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ORIGINAL PAPER

Influence of *Quercus alba* oak geographical origin on the colour characteristics and phenolic composition of Tempranillo wines

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Abstract

This research analyzes the influence of *Quercus alba* barrels from four different forests of the USA [Missouri (M), Ohio (O), Kentucky (K) and Pennsylvania (P)] on the colour parameters, phenolic composition, and visual and gustatory sensory characteristics of 12 Tempranillo red wines from different wineries after 12 months of aging. Oak origin significantly affected the colour parameters of the wines, except for tonality, and the content of anthocyanins, stilbenes and ellagitannins but had little effect on the rest of the phenolic compounds. Wines aged in K and O barrels showed the highest content of ellagitannins. Wines aged in P barrels had the lowest content of phenolic compounds and colour intensity (CI), while K and O barrels produced the highest levels of ellagitannins. K wines were the best value in the global perception of the taste analysis. The influence of the barrel origin on the phenolic composition of the wines was also evaluated according to their initial phenolic composition. Wines with the highest initial CI and phenol content also showed the highest content of ellagitannins after barrel aging. When aging wines with high CI and phenol content, M barrels provided the wines with the highest content of ellagitannins.

Keywords Red wine · Forest · American oak · Chemical composition · Sensory analysis · Aging

Introduction

Aging in oak barrels is a conventional technique used to improve the quality of red wines. Wine aging in oak barrels involves some complex and slow changes that have a positive impact on its flavor, aromatic complexity, and colour stability. During the aging process, the oak barrel allows a slow and continuous entry of oxygen and contributes to the gradual release of polyphenolic and volatile compounds into the wine. The content of volatile compounds, ellagitannins, hydroxybenzoic and hydroxycinnamic acids, and aldehydes of the oak heartwood varies significantly depending on the oak species and the geographical origin of the wood [1]. Wine aging in oak barrels includes many chemical reactions and changes in wine phenolic composition that may stabilize the red wine colour and decrease its astringency [2]. These reactions are due to the moderate wine oxidation in the oak barrel [3], to reactions of wine compounds with compounds extracted from the wood [4], and to adsorption of wine phenolic compounds on the wood surface [5].

Various studies on the chemical composition of oak woods have pointed out the great variability of phenolic extractable compounds as a function of their forest origin [6–8]. In fact, it has been observed that the content of ellagitannins allows the distinction of French and East European oak woods (*Quercus robur* and *Quercus petraea*) [6], as well as Portuguese and Spanish oak woods (*Quercus pyrenaica*) [9]. In addition, Feuillat and co-workers [10] also observed significant differences in the ellagitannin content from oak wood (*Quercus robur* and *Quercus petraea*) of different French forests. Geographical origin also affected the content of phenolic compounds of woods from Portuguese cooperage (*Quercus pyrenaica*) [7, 8].

Nevertheless, there are a limited number of studies on the influence of oak forest origin on the wine phenolic composition, being French oak woods the better studied in this regard. Therefore, the effect of oak forests from different French locations [11, 12] showed that wines aged in Limousin barrels had the highest concentration of ellagitannins

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and total polyphenols. Moreover, it has also been reported that wines aged in Spanish oak barrels showed higher levels of monomeric anthocyanins compared to those aged in French and Central European oak barrels, which may be attributed to the relatively weaker polymerization reactions in wines aged in Spanish oak, which are likely linked to less oxidative conditions [13]. Significant differences were also observed in the content of monomeric phenolics in wines aged in Spanish and French *Quercus robur* and *Quercus petraea* barrels [14].

One of the main oak species used for wine ageing is *Quercus alba*, which grows in various areas across the United States [15], encompasses the whole eastern part of the United States, stretching north into southern Ontario and Quebec, Canada, from the Gulf Coast to the Great Lakes and from the Atlantic Seaboard in the east to the Missouri River in the west [16].

Although *Quercus alba* is one of the most common and used oak in the eastern United States, there is little information about its region of origin. According to our knowledge, no studies have been published evaluating the influence of *Quercus alba* barrels from different geographical origins in the United States on the phenolic composition, colour characteristics and sensory analysis of wines. A recent study of our research group has shown that *Quercus alba* oak barrels from different geographical origins in the USA produced wines with different aromatic characteristics, being the wines aged in barrels from Missouri and Kentucky forests were the ones with the highest scores on global aromatic perception [17].

Therefore, this study aims to evaluate the influence of the USA geographical origin of *Quercus alba* oak barrels on the phenolic composition, colour properties, and visual and gustatory sensory properties of Tempranillo wines after twelve months of barrel aging. For this, *Quercus alba* oak barrels from Missouri, Ohio, Kentucky, and Pennsylvania forest were employed. Moreover, wines were grouped based on their initial polyphenolic content, total polyphenol index and colour intensity to assess whether the barrel origin had a different effect depending on the initial polyphenolic composition of the wines.

Materials and methods

Barrels

225-L oak barrels were manufactured in Murua Cooperage (Logroño-La Rioja, Spain) in 2018 as previously described [17]. *Quercus alba* staves from four different forests of the USA: Missouri (M), Kentucky (K), Ohio (O) and Pennsylvania (P) were used. Raw staves had the following measurements: 950 mm high × 27 mm thick × 50–100 mm wide with natural seasoning at the cooperage for 24 months. Barrels were introduced in water for 40 min to a maximum of 3 h at high temperatures to decrease bitter tannins and favors the toasting process. Finally, the barrels were toasted at medium intensity on oak firewood.

Vinifications and wine samples

Vinifications were carried out in 12 wine cellars from Spanish Qualified Designation of Origin (D.O.Ca) Rioja and Designation of Origin (D.O.) Ribera del Duero using *Vitis vinifera* cv Tempranillo by traditional red winemaking process [17]. Once malolactic fermentation was completed, wines were aged in triplicate in four different geographical origins of *Q. alba* oak barrels for a period of 12 months. The storage conditions were maintained at a temperature of 14–16 oC and a relative humidity of 70–75%. As 12 wine cellars were part of the investigation, 144 *Q. alba* oak barrels were employed. Samples were taken in the initial wines and after 12 months of aging. Wines after 12 months of aging were named M, K, O and P wines.

Determination of oenological parameters and colorimetric characteristics of wines

The classical oenological parameters, total polyphenol index (TPI) and colour intensity (CI) were analysed according to methodologies exposed by the OIV (2003) [18]. Malic acid and the sum of glucose and fructose were analyzed by the autoanalyzer BioSystems Y15 (Biosystem, Barcelona, Spain). The method described by Ayala and co-workers [19] was used to assess the CIELAB parameters. The colour differences between wines were determined as: $\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{0.5}$ [20]. All analyses were made in triplicate.

Analysis of wine monomeric phenolic compounds

Monomeric phenolic compounds were analyzed by high-performance liquid chromatography (HPLC) (Agilent Technologies, Waldbronn, Germany) with photodiode array detection (DAD) according to the methodology described in Gómez-Alonso and co-workers [21].

A modular 1100 Agilent liquid chromatograph (Agilent Technologies, Waldbronn, Germany) equipped with one G1311A quaternary pump, an on-line G1379A degasser, a G1316A column oven, a G1313A automatic injector, and a G1315B photodiode-array detector (DAD) controlled by the Chemstation Agilent software was used. Separation was done in an ACE HPLC column [5 C18-HL, particle size 5 µm; 250 × 4.6 mm (Teknokroma, Barcelona, Spain)]. The solvents used were: (A) 50 mmol/L ($NH_4H_2PO_4$ at pH 2.6), (B) acetonitrile/solvent A (80:20% v/v) and (C) 200 mmol/L (o-phosphoric acid at pH 1.5), establishing the following gradient: isocratic 0% B and 0% C during 5 min, from 0 to 8% B in 12 min, from 8 to 14% B and 0% to 86% C in 5 min, from 14 to 18% B and 86% to 82% C in 7 min, from 18 to 21% B and 82% to 79% C in 26 min, from 21 to 33% B and 79% to 67% C in 15 min, from 33 to 50% B and 67% to 50% C in 8 min, from 50 to 80% B and 50% to 0% C in 8 min, at a flow of 1 mL/min. Phenolic compounds were identified according to the retention times of pure compounds and the UV–visible characteristics obtained from chemical standards. Quantification was made using DAD chromatograms recorded at 280 nm (hydroxybenzoic acids and flavan-3-ol), 320 (hydroxycinnamic acids and stilbenes), 360 nm (flavonols), and 520 (anthocyanins). The sum of total hydroxybenzoic acids, hydroxycinnamic acids, flavonols, flavan-3-ol, stilbenes, ellagitannins, and anthocyanins was used to determine the total phenolics. All analyses were done in triplicate.

