1.73 b ± 0.32	1.81 b ± 0.19	1.43 b ± 0.55
kaempferol	0.87 d ± 0.08	0.70 c ± 0.05	0.36 b ± 0.01	0.42 b ± 0.06	0.34 b ± 0.04	0.37 b ± 0.07
global families						
total polyphenols	281 c ± 15.5	199 b ± 24.1	280 c ± 0.62	206 b ± 22.1	281 c ± 4.66	206 b ± 15.2
HCAD	101 c ± 4.20	57.2 b ± 11.5	104 c ± 2.14	65.1 b ± 10.1	105 c ± 0.16	64.8 b ± 7.59
flavonols	80.0 c ± 5.20	44.2 b ± 12.7	60.2 bc ± 0.45	38.4 b ± 7.96	61.4 bc ± 0.45	37.5 b ± 5.69
flavan-3-ols	21.8 e ± 2.16	13.9 cd ± 0.21	12.3 c ± 0.25	8.68 b ± 0.49	15.6 d ± 0.49	11.1 c ± 0.00
chromatic characteristics						
L^*	96.7 c ± 0.42	94.7 b ± 0.49	96.4 c ± 0.42	97.3 c ± 0.11	96.3 c ± 0.35	97.1 c ± 0.14
C^*_{ab}	10.9 b ± 0.63	11.8 b ± 0.05	14.0 c ± 0.44	10.9 b ± 0.41	14.6 c ± 0.84	11.8 b ± 0.16
h_{ab}	95.5 ± 0.68	94.0 ± 0.42	96.4 ± 1.98	97.9 ± 0.02	96.9 ± 1.79	97.4 ± 0.05
a^*	−1.03 ± 0.07	−0.82 ± 0.09	−1.57 ± 0.52	−1.50 ± 0.06	−1.75 ± 0.55	−1.51 ± 0.01
b^*	10.8 b ± 0.63	11.8 b ± 0.05	13.9 c ± 0.38	10.8 b ± 0.41	14.5 c ± 0.78	11.7 b ± 0.16

[a] Different letters in the same row denote significant differences according to Student−Newman−Keuls test ($p < 0.05$). nd, not detected; nq, not quantificable. GRP, grape reaction product, 2-*S*-glutathionylcaftaric acid; GSCf, 2-*S*-glutathionylcaffeic acid; GRP-Et 1, monoethyl ester of 2-*S*-glutathionylcaftaric acid at the carboxy group of the glycine terminal unit of the glutathionyl moiety; GRP-Et 2 and GRP-Et 3, monoethyl esters of 2-*S*-glutathionylcaftaric acid at the two carboxy groups of the caftaric acid moiety; Glu-cys-GRP, 2-*S*-(γ-glutamylcysteinyl)-*trans*-caftaric acid.

Polyphenolic Compounds Identified in Chardonnay Musts and Wines. In this research, the following types of polyphenolic compounds have been identified in Chardonnay musts and wines: benzoic acids, flavan-3-ols, flavonols, and hydroxycinnamic acid derivatives (Tables 2 and 3). The benzoic acids (gallic acid) and flavan-3-ols ((+)-catechin, (−)-epicatechin, and also (−)-epicatechin 3-gallate) identified were the expected, well-known, compounds usually present in white musts and wines. Among the flavonols, Chardonnay musts mainly contained flavonol 3-glycosides, predominantly derived from the aglycone quercetin (as 3-glucuronide and 3-glucoside) and then kaempferol (as 3-glucoside and 3-galactoside), and traces of isorhamnetin 3-glucoside, together with small amounts of the free aglycones quercetin and kaempferol released by

Table 4. Results of Principal Component (PC) Analysis Applied to the Concentrations of Polyphenol Compounds (Hydroxycinnamic Acid Derivatives, Benzoic Acids, Flavan-3-ols, and Flavonols) Identified in Control and Hyperoxygenated Chardonnay Musts and Wines and after a Year of Bottle Storage in Light and Dark Conditions

PC	% explained variance	% accumulated explained variance	variables more correlated to each PC	loading
1	43.80	43.80	*t*-GRP-Et-1	0.974
			t-GRP-Et-3	0.972
			t-GRP-Et-2	0.969
			t-GSCf	0.950
			c-GRP-Et-1	0.940
			p-coumaric acid	0.822
			quercetin-3-glucuronide	−0.927
			kaempferol-3-glucoside	−0.890
			(+)-catechin	−0.892
2	31.17	74.97	*c*-GRP	0.951
			c-fertaric acid	−0.940
			t-fertaric acid	0.921
			kaempferol	−0.946
			quercetin	−0.885
			(−)-epicatechin	−0.939
3	20.66	95.63	HCAD	0.965
			total polyphenols	0.904
			flavonols	0.863

hydrolysis. The white wines mainly contained the free aglycones quercetin and kaempferol, and only wines at the end of alcoholic fermentation contained small amounts of quercetin-3-glucuronide and kaempferol-3-glucoside.

With regard to hydroxycinnamic acid derivatives, Chardonnay musts contained the *trans* and *cis* isomers of the hydroxycinnamoyltartaric acid derived from caffeic, *p*-coumaric and ferulic acids (caftaric, coutaric, and fertaric acids, respectively), being also present the reaction product of glutathione with oxidized caftaric acid, 2-*S*-glutathionylcaftaric acid (also known as GRP, grape reaction product), formed during must obtaining and processing (Table 2). It is remarkable that GRP was found as both *trans* and *cis* isomers, as previously found by Cejudo-Bastante et al.[33] All of the aforementioned hydroxycinnamic acid derivatives were also found in Chardonnay wines, in addition to the expected hydrolysis products (free caffeic, *p*-coumaric, and ferulic acids) and recently described derivatives of GRP:[33] the hydrolysis products *trans*-2-*S*-glutathionylcaffeic acid (*t*-GSCf) and 2-*S*-(glutamyl)cysteinyl-*trans*-caftaric acid, and four monoethyl esters of GRP (*t*-GRP-Et-1, *c*-GRP-Et-1, *t*-GRP-Et-2, and *t*-GRP-Et-3).

Effects of Must Hyperoxygenation and Wine Bottle Storage on the Phenolic Composition and Color Characteristics of Chardonnay Wines. The hyperoxygenation treatment induced browning in the must, but after cold-settling and raking, the untreated (control must, CM) and treated must (hyperoxygenated must, HM) showed no significantly different color characteristics (Table 2). However, the phenolic composition of HM markedly changed in comparison to CM and was characterized by significantly lower content of almost all kinds of global (total polyphenols, hydroxycinnamic acid derivatives, and flavonols) and individual compounds belonging to these kinds of polyphenolics (Table 2), according to several authors.[3,6]

The wines elaborated from control and treated Chardonnay musts (CW and HW, respectively) maintained the differences in phenolic composition shown by their respective original musts (CM and HM). Moreover, these differences were extended to the new phenolic compound developed in wines, namely, free hydroxycinnamic acids and flavonol aglycones and also to the further formation of GRP derivatives in 1-year bottle-stored wines (Table 3).

PCA was applied to extract useful information from the complex matrix of phenolic compounds data corresponding to control and hyperoxygenated musts (CM and HM), control wines, and wines derived from hyperoxygenated musts (CW and HW), and after subsequent storage under light and dark conditions (CW-1 light, CW-1-dark, HW-1 light, and HW-1 dark). The first three principal components (PCs) explained nearly the total accumulated variance (Table 4). PC-1 and PC-2 mainly allowed the distinction between musts (M), wines just after alcoholic fermentation (W), and wines after 1 year of bottle storage (W-1), and only small differences could be denoted by these two PCs with regard to the treatment of hyperoxygenation (Figure 1A). However, PC-3 clearly differentiated the hyperoxygenation-treated must and the corresponding wines from the nontreated, control, ones (Figure 1B).

Following the time sequence of the groups depicted in Figure 1A, the most relevant changes affecting musts after alcoholic fermentation were the total consumption of *c*-GRP, the decrease of the concentration of *t*-fertaric acid together with the increase of its *cis* isomer, the increase of (−)-epicatechin concentration, and the increase of the content of the flavonol aglycones quercetin and kaempferol caused by the hydrolysis of their 3-glycoside precursors, which decreased in concentration (Tables 2 and 3). Depolymerization of the proanthocyanidin molecules (containing (−)-epicatechin as terminal units) could have occurred during the alcoholic fermentation, favored by the acidic conditions. Another novelty shown by wines in comparison to musts was the occurrence of hydroxycinnamic acids