Analysis of wine ellagitannins

The ellagitannin content, expressed as castalagin, was evaluated by HPLC–DAD after acid hydrolysis according to the method described by Peng and co-workers [22] with little modifications. Briefly, 10 mL of wine were concentrated using a rotavapor until dryness. Next, it was redissolved in methanol, and a fraction was analyzed using HPLC–DAD for the determination of free ellagic acid. Another fraction was hydrolyzed in an acidic HCl/MeOH medium and subsequently analyzed using HPLC–DAD for the determination of total ellagic acid. Separation was done in an Agilent XDB-C18 column (150 × 4.6 cm × 5 µm), with a wavelength of 370 nm.

Wine sensory analysis for the visual and gustatory phase

Sensory evaluation was performed in wines after 12 months of aging in a test room designed according to the ISO 8589:2010 Standard. K, M, O and P wines were tasted separately for each wine cellar. The tasting panel was formed by a group of 12 oenologists from different wineries in the D.O.Ca Rioja (7 males and 5 females, 25–40 years old). A standardized tasting sheet of descriptive analysis method ISO 11035 was used. Before the sensory analysis, the panelist established a common set of attributes for the visual and the gustatory phases by determining both qualitative and quantitative criteria.

For both the visual and gustatory phases, a structured numerical scale with 6 points was utilized. A score of 0 corresponded to the absence of intensity while a score of 5 indicated the highest intensity. The Geometric Mean (GM %) was utilized to classify the visual and gustatory descriptors, in accordance with ISO 11035 from the International Organization for Standardization.

Statistical procedures

Statistical analyses were performed using SPSS Statics 23 (IBM Corp., Armonk, NY, USA). Multivariate analysis of variance (MANOVA) was used to determine differences among barrel origins considering wine mean values as it considers both the factor of wine and the barrel origin (Table 3, 5 and 7). Analysis of variance (ANOVA) was used to determine differences among barrel origins considering individual data for each wine cellar (Table 4 and 6). Post hoc Duncan ($p < 0.05$) was used to determine the significant differences between samples.

Moreover, wines were grouped based on their initial polyphenolic content, total polyphenol index and colour intensity to assess whether the barrel origin had a different effect depending on the initial polyphenolic composition of the wines. This clustering was carried out using the *k*-means method for three groups. These results were computed using R version 4.1.0 (R Core Team, Vienna, Austria). Heatmaps were also performed with the R version.

Results and discussion

Oenological parameters and colorimetric characteristics of the initial wines

General oenological parameters and colorimetric characteristics of the wines before aging are shown in Table 1.

The results for volatile acidity after malolactic fermentation showed no microbial alterations. The values of ethanol content, pH, titratable acidity and CIELAB parameters were like those obtained in other Tempranillo wines before barrel aging [23, 24]. The total polyphenol index (TPI) and colour intensity (CI) values of the initial wines were both high, indicating favorable aging potential.

Phenolic composition of the initial wines

Table 2 presents the content of the different families of monomeric phenolic compounds in the wines before aging.

The main hydroxybenzoic acid detected in wines was gallic acid (Table 2), and it was in the range reported [23, 24]. Ellagic acid was not detected in the wines before aging since this compound originates from hydrolysable tannins, predominantly derived from oak tannins [25]. Caftaric and coutaric acid were the primary hydroxycinnamic acids detected in the wines before aging. Concentration of tartaric esters and free acids were in the range

Table 1 General oenological parameters and colorimetric characteristics of Tempranillo wines before wine aging

Parameters[a]	Wine 1	Wine 2	Wine 3	Wine 4	Wine 5	Wine 6	Wine 7	Wine 8	Wine 9	Wine 10	Wine 11	Wine 12	Mean value[b]
pH	3.67	3.72	3.53	3.65	3.63	3.90	3.74	3.58	3.48	3.80	3.73	3.55	3.67 ± 0.12
Total acidity (g/L)	4.53	4.47	5.01	4.56	4.60	3.98	4.09	4.63	5.51	3.89	4.21	5.05	4.54 ± 0.47
Volatile acidity (g/L)	0.30	0.33	0.33	0.50	0.56	0.54	0.49	0.46	0.39	0.46	0.31	0.43	0.43 ± 0.09
Alcoholic degree (% v/v)	14.50	14.02	13.92	13.80	14.26	15.50	13.70	13.91	15.00	14.80	14.50	14.13	14.3 ± 0.5
Free sulfur dioxide (mg/L)	30.00	57.00	19.00	30.00	17.20	35.00	38.00	43.00	40.00	40.00	34.00	38.00	35.11 ± 11
Total sulfur dioxide (mg/L)	50.00	88.00	44.00	66.00	44.00	54.00	82.00	68.00	73.00	75.00	57.00	78.00	65 ± 15
Glucose + fructose (g/L)	1.72	2.33	2.10	2.53	2.61	2.26	2.05	2.66	2.78	2.06	1.79	1.22	2.18 ± 0.45
Malic acid (g/L)	0.15	0.22	0.16	0.25	0.11	0.21	0.30	0.16	0.20	0.26	0.13	0.18	0.19 ± 0.06
L^*	52.96	67.86	62.46	55.76	55.27	60.61	70.52	53.20	53.24	57.72	62.47	66.84	59.91 ± 6.17
a^*	44.29	28.90	33.90	44.29	44.68	38.01	23.34	44.34	42.19	38.89	34.81	30.52	37.35 ± 7.12
b^*	1.97	2.10	1.88	3.09	3.93	2.22	7.29	1.70	8.80	3.12	1.00	3.23	3.36 ± 2.35
C^*	44.33	28.98	33.95	44.40	44.85	38.07	24.45	44.37	43.10	39.02	34.82	30.69	37.59 ± 7.00
h^*	2.54	4.15	3.17	3.99	5.02	3.34	17.35	2.20	11.79	4.59	1.64	6.05	5.49 ± 4.57
Colour intensity (CI)	20.01	12.19	14.81	18.85	18.75	15.72	11.31	19.21	19.78	17.01	14.79	12.46	16.24 ± 3.14
Tonality (T)	0.64	0.74	0.70	0.61	0.62	0.66	0.85	0.61	0.70	0.68	0.67	0.72	0.68 ± 0.07
Total polyphenol index (TPI)	80.61	80.84	70.23	91.13	79.09	86.72	71.92	81.85	86.69	82.35	77.03	72.07	80 ± 7

L^* lightness; a^* red-green colour component, b^* yellow-blue colour component, C^* chroma, h^* hue angle