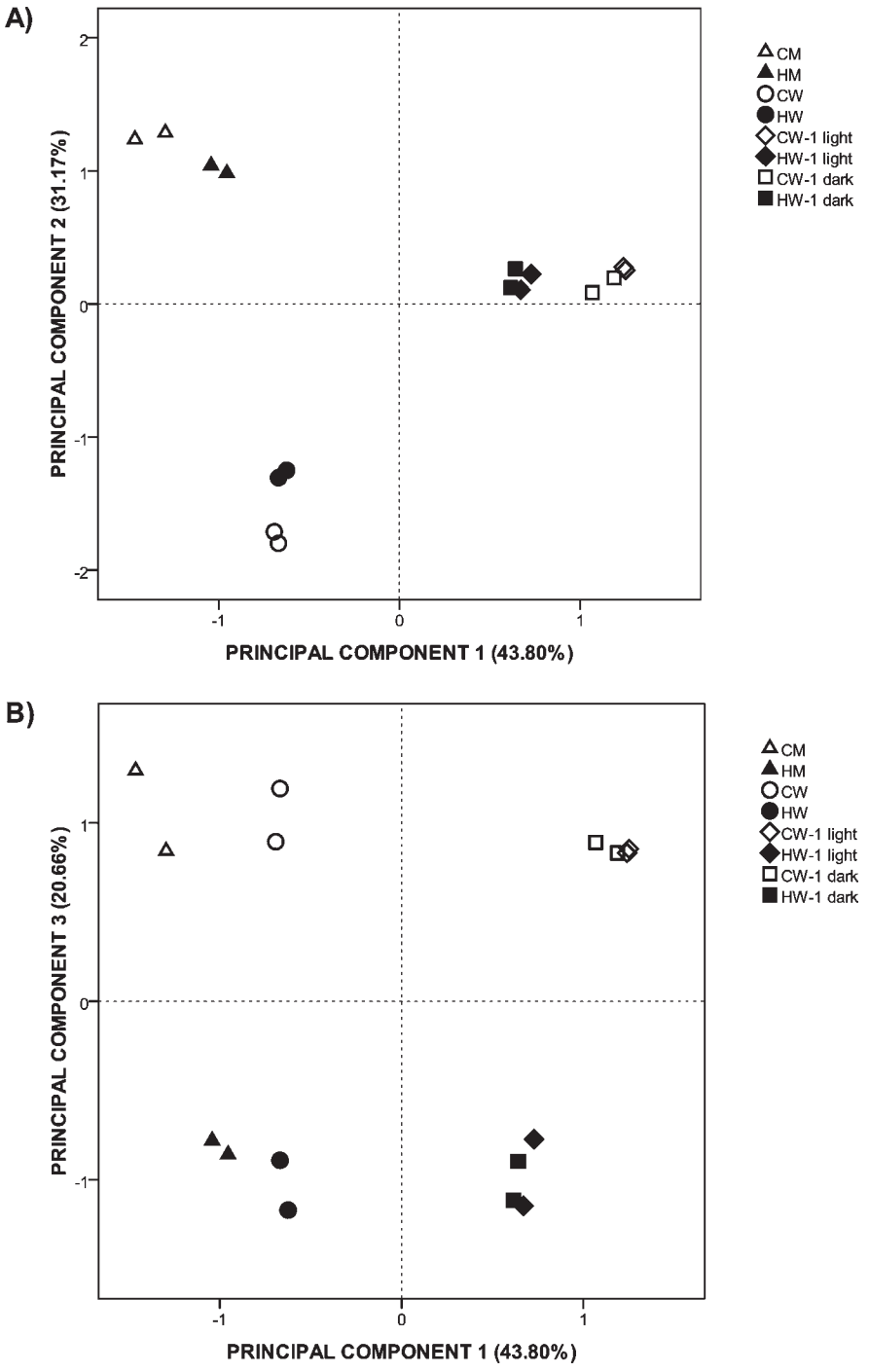

Figure 1. Plot of Chardonnay white wine samples in the space defined by principal components PC-1 versus PC-2 (A) and PC1 versus PC-3 (B): control (CM) and hyperoxygenated must (HM), control wines (CW), and wines from hyperoxygenated (HW) musts, and after 1 year of bottle storage (-1) in different conditions, light and dark, with regard to polyphenolic compounds and color parameters.

released by hydrolysis of their corresponding tartaric esters (hydroxycinnamoyltartaric acids) (Tables 2 and 3). After 1 year of bottle storage, the total amount of hydroxycinnamic acid derivatives measured by means of the absorbance at 320 nm (HCAD, Table 3) did not significantly change. However, whereas the content of the different hydroxycinnamoyltartaric acids did not follow a unique trend (increase of the concentrations of *t*-caftaric and *t*-fertaric acids, but decrease in the case of *c*-coutaric and *c*-fertaric acids) (PC-2 axis in Figure 1A; Table 3), a general increase of the free hydroxycinnamic acids (caffeic, *p*-coumaric, and ferulic acids) was observed, together with the formation of new GRP derivatives (hydrolysis of the tartaric or the glycinyl moieties; formation of monoethyl esters) (PC-1 axis in Figure 1A; Table 3). This behavior partially agreed with reported results for 1-year-stored Zalema and Colombard white wines.[18] Therefore, the identification of the recent five GRP derivatives was crucial to clarify the effect of different storage conditions in control wines and wines derived from

Table 5. Volatile Compound Concentration (Micrograms per Liter) and Standard Deviation (n = 2) of Control (CM) and Hyperoxygenated (HM) Chardonnay White Musts[a]

	CM	HM
esters		
isoamyl acetate	1.77 b ± 0.66	5.62 c ± 0.02
hexyl acetate	1.41 b ± 0.74	8.82 c ± 0.39
ethyl hexanoate	3.19 b ± 0.35	7.24 c ± 1.11
ethyl octanoate	11.1 ± 0.49	13.2 ± 0.21
ethyl decanoate	5.21 b ± 0.22	9.80 c ± 0.08
diethyl succinate	0.87 b ± 0.01	1.51 c ± 0.27
ethyl acetate	23.3 b ± 3.35	57.6 c ± 0.14
2-phenylethyl acetate	13.0 b ± 0.52	44.8 c ± 3.25
alcohols		
2-methyl-1-propanol	47.4 b ± 0.10	72.0 c ± 4.03
1-butanol	5.07 ± 0.35	5.23 ± 0.21
1-penten-3-ol	2.53 ± 0.47	2.92 ± 0.25
3-penten-2-ol	tr b	4.09 c ± 0.63
3-methyl-1-butanol	2.95 b ± 0.08	3.80 c ± 0.56
4-heptanol	0.80 ± 0.06	0.65 ± 0.06
(*Z*)-2-penten-1-ol	4.82 ± 0.05	5.19 ± 0.37
1,2-butanediol	0.66 b ± 0.06	1.16 c ± 0.17
3-octanol	tr b	1.37 c ± 0.16
1-octen-3-ol	tr b	1.72 c ± 0.04
1-heptanol	1.26 ± 0.25	1.25 ± 0.24
2-methoxy-1-butanol	tr b	1.16 c ± 0.04
2-methylthioethanol	0.24 ± 0.03	0.51 ± 0.02
2-ethyl-1-hexanol	1.22 b ± 0.66	1.83 c ± 0.16
3-methylthiopropanol	4.58 b ± 0.32	7.65 c ± 0.02
C_6 alcohols		
2-hexanol	2.62 ± 0.13	3.70 ± 0.72
1-hexanol	171 b ± 3.08	399 c ± 10.8
(*E*)-3-hexen-1-ol	nd b	8.33 c ± 0.15
(*Z*)-3-hexen-1-ol	nd b	9.61 c ± 0.06
(*E*)-2-hexen-1-ol	1.54 b ± 0.33	9.08 c ± 0.44
(*Z*)-2-hexen-1-ol	1.28 ± 0.27	1.68 ± 0.15
lactones		
γ-butyrolactone	0.21 ± 0.02	0.17 ± 0.00
pantolactone	5.67 b ± 1.20	7.42 c ± 0.10
terpenes		
α-terpineol	1.71 ± 0.16	1.61 ± 0.06
epoxy linalool	nd b	3.61 c ± 0.27
benzenic compounds		
benzaldehyde	3.05 ± 0.82	4.15 ± 0.42
benzyl alcohol	93.0 c ± 10.3	1.20 b ± 0.23
2-phenethyl alcohol	227 b ± 39.1	598 c ± 11.2
4-vinylguaiacol	13.5 ± 0.73	12.4 ± 0.28
benzoic acid	40.2 ± 11.8	47.2 ± 2.19
acids		
2-methylpropanoic acid	1.16 b ± 0.04	2.23 c ± 0.23
butyric acid	0.33 ± 0.02	0.50 ± 0.00
isovaleric acid	2.85 c ± 0.97	0.78 b ± 0.03
2-methylhexanoic acid	tr b	1.19 c ± 0.09
hexanoic acid	32.1 ± 1.27	37.0 ± 0.05
(*E*)-2-hexenoic acid	8.73 ± 0.34	9.35 ± 0.01
octanoic acid	13.5 ± 1.12	19.2 ± 0.15
decanoic acid	32.7 b ± 1.63	58.5 c ± 2.18
aldehydes		
2-hexenal	1.91 ± 1.02	9.81 ± 1.72
heptanal	0.75 b ± 0.06	2.68 c ± 0.07
miscellaneous		
3-hydroxy-2-butanone	0.80 ± 0.02	0.54 ± 0.07
6-methyl-5-hepten-2-one	0.23 ± 0.02	0.27 ± 0.04
5-ethyl-6-methyl 3-hepten-2-one	0.11 ± 0.03	0.11 ± 0.00
2-furanmethanol	4.22 b ± 1.36	5.46 c ± 0.61