[a] Total acidity as g/L tartaric acid; volatile acidity as g/L acetic acid

[b] Mean values and standard deviations of the wines from the 12 wineries

Table 2 Phenolic composition (mg/L) of the wines before aging

Compounds[a]	Wine 1	Wine 2	Wine 3	Wine 4	Wine 5	Wine 6	Wine 7	Wine 8	Wine 9	Wine 10	Wine 11	Wine 12	Mean value[b]
Hydroxybenzoic acids													
Gallic acid	60.11	99.16	62.57	84.85	62.19	61.34	51.59	61.28	69.79	85.19	82.15	88.08	72.36 ± 14.81
Syringic a cid	5.90	4.53	6.14	7.05	7.61	6.02	6.30	5.98	7.54	6.17	6.61	5.73	6.30 ± 0.84
Ellagic acid	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Total	66.01	103.69	68.71	91.90	69.80	67.36	57.89	67.26	77.33	91.36	88.76	93.81	78.67 ± 14.51
Hydroxycinnamic acids													
cis-Caftaric acid	5.09	4.47	5.25	5.37	5.82	3.79	4.10	4.66	4.54	5.78	6.09	2.91	4.83 ± 0.93
trans-Caftaric acid	27.86	31.99	28.70	44.96	35.42	44.89	22.02	38.83	37.69	34.14	15.54	31.02	32.86 ± 8.65
cis-Coutaric acid	5.17	4.39	5.33	8.63	4.96	6.57	4.22	5.06	4.92	4.84	2.77	5.18	5.17 ± 1.40
trans-Coutaric acid	23.31	20.27	24.03	41.81	31.50	34.52	21.38	41.00	39.78	30.20	16.92	22.92	28.97 ± 8.72
Caffeic acid	1.81	7.25	1.87	2.78	4.89	5.18	12.30	3.46	3.36	4.77	17.25	2.34	5.61 ± 4.68
trans-Fertaric acid	1.36	1.34	1.40	1.46	1.64	1.36	1.14	1.29	1.25	1.61	1.98	1.07	1.41 ± 0.24
p-Coumaric acid	1.30	1.56	1.34	1.97	1.51	0.92	1.79	0.65	0.63	1.48	1.28	1.04	1.29 ± 0.42
Total	65.91	71.27	67.91	106.98	85.74	97.23	66.95	94.96	92.16	82.82	61.83	66.48	80.02 ± 15.20
Flavonols													
Myricetin-3-gal	5.55	3.85	5.90	5.84	4.82	4.86	1.48	3.76	2.94	5.26	4.71	1.88	4.24 ± 1.49
Myricetin-3-glc	23.61	19.74	17.45	20.87	15.51	9.35	22.18	23.74	20.37	19.04	18.99	12.01	18.57 ± 4.41
Quercetin-3-gal	4.82	3.38	2.49	6.96	2.21	3.42	1.81	2.85	3.59	2.31	2.44	2.46	3.23 ± 1.43
Quercetin-3-glc	5.41	4.81	7.74	5.82	6.79	3.78	4.04	7.34	5.07	4.40	4.64	4.24	5.34 ± 1.32
Quercetin-3-glcU	4.30	3.83	6.16	4.64	5.40	3.01	3.21	5.85	4.03	3.50	3.69	3.38	4.25 ± 1.05
Isorhamnetin-3-glc	2.27	1.91	2.86	2.29	4.78	2.11	1.91	2.42	2.79	2.54	2.07	1.31	2.44 ± 0.85
Myricetin	19.05	10.73	11.00	21.12	10.93	15.65	10.21	16.26	15.05	16.03	10.71	9.70	13.87 ± 3.84
Quercetin	9.68	12.83	12.69	17.19	10.72	15.86	4.97	17.74	14.25	7.88	4.37	8.39	11.38 ± 4.48
Kaempferol	0.57	2.87	2.17	4.18	1.57	2.40	0.39	4.02	3.06	0.87	0.39	1.34	1.99 ± 1.35
Isorhamnetin	0.68	1.00	0.99	1.27	1.75	1.07	0.49	1.05	0.60	0.50	0.24	0.56	0.85 ± 0.42
Total	75.94	64.95	69.45	90.18	64.48	61.51	50.69	85.03	71.75	62.33	52.25	45.27	66.15 ± 13.42
Flavan-3-ol													
Catechin	85.03	116.81	86.75	190.42	100.63	85.91	77.14	85.93	118.10	84.33	82.12	113.22	102.20 ± 31.32
Stilbenes													
trans-Piceid	2.11	2.48	2.19	2.08	2.88	5.06	2.31	1.81	1.73	2.52	2.29	2.53	2.50 ± 0.87
trans-Resveratrol	0.85	1.56	0.89	1.60	1.10	2.06	1.19	1.80	1.72	1.27	0.81	0.42	1.27 ± 0.49
Total	2.96	4.04	3.08	3.68	3.98	7.12	3.50	3.62	3.44	3.79	3.10	2.95	3.77 ± 1.12
Ellagitannins	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Anthocyanins													
Delphinidin-3-glc	36.61	21.12	37.73	63.74	17.07	30.82	41.22	40.64	17.52	39.80	37.48	32.45	34.68 ± 12.71
Cyanidin-3-glc	3.37	2.20	3.55	7.11	2.14	4.11	5.00	4.64	1.93	4.39	3.78	4.44	3.89 ± 1.44
Petunidin-3-glc	36.74	22.79	37.86	57.42	19.75	28.79	46.57	36.70	18.85	39.39	40.54	31.03	34.70 ± 11.26

published for Tempranillo wines after malolactic fermentation [26]. Wines showed values in flavonol content in the range obtained by other authors [24, 27]. Quercetin and myricetin-type flavonols were the main flavanols detected, representing together 87–95% of the total flavonol content in the wines. The content of kaempferol, isorhamnetin and their derivatives accounted for 5–13% of total flavonols in wines. Similar percentages were observed by Gómez-Alonso et al. [21] in Tempranillo young wines. The presence of flavonols in grape skins is exclusively in the form of glycosides [28]; and therefore, the occurrence of free flavonols in wines may result from the hydrolysis of their glycosides during the winemaking process. In fact, the average percentage of free flavonols found in wines was 30% to 57% of the total flavonols, which is in the range reported for red wines [29]. Catechin was the only flavan-3-ol detected in wines. Its concentration was slightly higher than that described in Tempranillo wines after malolactic fermentation [26]. The content of stilbenes agreed with the values found described in the bibliography [27]. *Trans*-piceid (resveratrol glucoside or polydatin) was the main stilbene in the wine samples, representing around 50–86% of the total content of stilbenes. Ellagitannins were not found in the initial wines because they are detected in wines as a result of oak barrel aging or the use of oak chips in stainless steel tanks [30], representing up to 10% of the dry weight of the wood core [31]. The total concentration of anthocyanins in the wines was within the range of values reported for young Tempranillo wines [26]. Therefore, malvidin 3-*O*-glucoside and its derivatives were identified as the most prevalent anthocyanins in the wines, being around 53–62% of the total anthocyanin content. Non-acylated anthocyanins were 81–87% of the total anthocyanin content. Within the acylated anthocyanins, *p*-coumaroyl derivatives were predominant, as typically observed in this grape variety [21].

Colour parameters and total polyphenol index of the wines after 12 months of aging

Table 3 shows the mean values of colour parameters and total polyphenol index of the 12 wines aged in K, M, O and P barrels for 12 months. Table 4 shows the individual values of all the wines from the 12 wineries.

All the wines showed similar CIELAB and CI parameters after 12 months of aging than those shown in the bibliography for Tempranillo wines aged in oak barrels for 12 months [20, 32].

Barrel origin significantly affected the CIELAB parameters of the wines. K, M, O and P wines showed differences in their CIELAB parameters, both when considering the mean values (Table 3) and the individual data of the

Table 2 (continued)

Compounds[a]	Wine 1	Wine 2	Wine 3	Wine 4	Wine 5	Wine 6	Wine 7	Wine 8	Wine 9	Wine 10	Wine 11	Wine 12	Mean value[b]
Peonidin-3-glc	8.67	6.60	9.11	14.32	19.72	11.45	11.28	11.15	3.59	10.46	11.36	11.16	10.74 ± 3.93
Malvidin-3-glc	124.81	88.10	128.61	172.53	103.28	87.09	155.5	110.2	57.28	128.7	134.8	102.7	116.13 ± 31.53
Delphinidin-3-acglc	1.28	1.77	1.34	3.84	1.47	1.39	2.59	1.98	1.26	2.35	1.88	1.70	1.90 ± 0.74
Cyanidin-3-acglc	1.42	1.47	1.50	6.67	1.08	0.71	3.27	2.47	0.82	1.28	1.84	1.12	1.97 ± 1.64
Petunidin-3-acglc	1.43	1.54	1.51	6.30	0.91	0.97	3.93	1.79	1.04	1.75	1.32	0.86	1.95 ± 1.60
Peonidin-3-acglc	0.77	1.20	0.81	1.05	2.41	1.02	1.74	0.91	1.77	0.81	0.85	0.83	1.18 ± 0.52
Malvidin-3-acglc	5.18	6.80	5.44	9.91	7.22	4.62	7.87	4.10	0.87	6.70	5.48	4.38	5.71 ± 2.26
Delphinidin-3-cmglc	3.86	2.42	4.06	7.80	1.64	2.91	6.11	4.17	1.81	4.67	3.78	2.91	3.85 ± 1.77
Cyanidin-3-cmglc	1.29	0.84	1.35	2.62	1.08	1.05	1.85	1.54	0.61	1.55	1.15	0.92	1.32 ± 0.53
Petunidin-3-cmglc	3.84	3.12	4.04	7.06	2.40	3.24	5.57	3.82	1.78	4.93	4.01	2.81	3.89 ± 1.44
Peonidin-3-cmglc	1.94	1.81	2.04	3.58	4.57	1.84	3.07	1.91	0.66	2.22	1.90	1.68	2.27 ± 1.02
Malvidin-3-cmglc	13.06	9.38	13.72	23.30	12.85	11.39	19.40	11.86	6.49	18.40	15.08	10.39	13.78 ± 4.67
Total	242.29	171.20	254.71	387.25	197.59	191.40	314.97	237.88	116.28	267.40	265.25	209.38	237.96 ± 70.07

glc glucoside, *acglc* acetyl-glucoside, *cmglc* coumaroyl-glucoside, *gal* galactoside, *glcU* glucuronide, *nd* not detected

[a] Mean values and standard deviations of the wines from the 12 wineries

Table 3 Mean colour parameters and total polyphenol index of the wines after 12 months of aging

	*L**	*a**	*b**	*C**	*h**	CI	T	TPI
K	54.27 a	37.61 c	10.03 b	39.66 c	13.75 bc	14.02 b	0.740	79.75
M	54.38 a	37.32 b	9.92 b	39.14 b	13.69 b	14.03 b	0.741	79.18
O	54.16 a	37.78 c	10.12 b	39.68 c	13.85 c	14.10 b	0.739	79.29
P	55.65 b	36.72 a	9.06 a	38.36 a	13.11 a	13.76 a	0.743	78.38

Mean values of the 12 wines are shown for each barrel origin. For each parameter, values with different letters are significantly different between the samples ($p \leq 0.05$)

*L** lightness, a^* red-green colour component, *b** yellow-blue colour component, *C** chroma, *h** hue angle, *CI* colour intensity, *T* tonality, *TPI* total polyphenol index, *K* wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels

wines (Table 4). Therefore, *L** values showed significant differences among origins in the 12 wines studied, while *b** showed differences in 11, *h** in 10, and *a** and *C** in 8 of the wines.

Wines aged in K and O barrels showed significantly higher mean chroma (*C**) values than wines aged in M and P barrels (Table 3), which means higher colour vividness. Therefore, wines aged in K and O barrels showed the highest *C** values in most of the wineries which showed significant differences in *C** (Table 4). In contrast, wines aged on P barrels had the lowest mean chroma (*C**) and hue angle (*h**), but the highest lightness (*L**) values (Table 3). These findings showed that P wines were more luminous (higher *L** values) but less colorfulness (lower *C**) wines, and were in agreement with the data observed for the wines in each individual winery. Hence, wines aged in P barrels showed the highest *L** value in 11 of the 12 wines studied (Table 4).

The *a** CIELAB parameter showed significant differences in the mean values among the wines (Table 3). Hence, K and O wines showed the highest mean contribution of redness (*a**) component, but this effect was not observed when observing each individual wine (Table 4). The lowest mean *a** value was obtained in P wines (Table 3), which also showed the lowest values in half of the wines with differences in the a* value (Table 4). Higher *a** values could be associated with a higher formation of anthocyanin-derived pigments, which could contribute to the stabilization of the flavylium red-colored form in wines aged in K and O barrels [33]. The lowest mean *a** values in wines aged in P barrels could be related to a higher degradation of anthocyanins and related pigments, as well as the precipitation of insoluble polymeric anthocyanin-derived pigments in these wines [34]. With regards to the *b** component, wines aged in M barrels showed the highest *b** values in 7 of the 11 wineries with differences (Table 4), which could be attributed to a greater loss of copigmentation effects which may be accompanied by the formation of red-orangish pigments derived from anthocyanin as pyranoanthocyanins [35], as well as higher oxidation phenomena of red wine pigments [34]. The lowest mean *b** value in wines aged in P barrels (Table 3) could be due to the fact that these wines maintained more of the anthocyanin-alkyl-catechin pigments that contribute to the purple colour [36].

Barrel origin also significantly affected the colour intensity (CI) of the wines but not the tonality (T). Hence, CI showed significant differences among origins in 9 of the wines studied, while T values showed differences in only 2 wines (Table 4). K, O and M wines showed the highest mean CI values (Table 3), in good agreement with the data obtained for each individual wine. Therefore, wines aged in M and O barrels showed the highest CI in the majority of wineries studied, followed by K wines (Table 4).

The colour difference values (ΔE^*) were calculated to determinate the general colorimetric differences among the K, M, O and P wines. These values were in the 0.22–2.13 CIELAB unit range (data not shown). In general, the human eye can differentiate between two colours when the difference in ΔE^* is equal to or greater than 1.0 CIELAB unit [37]. Therefore, the colour differences between K/M, M/O and O/K pairs were not detectable by the human eye (0.34, 0.55, and 0.22, respectively), and only the K/P, M/P, and O/P pairs of barrels could be clearly detected (2.09, 2.02, and 2.13, respectively).

Variations in the chromatic properties of wines observed at the end of the aging process have been associated with variations in the structural and chemical properties of the used wood [14, 32].

Our results indicated that P wines showed lower *a** CIELAB values, and colour intensity, but the highest luminosity, which could indicate that these barrels have larger pore size and/or lower content of ellagitannins, and both factors impact the oxidation processes, as well as the polymerization and condensation reactions of the anthocyanins [32]. In previous studies, we have already observed that P barrels had a finer grain than the rest of barrels used [17]. In fact, the OTR (oxygen transfer rate) is affected by grain size as smaller grains allow for greater oxygen influx. Moreover, ellagitannins derived from oak wood have a significant role

Table 4 Colour parameters and total polyphenol index of all the wines after 12 months of aging

		*L**	*a**	*b**	*C**	*h**	CI	T	TPI
Wine 1	K	53.80a	40.50 ab	2.10b	40.60ab	3.00c	18.30ab	0.68	91.75c
	M	55.90b	39.60a	2.20c	39.60a	3.10d	18.60b	0.68	91.15c
	O	53.60a	40.90b	2.10b	41.00b	2.90b	18.50b	0.67	88.92b
	P	56.40b	41.00b	2.00a	41.10b	2.80a	17.40a	0.67	88.30a
Wine 2	K	70.90a	24.90	3.30a	25.10	7.70a	10.50	0.80	89.75c
	M	71.50a	24.80	3.40b	25.00	8.20b	10.40	0.80	86.75b
	O	71.10a	24.80	3.30a	25.00	7.60a	10.50	0.79	91.92d
	P	74.30b	24.80	3.30a	25.00	7.70a	10.20	0.79	84.20a
Wine 3	K	63.80a	32.00b	2.40a	32.18b	4.20a	13.40ab	0.72	82.95b
	M	65.90b	30.50a	2.40a	30.60a	4.60b	13.90b	0.72	81.45a
	O	63.30a	31.90b	2.60b	32.00b	4.60b	13.60b	0.73	82.62b
	P	66.30b	31.90b	3.30c	32.10b	5.90c	12.70a	0.72	81.00a
Wine 4	K	71.14a	36.30	15.04a	39.29	22.51a	12.49	0.77	82.62b
	M	70.68a	36.72	15.35ab	39.79	22.69a	12.83	0.77	82.83b
	O	70.50a	36.22	15.70b	39.48	23.43b	12.88	0.79	81.95a
	P	73.66b	35.89	15.79b	39.21	23.74b	12.72	0.79	81.00a
Wine 5	K	69.54ab	38.71a	13.64a	41.05a	19.42	13.13a	0.72	71.08c
	M	68.59ab	40.18b	14.19b	42.57b	19.29	14.28b	0.71	70.68b
	O	67.91a	40.22b	14.01ab	42.59b	19.21	13.98b	0.71	69.13a
	P	70.35b	40.81b	14.07b	43.18b	19.05	13.80ab	0.70	68.75a
Wine 6	K	61.50a	31.50ab	3.90b	33.80c	7.10b	15.10b	0.76b	91.35a
	M	61.50a	32.90c	3.70a	33.10bc	6.40a	14.80ab	0.75a	93.45b
	O	61.60a	32.40bc	4.20c	32.60ab	7.40c	14.60ab	0.76ab	92.92ab
	P	64.60b	31.40a	5.90d	32.00a	10.60d	14.10a	0.78b	91.50a
Wine 7	K	11.66b	42.28bc	20.04c	46.79b	25.36b	10.85ab	0.81	65.10c
	M	12.64c	43.38c	21.70d	48.50c	26.57c	10.82ab	0.83	63.87b
	O	11.22a	41.83b	19.29b	46.07b	24.76b	11.01b	0.81	62.61a
	P	12.77c	39.14a	16.80a	42.60a	23.22a	10.28a	0.83	63.03ab
Wine 8	K	53.20ab	42.90	1.30b	42.90	1.70b	13.35ab	0.72	94.05d
	M	52.20a	41.80	1.80c	41.80	2.40d	13.82b	0.72	87.75a
	O	54.00b	42.40	1.10a	42.40	1.50a	13.54b	0.73	91.52c
	P	56.00c	42.80	1.30b	42.30	1.80c	12.49a	0.72	89.10b
Wine 9	K	62.39a	44.89	22.56	50.24	26.68	17.74	0.76	76.62
	M	61.73a	44.08	22.07	49.30	26.59	17.88	0.77	75.00
	O	62.16a	44.62	22.11	49.80	26.36	17.74	0.76	74.79
	P	64.91b	44.09	22.03	49.22	26.55	17.96	0.76	75.39
Wine 10	K	60.62b	43.43b	16.33b	46.40b	20.60b	17.79b	0.70a	70.88a
	M	61.92b	43.28b	16.84b	46.30b	20.80b	13.94a	0.70a	73.61b
	O	62.19b	43.10b	16.72b	46.23b	21.20b	17.13b	0.71a	72.66ab
	P	54.10a	36.22a	9.02a	38.18a	13.09a	17.17b	0.74b	78.16c
Wine 11	K	62.50ab	33.20a	2.00b	33.30a	3.40c	13.90a	0.70	81.35bc
	M	62.10ab	33.30a	2.10c	33.40a	3.60d	14.70b	0.70	82.35c
	O	61.70a	33.70a	1.90a	33.80a	3.30b	14.20ab	0.69	79.92b
	P	63.70b	35.40b	1.90a	35.40b	3.10a	14.10ab	0.68	78.30a
Wine 12	K	10.25b	40.77b	17.63b	45.42b	23.38b	11.63a	0.73	60.83
	M	7.92a	37.26a	13.62a	39.67a	20.08a	12.37b	0.74	61.29
	O	10.68c	41.31b	18.35c	45.21b	23.95b	11.48a	0.72	62.28
	P	10.74c	37.12a	13.33a	39.44a	19.75a	11.98ab	0.72	61.87