[a] Different letters in the same row denote significant differences according to Student's t test ($p < 0.05$) between CM and HM. nd, not detected; tr, traces.

hyperoxygenated musts. Moreover, glycosylated flavonols completely disappeared in the stored white wines (PC-1 axis in Figure 1A; Table 3), but the concentration of their free aglycones also decreased (PC-2 axis in Figure 1A; Table 3).

With regard to the chromatic characteristics, it is highlighted that the lightness (L^*) of the wines derived from hyperoxygenated musts slightly increased as a consequence of the storage treatment (Table 3), regardless of the illumination conditions to which they were submitted.[18] Moreover, the yellow color component (b^*) of the stored control wines increased during storage, whereas changes were not observed in wines derived from hyperoxygenated musts. Therefore, wines submitted to hyperoxygenation treatment and further bottle storage greatly maintained the initial wine color, having greater resistance to browning. This result agreed with those of Castro et al.,[2] who demonstrated, by measurement of the absorbance at 490 nm, that browning of Fino Sherry wines obtained from hyperoxygenated must was lower after 1 year of storage in bottles. In contrast, no significant differences were observed in chromatic characteristics attributable to the different storage conditions (lightness vs darkness), contrarily to Maury et al.,[34] who described that white wines stored under dark conditions had lower resistance to browning than those stored under light conditions.

As depicted in Figure 1B, hyperoxygenated musts and their corresponding wines could be easily differentiable. The hyperoxygenation treatment produced a significant decrease of the concentration of virtually all phenolic compounds in HM with regard to CM, mainly all of the *trans* isomers of hydroxycinnamic acid derivatives, gallic acid, flavonols, and flavan-3-ols (Table 2). The aforementioned differences were maintained in the wines elaborated from CM and HM (Table 3), in agreement with results reported for other white wines obtained by hyperoxygenation of must.[35] Regardless of the storage conditions, the fraction of wine phenolic compounds more affected by the hyperoxygenation treatment were hydroxycinnamic acid derivatives, in agreement with previous results,[2] giving rise to significantly lower concentrations of virtually all of them, whereas benzoic acids and both flavonol glycosydes and aglycones were not so much affected.

Volatile Compounds Identified in Chardonnay Musts and Wines. A total of 95 volatile compounds were identified in Chardonnay white musts, wines, and aged wines, belonging to different chemical families (esters, alcohols, lactones, terpenes, benzenic and furanic compounds, dioxanes and dioxolanes, acids, and C_{13}-norisoprenoids) (Tables 5 and 6).

Table 6. Volatile Compound Concentration (Micrograms per Liter) and Standard Deviation (n = 2) of Control Chardonnay White Wines (CW) and Wines from Hyperoxygenated Musts (HW) and after 1 Year of Bottle Storage (-1) in Different Conditions, Light and Dark [a]