Different letters in the same column and the same wine indicate statistically significant differences ($p < 0.05$). Data from different wines have not been statistically compared

*L** lightness, a^* red-green colour component, *b** yellow-blue colour component, *C** chroma, *h** hue angle, *CI* colour intensity, *T* tonality, *TPI* total polyphenol index, *K* wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels

Table 5 Mean phenolic composition (mg/L) of the wines after 12 months of aging

	K	M	O	P
Hydroxybenzoic acids				
Gallic acid	86.95b	85.64b	85.11b	80.84a
Syringic acid	3.96d	3.65b	3.31a	3.81c
Ellagic acid	2.03c	1.98b	2.19d	1.87a
Total	92.94c	91.27bc	90.61b	86.52a
Hydroxycinnamic acids				
cis-Caftaric acid	4.87	4.82	4.88	4.91
trans-Caftaric acid	32.26b	31.67ab	31.95b	31.13a
cis-Coutaric acid	4.78c	4.6ab	4.69bc	4.52a
trans-Coutaric acid	27.061b	27.18b	27.38b	26.08a
Caffeic acid	6.858b	6.58a	7.64d	7.41c
trans-Fertaric acid	4.17a	4.32b	4.30b	4.25ab
p-Coumaric acid	3.45b	3.40b	3.28a	3.19a
Total	83.451b	82.57ab	84.14b	81.49a
Flavonols				
Myricetin-3-gal	1.95b	2.03c	1.84a	1.92b
Myricetin-3-glc	11.17b	11.05b	11.28b	10.68a
Quercetin-3-gal	4.06bc	3.87a	4.10c	3.97b
Quercetin-3-glc	4.25b	4.22a	4.07b	4.04ab
Quercetin-3-glcU	3.18b	3.02a	3.37b	3.26ab
Isorhamnetin-3-glc	1.86b	1.84ab	1.87b	1.81a
Myricetin	11.24ab	11.08ab	11.30b	10.99a
Quercetin	6.85b	6.78b	6.77b	6.58a
Kaempferol	1.00b	0.98ab	0.97a	0.99ab
Isorhamnetin	0.56	0.54	0.56	0.55
Total	46.11b	45.42ab	46.12b	44.77a
Flavan-3-ol				
Catechin	30.82b	31.14b	30.59b	29.91a
Stilbenes				
trans-Piceid	1.60a	1.66b	1.61ab	1.56a
trans-Resveratrol	1.22b	1.25b	1.25b	1.06a
Total	2.82b	2.91c	2.86bc	2.62a
Ellagitannins	10.10c	9.43b	10.32c	8.96a
Anthocyanins				
Delphinidin-3-glc	33.10b	34.09c	33.13b	30.97a
Cyanidin-3-glc	2.46b	2.46b	2.65c	2.19a
Petunidin-3-glc	29.22b	29.96c	29.55bc	27.83a
Peonidin-3-glc	7.81b	8.00c	7.95bc	7.45a
Malvidin-3-glc	91.45b	93.52c	91.86bc	86.66a
Delphinidin-3-acglc	1.41b	1.39b	1.55c	1.28a
Cyanidin-3-acglc	1.15c	1.12b	1.24d	1.07a
Petunidin-3-acglc	1.47b	1.39a	1.62c	1.45b
Peonidin-3-acglc	0.75c	0.69a	0.80d	0.73b
Malvidin-3-acglc	5.88c	5.84c	5.65b	5.48a
Delphinidin-3-cmglc	2.79b	2.93d	2.86c	2.72a
Cyanidin-3-cmglc	0.64b	0.64b	0.62a	0.64b
Petunidin-3-cmglc	2.70b	2.79c	2.65ab	2.63a
Peonidin-3-cmglc	1.46b	1.56c	1.37a	1.37a
Malvidin-3-cmglc	10.09b	10.43c	10.15b	9.46a
Total	192.38b	196.82c	193.66bc	181.91a

Mean values of the 12 wines are shown for each barrel origin. For each parameter, values with different letters are significantly different between the samples ($p < 0.05$)

glc glucoside, *acglc* acetyl-glucoside, *cmglc* coumaroyl-glucoside, *gal* galactoside, *glcU* glucuronide, *K* wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels

Table 6 Composition of phenolic families (mg/L) of all the wines after 12 months of aging

		HB acids	HC acids	Flavonols	Flavan-3-ol	Stilbenes	Ell	Ant
Wine 1	K	92.37c	74.55c	64.90b	30.82	1.16b	10.93c	222.6b
	M	69.60a	56.74a	51.90a	31.14	0.85a	9.67b	205.1a
	O	81.54b	67.08b	60.94b	30.59	1.41c	11.70c	192.9a
	P	81.68b	66.65b	59.87b	29.91	1.42c	7.53a	189.6a
Wine 2	K	117.0ab	76.79	44.30	31.71	1.42ab	13.28	331.0ab
	M	121.7b	75.83	45.79	29.03	1.53b	13.07	358.3c
	O	116.5ab	78.60	46.01	31.62	1.99c	13.78	340.7bc
	P	106.3a	72.81	41.93	28.51	1.34a	13.69	310.0a
Wine 3	K	75.59b	58.96a	52.52b	30.13	0.86a	6.61a	204.0c
	M	73.40b	59.77a	53.34b	31.07	0.90a	7.42a	226.4d
	O	72.98b	68.14b	51.77b	28.93	0.97a	9.85b	184.8b
	P	61.77a	57.66a	41.52a	29.56	1.80b	7.09a	148.6a
Wine 4	K	107.7	104.2	63.34	48.19c	4.11a	12.53a	315.7
	M	108.4	103.5	62.23	36.38a	5.61b	17.16c	296.4
	O	104.9	102.3	61.90	41.79b	4.43a	14.59b	312.1
	P	107.8	103.7	61.30	37.69a	4.43a	14.96b	293.9
Wine 5	K	89.08	102.7	45.17	42.27	6.40	8.58c	199.2b
	M	85.46	102.8	46.00	40.07	6.08	4.02a	187.4ab
	O	83.81	104.0	44.23	42.63	6.24	6.22b	187.4ab
	P	84.10	102.7	44.53	41.36	6.12	8.37c	180.1a
Wine 6	K	96.21	92.74	39.19	34.62	2.00b	7.75a	92.90b
	M	99.66	101.0	42.87	35.17	2.04b	9.67b	123.8d
	O	100.2	100.1	40.36	29.33	1.26a	8.52a	114.20c
	P	100.1	97.41	39.14	30.48	1.26a	8.56a	83.46a
Wine 7	K	75.28	75.01	34.90	27.23	3.10b	9.43c	303.0
	M	74.52	74.52	33.75	27.75	2.86ab	7.82b	310.4
	O	70.19	73.85	34.76	26.23	2.73a	6.98b	292.6
	P	71.27	73.78	34.82	28.02	2.91ab	5.51a	309.9
Wine 8	K	61.12	101.5	63.14	29.82	1.44a	6.71a	200.2ab
	M	61.18	102.8	63.27	28.42	2.27c	6.74a	212.3bc
	O	58.68	103.1	64.55	34.02	2.14bc	9.33b	222.2c
	P	58.04	100.9	62.47	32.56	2.03b	6.46a	191.1a
Wine 9	K	84.71	93.68	47.17	24.83c	2.66b	14.90b	46.51c
	M	83.06	93.67	45.64	19.88b	2.07a	10.98a	31.31a
	O	81.50	92.07	45.95	17.37a	2.84b	11.97a	36.96b
	P	81.39	91.61	45.47	23.21c	2.60b	11.83a	32.91a
Wine 10	K	101.3b	82.59	31.62a	13.13a	4.94bc	8.15b	57.30a
	M	106.2b	83.15	34.78ab	31.14d	4.71b	5.52a	91.40b
	O	102.9b	82.83	36.74b	17.91c	5.21c	8.62b	116.0c
	P	86.52a	81.49	44.77c	15.70b	2.62a	8.96b	181.9d
Wine 11	K	99.71b	70.43b	37.27b	27.22	1.87b	11.86c	210.9c
	M	97.71b	68.49b	36.29b	32.01	2.40c	11.84c	204.0bc
	O	96.70b	69.29b	36.11b	29.05	1.70b	10.57b	190.6b
	P	87.29a	61.02a	32.72a	28.44	1.44a	8.18a	160.8a
Wine 12	K	115.3	68.15	29.81	29.26a	3.88b	10.50c	125.2c
	M	114.3	68.51	29.24	31.62ab	3.63ab	9.28b	115.2b
	O	117.5	68.33	30.17	37.60c	3.37a	11.67d	133.5c
	P	112.0	68.13	28.73	33.48b	3.45a	6.37a	100.8a