	CW	HW	CW-1 light	HW-1 light	CW-1 dark	HW-1 dark
major volatile compounds						
acetaldehyde[b]	179 c ± 21.4	98.4 b ± 21.2	162 bc ± 16.8	111 b ± 15.1	158 bc ± 13.2	141 bc ± 14.9
ethyl acetate[b]	38.6 b ± 1.89	38.4 b ± 3.37	56.7 c ± 5.65	54.9 c ± 2.91	67.4 c ± 7.05	67.3 c ± 6.68
methanol[b]	61.0 ± 0.92	54.3 ± 10.4	43.5 ± 0.77	41.0 ± 4.38	45.6 ± 3.35	42.7 ± 3.36
1-propanol[b]	43.4 ± 2.90	46.1 ± 0.58	42.8 ± 1.71	44.2 ± 2.82	43.6 ± 3.98	45.4 ± 2.11
isobutanol[b]	31.6 ± 5.48	33.3 ± 1.97	31.2 ± 3.08	31.5 ± 0.65	31.8 ± 4.45	32.3 ± 3.05
2-methyl-1-butanol[b]	71.7 d ± 2.84	60.6 c ± 4.11	28.7 b ± 0.41	22.7 b ± 0.04	29.0 b ± 1.36	25.3 b ± 0.46
3-methyl-1-butanol[b]	133 ± 9.95	111 ± 2.50	127 ± 6.75	103 ± 7.44	129 ± 11.6	108 ± 2.72
minor volatile compounds						
esters						
isoamyl acetate	1225 c ± 16.1	1265 c ± 13.4	120 b ± 2.11	131 b ± 1.21	135 b ± 4.77	154 b ± 0.34
hexyl acetate	56.8 c ± 3.97	88.6 d ± 0.47	4.95 b ± 0.05	5.46 b ± 0.07	5.60 b ± 0.14	6.38 b ± 0.10
ethyl butyrate	81.5 b ± 2.11	92.6 c ± 2.95	98.3 c ± 3.37	114 d ± 3.83	91.1 c ± 2.23	111 d ± 1.04
ethyl hexanoate	655 b ± 17.7	693 c ± 2.70	813 d ± 8.99	946 e ± 21.6	787 d ± 16.1	936 e ± 2.55
ethyl pyruvate	4.46 ± 2.88	3.81 ± 2.76	3.91 ± 1.60	3.22 ± 0.23	8.14 ± 0.07	7.43 ± 2.39
ethyl heptanoate	5.96 b ± 0.88	6.93 b ± 0.81	17.2 c ± 0.25	17.8 c ± 0.73	18.2 cd ± 0.56	19.0 d ± 1.70
ethyl lactate	13.8 b ± 2.42	10.2 b ± 1.22	68.9 c ± 9.05	68.8 c ± 0.97	73.5 c ± 1.01	39.9 b ± 34.2
ethyl octanoate	1106 c ± 13.5	1388 d ± 15.6	698 b ± 11.8	776 b ± 11.0	763 b ± 14.8	812 b ± 3.84
ethyl 2-hydroxy-3-methylbutanoate	2.81 b ± 0.13	3.54 b ± 0.74	19.9 c ± 2.18	23.2 c ± 0.15	27.9 d ± 1.11	20.6 c ± 3.08
ethyl 4-methyl-2-hydroxypentanoate	13.6 b ± 2.42	11.2 b ± 0.18	51.4 c ± 3.53	54.7 c ± 0.37	59.2 c ± 0.37	50.7 c ± 4.93
ethyl decanoate	454 d ± 4.85	504 e ± 5.94	80.2 b ± 2.22	91.8 bc ± 3.67	95.7 bc ± 2.47	119 c ± 4.19
ethyl 4-oxopentanoate	nd b	nd b	4.82 c ± 0.72	4.21 c ± 0.40	4.33 c ± 0.09	4.72 c ± 0.68
ethyl methyl succinate	2.02 b ± 0.32	2.69 b ± 0.23	51.1 c ± 2.78	48.8 c ± 1.05	56.1 d ± 0.53	47.6 c ± 3.33
diethyl succinate[b]	0.86 b ± 0.11	0.80 b ± 0.00	9.95 c ± 1.58	7.91 c ± 0.11	9.12 c ± 0.05	8.73 c ± 0.67
ethyl 4-hydroxybutyrate	628 d ± 7.82	567 c ± 6.73	131 b ± 7.56	120 b ± 7.23	122 b ± 3.65	112 b ± 6.87
ethyl laurate	638 d ± 13.86	572 c ± 6.73	134 b ± 4.04	123 b ± 3.75	125 b ± 3.76	110 b ± 2.46
ethyl 3-methylbutyl succinate	80.1 d ± 6.12	73.5 d ± 6.80	42.0 c ± 8.69	47.2 c ± 2.58	30.1 b ± 7.15	34.9 b ± 5.17
diethyl malate[b]	0.12 b ± 0.01	0.11 b ± 0.01	1.29 c ± 0.01	1.35 c ± 0.01	0.47 b ± 0.00	1.65 c ± 0.31
ethyl glutarate	107 b ± 5.52	82.8 b ± 3.82	905 d ± 68.6	782 c ± 14.8	876 d ± 5.18	797 c ± 8.45
diethyl hydroxyglutarate	207 b ± 4.98	151 c ± 6.21	1239 d ± 10.5	1197 d ± 14.3	1295 d ± 13.5	1242 d ± 15.7
diethyl monosuccinate[b]	3.96 b ± 0.37	4.03 b ± 0.18	15.2 c ± 2.89	11.1 c ± 0.01	12.9 c ± 0.00	11.7 c ± 0.89
2-phenylethyl acetate	363 c ± 2.01	529 d ± 47.0	40.8 b ± 0.06	25.7 b ± 2.40	21.5 b ± 1.30	21.8 b ± 2.51
alcohols						
2-methyl-1-propanol	147 ± 6.52	163 ± 6.33	189 ± 11.5	234 ± 2.75	239 ± 9.24	159 ± 0.11
1-butanol	16.7 bc ± 0.38	13.6 b ± 1.75	21.7 d ± 0.10	18.3 bcd ± 1.13	19.3 cd ± 0.28	14.2 b ± 2.81
3-methyl-3-buten-1-ol	0.85 b ± 0.06	0.64 c ± 0.00	nd b	nd b	nd b	nd b
(*Z*)-2-penten-1-ol	1.20 c ± 0.01	2.29 d ± 0.59	nd b	2.04 cd ± 0.34	1.73 cd ± 0.01	1.93 cd ± 0.02
C_6 alcohols						
1-hexanol	206 b ± 2.15	330 c ± 17.8	315 c ± 4.96	483 d ± 4.35	317 c ± 6.73	490 d ± 21.7
(*E*)-3-hexen-1-ol	15.5 c ± 0.21	11.8 bc ± 2.11	20.4 d ± 3.57	8.79 b ± 0.38	14.7 c ± 1.02	8.48 b ± 0.20
(*Z*)-2-hexen-1-ol	1.17 c ± 0.02	1.95 c ± 0.79	nd b	nd b	nd b	nd b
lactones						
γ-butyrolactone	19.4 c ± 1.56	20.4 c ± 0.65	15.3 b ± 0.72	19.3 c ± 1.41	16.8 b ± 0.90	21.9 c ± 1.12
4-hydroxyhexanoic acid lactone	nd b	nd b	12.8 c ± 1.46	12.5 c ± 0.67	15.3 d ± 0.97	11.9 c ± 0.40
pantolactone	29.1 e ± 0.57	23.2 d ± 1.34	15.3 b ± 0.71	14.9 b ± 0.90	17.4 c ± 0.57	16.9 c ± 1.05
δ-decalactone	19.0 c ± 0.49	22.9 d ± 0.28	13.1 b ± 1.05	16.7 bc ± 0.54	14.6 b ± 0.87	17.5 c ± 1.17
γ-undecalactone	247 b ± 9.65	205 b ± 13.2	518 c ± 2.94	500 c ± 7.67	561 c ± 7.56	533 c ± 6.07
terpenes						
linalool	5.07 c ± 0.35	6.80 d ± 0.39	3.44 b ± 0.02	5.48 c ± 0.63	2.12 b ± 0.42	3.35 b ± 0.53
citronellol	4.10 c ± 0.17	4.57 d ± 0.14	nd b	nd b	nd b	nd b
geranic acid	60.1 bc ± 12.9	74.1 c ± 12.4	nd b	nd b	nd b	nd b
benzenic compounds						

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Table 6. Continued

	CW	HW	CW-1 light	HW-1 light	CW-1 dark	HW-1 dark
benzaldehyde	9.62 c ± 2.35	13.4 c ± 1.30	3.77 b ± 0.47	4.76 b ± 0.40	5.10 b ± 0.10	5.99 b ± 0.61
benzyl alcohol	103 b ± 2.73	121 c ± 3.50	144 d ± 5.37	165 e ± 1.55	174 e ± 5.95	192 f ± 6.88
2-phenylethyl alcohol	5343 b ± 15.3	5362 b ± 23.9	7135 c ± 16.2	7399 d ± 12.6	8394 e ± 17.0	8577 f ± 14.0
benzofuran	nd b	nd b	6.95 c ± 0.88	10.2 d ± 0.47	8.70 cd ± 0.88	8.34 cd ± 1.61
4-vinylguaiacol	116 b ± 5.31	137 b ± 1.46	237 c ± 41.7	199 c ± 9.77	243 c ± 5.13	224 c ± 32.1
benzoic acid	80.8 c ± 2.53	106 d ± 9.19	84.3 c ± 2.99	87.5 cd ± 4.04	53.6 b ± 3.75	78.1 c ± 5.59
furanic compounds						
furfural	5.62 b ± 1.69	0.40 b ± 0.11	72.1 d ± 3.31	56.5 c ± 1.35	85.4 e ± 0.62	62.7 c ± 5.21
5-methyl-2-furancarboxaldehyde	nd b	nd b	14.3 c ± 1.75	10.7 c ± 0.56	14.8 c ± 0.63	13.8 c ± 2.70
ethyl furoate	5.24 b ± 0.86	4.95 b ± 0.13	30.2 d ± 2.63	21.4 c ± 1.87	23.5 c ± 0.82	26.4 cd ± 2.23
2-furanmethanol	2.26 c ± 0.63	3.73 d ± 0.03	nd b	nd b	nd b	nd b
dioxanes and dioxolanes						
2-ethyl-2,4,5-trimethyl-1,3-dioxolane	nd b	nd b	37.6 e ± 2.67	32.8 d ± 0.16	38.1 e ± 0.16	15.3 c ± 1.73
c-5-hydroxy-2-methyl-1,3-dioxane	nd b	nd b	15.1 d ± 2.53	17.3 d ± 2.80	18.8 d ± 3.28	7.25 c ± 1.10
c-4-hydroxy-2-methyl-1,3-dioxolane	nd b	nd b	2.74 c ± 0.42	3.42 c ± 0.44	5.15 d ± 0.99	2.52 c ± 0.48
t-4-hydroxy-2-methyl-1,3-dioxolane	nd b	nd b	3.32 c ± 0.11	5.96 d ± 1.16	2.88 c ± 0.38	5.79 d ± 1.00
t-5-hydroxy-2-methyl-1,3-dioxane	nd b	nd b	41.0 d ± 3.18	24.6 c ± 0.59	49.2 e ± 0.34	19.8 c ± 3.87
acids						
butyric acid	84.5 b ± 3.00	96.4 c ± 2.05	101 c ± 2.24	120 d ± 3.83	89.2 b ± 3.51	82.6 b ± 3.71
hexanoic acid[b]	0.63 b ± 0.01	0.74 b ± 0.05	1.06 c ± 0.17	1.02 c ± 0.01	0.93 c ± 0.00	1.07 c ± 0.04
octanoic acid[b]	2.49 b ± 0.02	2.58 c ± 0.02	2.63 d ± 0.02	2.72 e ± 0.01	2.56 c ± 0.02	2.62 d ± 0.01
decanoic acid	557 c ± 23.9	559 c ± 20.0	410 b ± 34.73	337 b ± 3.61	389 b ± 12.1	390 b ± 36.2
dodecanoic acid	147 d ± 0.25	148 d ± 20.9	51.0 c ± 1.17	33.2 bc ± 1.03	54.0 c ± 6.38	17.9 b ± 1.80
C_{13}-norisoprenoids						
β-damascenone	15.6 cd ± 2.29	6.32 b ± 1.28	18.6 d ± 0.22	13.0 c ± 0.29	12.0 c ± 1.68	14.0 c ± 0.90
3-hydroxy-β-damascone	24.3 d ± 2.84	31.9 de ± 1.63	18.0 c ± 0.15	15.8 b ± 0.65	18.1 c ± 1.30	19.8 c ± 1.71
3-oxo-α-ionol	135 d ± 6.11	173 e ± 5.71	82.6 b ± 4.47	89.1 bc ± 2.81	87.8 b ± 1.71	111 c ± 9.50
miscellaneous						
2-methyldihydro-3(2*H*)thiophenone	3.99 ± 0.31	3.41 ± 0.51	4.56 ± 0.01	3.86 ± 0.04	3.42 ± 0.00	3.60 ± 0.56
TDN	nd b	nd b	6.09 cd ± 0.30	4.99 c ± 0.07	7.02 d ± 0.57	6.48 cd ± 1.03

[a] Different letters in the same row denote significant differences ($p < 0.05$) according to Student−Newman−Keuls test. nd, not detected. [b] mg/L.