Different letters in the same column and the same wine indicate statistically significant differences ($p < 0.05$). Data from different wines have not been statistically compared

Ant anthocyanins, *HB acids* hydroxybenzoic acids, *HC acids* hydroxycinnamic acids, *Ell* ellagitannins, *K* wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels in wine colour properties. They function as antioxidants, due to their capability to consume large amounts of oxygen, thus regulating the oxidation reactions [32].

Although no significant differences were observed in mean TPI values (Table 3), K, M, O and P wines showed differences in TPI in 10 of the wines analyzed (Table 4), indicating that barrel origin significantly affected the TPI of the wines. Wines aged in K barrels showed the highest TPI in 6 of the wines studied (Table 4), while P wines showed the lowest value in the majority of the wines.

Phenolic composition of the wines after 12 months of aging

Table 5 shows the mean content of phenolic compounds of the 12 wines aged in K, M, O and P barrels for 12 months. Table 6 shows the individual values of all the wines from the 12 wine cellars. Data of individual phenolic compounds are shown in Annex Table 8.

Results showed that the influence of the geographic origin of *Q. alba* on the monomeric phenolic compounds was mainly significant for anthocyanins, stilbenes and ellagitannins. Barrel origin significantly affected the content of anthocyanins in 10 of the 12 wines studied, and ellagitannin and stilbene content was affected in 11 of the wines (Table 6). However, barrel origin only influenced the content of hydroxybenzoic acids and hydroxycinnamic acids in 5 and 3 of the wines studied, respectively, and flavonol and flavan-3-ols content showed differences in only 4 of the 12 wines (Table 6).

In general, P wines showed the lowest concentration of total hydroxycinnamic, hydroxybenzoic acids, flavonols, stilbenes, ellagitannins, and anthocyanins, both in mean and individual data (Tables 5 and 6), and K and O wines showed in total the highest concentration of ellagitannins in 8 of the wines (Table 6). This may be explained by the different types of compounds that are released from wood originating from different geographic regions [7, 12]. Moreover, the content of low molecular weight compounds and their transfer to the wine have been associated with the amount of oxygen that the wines are exposed to during the process of wood aging [38], suggesting our results a different oxygen permeability among barrels.

Regarding the concentrations of anthocyanins, a decreased was observed in most of the wines after 12 months of aging (Tables 2 and 6). The mean anthocyanin content decreased 17 to 19% in K, M and O wines, and 24% in P wines (Tables 2 and 5). The reduction in the concentration of anthocyanins in red wines aged in oak wood barrels has also been reported in the bibliography [39]. Our results indicated that there were more aging reactions leading to a decrease in anthocyanins in P wines than in K, M and O wines. Therefore, P wines showed a quicker evolution than wines from the other barrels because there was a higher loss of anthocyanins. Therefore, P wines showed significantly lower values of total anthocyanins in 9 of the wineries studied (Table 6), which was related to their lowest colour intensity, C^* and a^* values (see Sect. "Colour parameters and total polyphenol index of the wines after 12 months of aging").

Regarding stilbenes, the mean concentration of *trans*-resveratrol and its glucoside decreased during oak aging (Tables 2 and 5). The decrease in the content of stilbenes, attributed in the bibliography to their enzymatic conversion to their isomers *cis*-resveratrol and *cis*-resveratrol glucoside [40] and to adsorption processes on the oak surface [5], was more accused in P wines (23 to 26% for K, M and O wines vs 32% for P wines) (Tables 2 and 5). After 12 months of aging, wines aged in P barrels had the lowest contents of stilbenes in the majority of the wines, which could indicate a higher adsorption capacity of P barrels.

Wines from K were the richest in ellagitannins in 6 of the 11 wines which showed significant differences (Table 6), probably due to the different content in the heart wood, which undergo alterations based on the different geographic origin [7, 11]. Once ellagitannins are extracted into wine, they undergo a series of reactions, e.g. reacting with anthocyanins or flavanols to form anthocyanin-ellagitannin or flavano-ellagitannins; consuming oxygen, regulating oxidation reactions due to their strong antioxidant properties; and they can also be hydrolyzed [4, 41]. Our results seemed to indicate that the release of ellagitannins from the barrels was higher that its consumption by participating in the reactions resulting in an increase in their concentration. The lowest ellagitannin content in P wines (Tables 5 and 6) may be due to a higher oxygen permeability of the barrel or to the fact that the barrel releases less ellagitannins. The higher oxygen supply in P wines could result in more intense oxidation reactions. Thus, ellagitannins could be more easily oxidized, and as result, the decrease in their concentration would be more pronounced than in others barrels. Our previous results indicated that P barrels probably had a finer grain [17], and thus, they would be more permeable to oxygen.

As observed in bibliography [2, 38], the most abundant benzoic acid was gallic acid and its content was higher in aged wines than in non-aged wines (Tables 2 and 5). Gallic acid may be released by hydrolysis of gallate esters from hydrolysable tannins present in the wood and exhibits strong antioxidant properties even at very low concentrations [42]. Generally, a higher content of gallic acid was found in K, M and O wines (Annex Table 8). Recent studies have shown that the wines aged in low OTR barrels extracted more gallic acid from the wood, and at a faster rate when compared to wines aged in high

OTR barrels. This may be attributed to the fact that wood classification by OTR involves a distinction of compositional and anatomical and traits [38]. Ellagic acid increased in the wines during oak aging because of the contact with wood (Tables 2 and 5). The most pronounced increase was observed in O and K wines. The oak wood has a high content of water-extractable ellagitannins, which can be extracted into wine during the oak aging process. However, the mean content of syringic acid decreased with respect to the initial wines (Tables 2 and 5), being the wines aged in O barrels the ones with the lowest concentrations in 6 of the 12 wines (Annex Table 8). A decrease in the concentrations of syringic acid during oak aging has been previously reported [43]. The decrease in the concentrations of syringic acid may be attributed to the equilibration of concentrations between the oak wood and the wine. Additionally, adsorption on the barrel surface, as well as a relatively slow penetration of phenolic compounds into the oak barrels could occur during the wine aging process [43]. The reduction of syringic acid during oak aging could also be attributed to enzymatic or chemical degradation [43].

Hydroxycinnamic acids evolved differently during oak aging. Fertaric acid content increased in all the wines (Tables 2 and Annex Table 8), probably due to aging in oak barrels favors the esterification processes [44]. Caftaric and coutaric acids diminished or maintained their concentration in most of the wines (Tables 2 and Annex Table 8), as they are described to be highly reactive compounds that contribute to oxidation processes [42]. An increase in the mean value of caffeic and *p*-coumaric acids was also observed in the wines during aging (Tables 2 and 5). The increase in the content of caffeic and *p*-coumaric acids could be attributed to various factors, including the hydrolysis of hydroxycinnamoyl-tartaric acid grape precursors in wine to produce caffeic and *p*-coumaric acids. Additionally, the release of these acids from oak wood and their consumption during the formation of pyranoanthocyanins, which result from the reaction between free hydroxycinnamic acids and anthocyanins, may also contribute to this phenomenon [45].