Among the varietal volatile fractions, C_6 alcohols (e.g., 1-hexanol, (*E*)-3-hexen-1-ol, (*Z*)-2-hexen-1-ol), benzenic (e.g., benzaldehyde, benzyl alcohol, 2-phenylethyl alcohol), terpenes (e.g., linalool, citronellol, geraniol), and C_{13}-norisoprenoids compounds (e.g., β-damascenone) have been identified in Chardonnay musts and wines. The latter families of volatile compounds play an important role in the varietal character of wines because they have pleasant aroma (fruity and floral) and very low odor threshold. Benzenic compounds are also an important group within varietal aromas, including aromatic alcohols, aldehydes, volatile phenols, and shikimic acid derivatives in Chardonnay musts and wines.[36]

In addition, several volatile compounds formed during the alcoholic fermentation have been also identified in this study. Among these, it is worth mentioning alcohols, fatty acids, and lactones. Moreover, Chardonnay wines contained a large extent of ethyl esters of fatty acids and acetates (short-, medium-, and long-chain ethyl esters). The aforementioned compounds have long been considered important contributors to wine aroma from neutral varieties,[37] because they occur in wines as major volatile constituents and in many cases above their odor threshold values. Also, acetaldehyde is highlighted as the dominating aldehyde present in these wines, due to its formation by the metabolism of yeasts. All of these compounds have been previously described by several authors, as usual volatile constituents of wines.[38−41]

Effects of Must Hyperoxygenation and Wine Bottle Storage on the Volatile Composition of Chardonnay Wines. According to Student *t* test ($p < 0.05$), the most relevant effect of hyperoxygenation treatment applied to Chardonnay musts was the significant increase of the concentration of C_6 alcohols and aldehydes, such as 1-hexanol and 2-hexenal (Table 5), due to the oxidation conditions which provoke the formation of these compounds from their precursors linoleic and linolenic acids.[42] Also, the synthesis of high alcohols such as 2-phenylethanol are favored by the presence of oxygen.[43] In general, volatile compound concentrations were higher in musts and wines derived from hyperoxygenated white musts as was described by other authors.[1,9,10] An exception was acetaldehyde and isoamylic alcohols, the contents of which were significantly decreased in wines derived from hyperoxygenated musts (HW), according to the Student−Newman−Keuls test ($p < 0.05$), which can be of great importance in the quality aroma of these wines (Table 6).

With regard to the minor volatile compounds, a significant increase in the concentration of some acetates and fatty acids esters, characterized by present fresh and fruity aromas, was observed in HW (isoamyl acetate, hexyl acetate, ethyl butyrate, ethyl hexanoate, ethyl decanoate, and 2-phenylethyl acetate).

The same fact was observed in terpenic and benzenic compounds with great impact on wine flavor, such as linalool, citronellol, benzaldehyde, benzyl alcohol, and benzoic acid. The concentration of 1-hexanol was also higher in wines derived from hyperoxygenated musts due to the already higher content in the oxygen-treated must (Table 6).

The concentration of the C_{13}-norisoprenoids 3-hydroxy-β-damascone and 3-oxo-α-ionol increased in HW. However, although above its odor threshold, β-damascenone presented a lower concentration in treated wines, probably due to its transformation in 3-hydroxy-β-damascone with a lesser sensorial impact.

The increase of the concentration of virtually all volatile compounds as a consequence of hyperoxygenation treatment could be due to the activation of yeast metabolism as a consequence of the initial oxygen addition and/or to the increase of the "quality" of the must clarification obtained under hyperoxygenation conditions, which probably oriented the yeast metabolism to produce more esters than higher alcohol components.

To study the hyperoxygenation effect on the volatile fraction of aged white wines, 1 year of storage in different conditions (light and dark) was carried out (Table 6).

On the one hand, 1-year-aged Chardonnay wines had a lower concentration of the major isoamylic alcohols and acetaldehyde, according to the results obtained by Escudero et al.[19] and Silva-Ferreira et al.[44] in several wines from different parts of Spain (Macabeo, Airén, Viura, Parrellada, Chardonnay, and Moscatel), aged under oxygen conditions. Nevertheless, ethyl acetate increased in the same way in control and treated wines (CW-1 and HW-1). The concentration of the majority of acetates and fatty acids ethyl esters of octanoic, decanoic, and dodecanoic acids decreased in white aged wines, because they are slowly hydrolyzed during storage to attain equilibrium concentration.[45,46] However, other esters had higher concentrations after storage, especially esters of succinic acid, ethyl lactate, ethyl glutarate, and hydroxy esters, because of the high concentration of their corresponding acids in wines. The same behavior was observed in the case of the benzenic compounds (benzyl alcohol, 2-phenylethanol, and 4-vinylguaiacol).