Regarding barrel origin, P wines exhibited the lowest mean concentrations of hydroxycinnamic acids (Table 5), but only 3 wines showed significant differences when considering the individual wines (Table 6), indicating that barrel origin had very little effect on these compounds.

With regards to the flavonol content, quercetin, and myricetin-type flavonols were the most abundant in all the wines. A decrease in the mean values of both flavonol glycosides and aglycones content was observed after 12 months of aging (Tables 2 and 5). As previously reported [1], these results suggest that flavonol glycosides underwent significant acid hydrolysis during the oak aging process. However, an increase in the aglycone content was not observed, probably due to the involvement of flavonols in oxidation and condensation reactions, as well as their insolubility [46]. The mean content of catechin also decreased in the wines during aging (Tables 2 and 5). Flavan-3-ol content in the wines after 12 months of aging was in the range described by other authors [14, 42]. The loss of catechin during wine barrel aging may be attributed to its participation in polymerisation and condensation reactions with anthocyanins, which are favored by the diffusion of oxygen from the barrels and its adsorption on the wood surface [38]. Regarding barrel origin, the higher mean content of flavanols and flavan-3-ols were obtained in K, M and O wines (Table 5). However, only 4 wineries showed significant differences in these compounds when considering the individual wines (Table 6), indicating that barrel origin practically did not affect the content of these compounds.

The results of this research revealed that the origin of barrels made of *Quercus alba* had a great effect on the content of anthocyanins, stilbenes and ellagitannins but it practically did not affect the rest of the phenolic compounds. The lower content of TPI and phenolic compounds in P wines suggested a slower release of compounds during aging, and/or a higher adsorption capacity and/or oxidation level than barrels from K, M and O origins. By contrast, the higher ellagitannin concentration of K and O wines would provide greater colour stability during wine maturation and aging, protecting the wine against oxidation.

Wine sensory analysis after 12 months of aging

K, M, O and P wines were tasted separately for each winery. ANOVA was applied to the mean data of the 12 wines to analyze differences among wines according to barrel origin. All the sensory attributes evaluated showed GM > 40% (data not shown), so all of them were included in the statistical analysis. The ANOVA results are shown in Fig. 1. Figure 1 shows the mean values of the visual and gustatory phases of sensory analysis of Tempranillo wines aged for 12 months in K, M, O, and P oak barrels. Figure 2 shows the heatmap plot of all the individual wines identifying in red the highest values for each wood origin.

The Tempranillo wines used in this study were characterized by high colour intensity, limpidity, and brightness (Fig. 1A). Regarding the judge's comments on colour, it is worth noting that the wines were described as exhibiting ruby and cherry red tonalities.

Despite the wines aged in the different barrels showed significant differences in the colorimetric parameters (see Sect. "Colour parameters and total polyphenol index of the wines after 12 months of aging"), no significant differences were observed in any descriptor of visual phase when analyzing the mean values (Fig. 1A). Expect for brightness, the origin of the barrel also showed no clear effect on the visual parameters of colour when considering the individual results of the 12 wines (Fig. 2). This fact may be attributed to the diminished ability of tasters to discern colour variations of up to five units in a to Δ*E** when observing the wine through a glass [47]. M wines showed the highest brightness scores in 7 of the 12 wines studied.

Regarding the gustatory phase (Fig. 1B), wines showed high values of smoothness, tannin level, body, length, and balance. Acidity, tannin level, astringent tannin, and balance descriptors showed significant differences among wines aged in barrels from different origins. P wines achieved the mean highest scores in astringent tannins. The higher astringent tannin perceived in P wines increased the perception of acidity with respect to other wines. These results agreed with the sensory data of the individual wines. Therefore, P wines achieved the highest punctuations in astringent tannins and acidity in 6 and 7 of the 12 wines, respectively (Fig. 2).

K and O wines showed the mean highest punctuations of tannin level, and the highest values for balance descriptor (Fig. 1B), in agreement with the individual data where K and O wines showed in total the highest values of tannin level in 8 of the wines, and ripe tannin and balance in 10 and 11 of the wines, respectively (Fig. 2). Wines from P barrels showed the lowest mean scores for the balance descriptor. The higher levels of tannins and ripe tannins in O and K wines made wines more appreciated by judges for their higher balance. The lower content of hydroxybenzoic acids, hydroxycinnamic acids, flavonols, flavan-3-ol, stilbenes, ellagitannins, and anthocyanins in P wines, and their higher values in astringent tannins and acidity, could have led to a lower score for the balance descriptor.

Finally, the tasting panel assessed the overall perception for both the visual and gustatory phases. All wines obtained high punctuations for global perception in the visual phase, 3.64, 3.63, 3.70 and 3.68 for K, M, O, and P, respectively, and no significant differences were observed (data not shown). However, in the global perception of the gustatory phase, the wines differed significantly as the panelist rating for overall acceptance ranged from 3.64 to 2.85. K and M wines had the highest scores (3.64 and 3.26, respectively), followed by the O wines (3.09), and finally, the P wines (2.85) (data not shown). Figure 2 shows that wines aged in K barrels were the bests valued un the gustatory phase in 9 of the wines tasted while M wines were the best valued in 3 of the 12 wines. It is important to highlight that a previous work of our research group showed that the K wine, together with the M wine, were the most highly valued regarding global olfactory perception [17], indicating that these forests would be the most suitable for making barrels providing wines with higher sensory qualities.

Classification of the wines according to their initial phenolic composition and effect of barrel origin

To evaluate the influence of the barrel forest origin according to the initial phenol composition of the wines, the wines were grouped by their initial polyphenol content (measured by HPLC–DAD), TPI and CI parameters. The clustering analysis was performed by the so-called k-means method, for k = 3 groups. The number of groups was previously set by the authors but also confirmed as appropriate by the "elbow" method.

Group 1 was composed of wines with the highest content of polyphenols (870.35 mg/L) as well as the highest value of TPI (90.94) and CI (18.85). Group 2 of wines was characterized by a low content of polyphenols (545.70 mg/L) and the lowest TPI (74.54) and CI (13.11). Group 3 of wines was characterized by low content of polyphenols (538.24 mg/L) and high TPI (82.85) and CI values (18.41).

Table 7 shows the concentration of phenolic compounds in the different groups of wines. MANOVA analysis confirmed the impact of the barrel´s origin, wine, and their interaction. The effect of the origin of the barrel (percentage of variance calculated by MANOVA analysis) increased when the wines were classified according to their initial phenolic (Table 7). As previously stated, the content of ellagitannins was the most affected by the barrel origin. Generally, the barrel origin had a greater influence on the wines with the highest content of phenolic compounds, and CI and TPI values. When considering all the wines, P barrels produced the wines with the lowest content of phenolic compounds (hydroxycinnamic acids, hydroxybenzoic acids, flavanols, flavan-3-ol, stilbenes, ellagitannins and anthocyanins) and CI values (Tables 3 and 5). K and O wines showed the highest content of ellagitannins (Table 5), which would enhance the colour stability of the wines during maturation and aging and protecting the wine against oxidation. This same effect was observed in the wines from group 2 and 3, but not in group 1. Wines forming group 2 were characterized by the lowest initial TPI and CI values. In this group, the effect of the barrel origin on the phenolic composition was the same as it was observed when considering all the wines. Wines from group 3 showed intermediate values of TPI and CI. In this group, P wines showed low contents of hydroxycinnamic acids, hydroxybenzoic acids, and stilbenes, and K and O wines had again the highest content of ellagitannins. In