As described by many authors, monoterpenes were degraded during aging, and only small amounts of linalool were detected in aged wines.[47] On the contrary, the only C_{13}-norisoprenoid for which the concentration increased with aging was β-damascenone, probably due to the acid hydrolysis of precursors. Changes due to the storage affected in the same way CW-1 and HW-1; therefore, the general higher concentration of many volatile compounds in the latter was maintained. It is important to highlight the increase undergone by virtually all furan derivatives in wines after 1 year of storage (although it was less evident for HW-1), which have been related to the untypical flavor of white wines aged in bottle.[46,48]

On the other hand, new volatile compounds were identified in CW and HW aged Chardonnay wines, regardless of the storage conditions, such as 1,2-dihydro-1,1,6-trimethylnaphthalene (TDN), dioxanes and dioxolanes (2-ethyl-2,4,5-trimethyl-1,3-dioxolane, *cis* and *trans* 5-hydroxy-2-methyl-1,3-dioxane, and 4-hydroxy-2-methyl-1,3-dioxolane), and furanic and benzenic compounds (5-methyl-2-furancarboxaldehyde and benzofuran) (Table 6).

TDN can be released from precursors in wines by acid hydrolysis during storage and has been described as responsible for the kerosene-like odor of Riesling aged wines.[49] TDN concentrations of Chardonnay stored wines were in all cases below their odor threshold (20 μg/L), and no significant differences were found between stored wines.

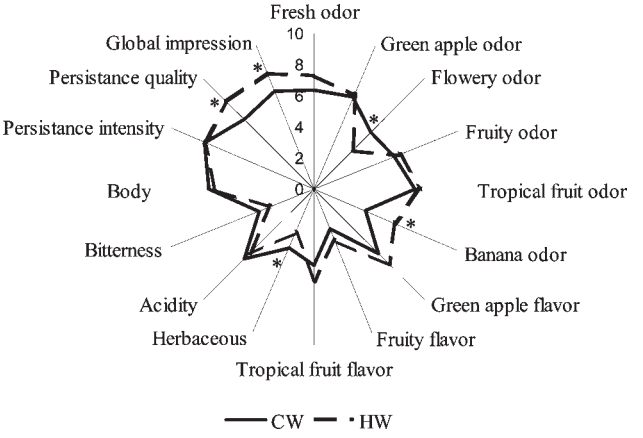

Figure 2. Olfactive and gustative attribute mean scores of Chardonnay control wine (CW) and wine from hyperoxygenated must (HW). *, significant differences according to Student's t test ($p < 0.05$) between CW and HW.

cis and *trans* isomers of 5-hydroxy-2-methyl-1,3-dioxane and 4-hydroxy-2-methyl-1,3-dioxolane are formed by reaction between glycerol and acetaldehyde in acid conditions.[50] Their aroma descriptors have been reported as "sweety" and "old port-like", but the concentration for the odor threshold of the total dioxanes and dioxolanes is higher in comparison to the content present in our stored wines (100 mg/L). The prevalence of dioxanes against dioxolanes in Chardonnay aged wines is noted. Their concentration was significantly lower in HW-1, even under aging in dark conditions. Their significantly lower concentration in wines derived from hyperoxygenated musts demonstrated the protection against oxidation in these wines, mainly without the presence of light.[48] Chemical oxidation of phenolic compounds can happen during storage in the presence of small amounts of oxygen, releasing oxygen peroxide that, by the Fenton reaction, can also generate hydroxyl radicals. Hydroxyl radicals are involved in the ethanol oxidation to acetaldehyde.[51] The low amounts of phenolic compounds observed in wines derived from hyperoxygenated musts could justify the lower amount of acetaldehyde available in these wines to produce dioxanes and dioxolanes.

The isomer *trans*-5-hydroxy-2-methyl-1,3-dioxane was the most abundant in control wines, presenting significantly lower concentration in HW-1, regardless of aging conditions.

Descriptive Sensorial Analysis. Figure 2 shows the attributes selected by descriptive sensorial analysis to describe the samples, together with the mean scores for each. With the aim of elucidating the significant sensory differences as a result of hyperoxygenation treatment, Student's t test was applied to the set of data. On the one hand, with regard to the olfactory analysis, oxygen addition provoked a significant increase of banana aroma that can be related with the higher concentration of isoamyl acetate in these wines. In addition, fruity and tropical fruit notes were slightly improved as a result of oxygen addition, probably due to the increase in short- and medium-chain fatty acid esters (Table 6). The same happened with the increase of fresh odor attribute in HW, which could be associated with the higher concentration of C_6 alcohols previously commented.

On the other hand, a significant diminution of floral aroma was observed, probably related to the lower concentration of β-damascenone (Table 6), responsible for the flowery character.[52]

Herbaceous and bitterness were lower in oxygen-treated wines as a result of the precipitation of polyphenolic compounds after the must hyperoxygenation. Consequently, Chardonnay white wines derived from hyperoxygenated musts were significantly and positively valuated, in agreement with the results obtained by Cheynier et al.[1] in oxygen-treated wines of the same variety.

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ABBREVIATIONS USED

HCAD , hydroxycinnamic acid derivatives; *t* , *trans*; *c* , *cis*; GRP , grape reaction product, 2-*S*-glutathionylcaftaric acid; GSCf , 2-*S*-glutathionylcaffeic acid; GRP-Et 1 , monoethyl ester of 2-*S*-glutathionylcaftaric acid at the carboxy group of the glycine terminal unit of the glutathionyl moiety; GRP-Et 2 and GRP-Et 3 , monoethyl esters of 2-*S*-glutathionylcaftaric acid at the two carboxy groups of the caftaric acid moiety; Glu-cys-GRP , 2-*S*-(γ-glutamylcysteinyl)-*trans*-caftaric acid; TDN , 1,2-dihydro-1,1,6-trimethylnaphthalene.

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