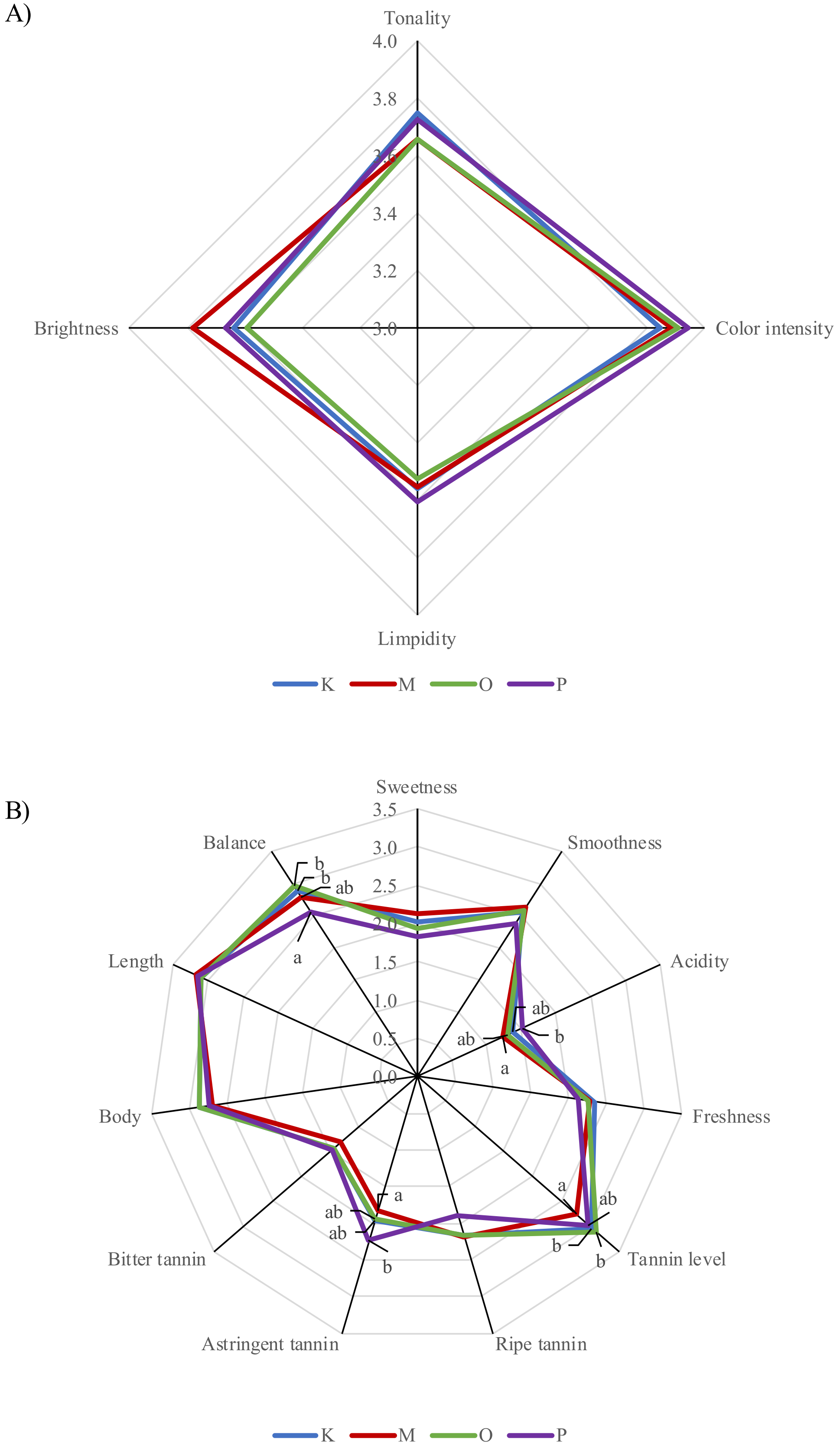

Fig. 1 Visual (**A**) and gustatory (**B**) phases of sensory analysis of the Tempranillo wines aged for 12 months in oak barrels from Kentucky, Missouri, Ohio and Pennsylvania forests. Mean values of the 12 wines are shown. For each parameter, values with different letters are significantly different between the samples ($p \leq 0.05$). Blue line: wines aged in Kentucky barrels; red line: wines aged in Missouri barrels; Green line: wines aged in Ohio barrels; Violet line: wines aged in P barrels

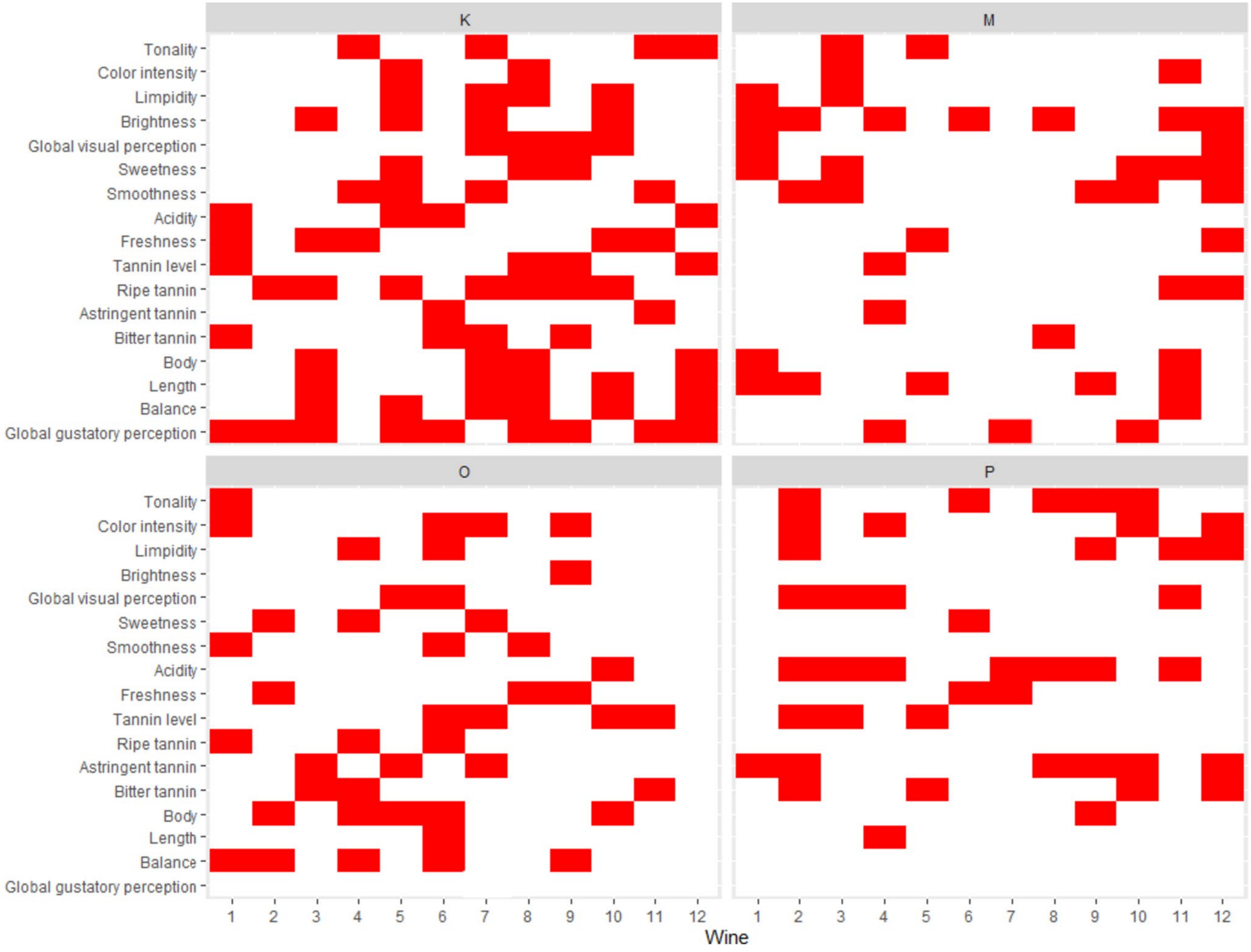

Fig. 2 Heatmap plots identifying for each barrel origin the highest values of the visual and gustatory attributes of the wines after 12 months of aging. The rows in the heatmap represent sensory attributes and the columns indicate wines. Red colour of the heatmap cells indicates the highest value of parameter across different samples. K: wines aged in Kentucky barrels; M: wines aged in Missouri barrels; O: wines aged in Ohio barrels; P: wines aged in P barrels

group 1 of wines, characterized by the highest initial polyphenol content, TPI and CI values, no significant differences were observed in the total content of hydroxybenzoic acids, hydroxycinnamic acids, flavanols and anthocyanins among the wines aged in the different barrels. However, wines aged in M barrels had the highest content of ellagitannins and stilbenes, indicating that M barrels could be the most appropriate for aging wines with a higher content of total polyphenols. Ellagitannins possess natural antioxidant properties and can protect other wine compounds, as stilbenes, from oxidation by reacting effectively with oxygen. Regardless of the origin of the barrel, the content of ellagitannins was the highest in wines of group 1. The content of ellagitannins would depend on an equilibrium between their release form the wood and its consumption, which is liked to its oxidation. Nikolantonaki and co-workers [48] observed that the time required for reaching the highest concentration of ellagitannin in the wine is related to its matrix.

Conclusions

This work studied the phenolic composition, colour parameters, and visual and gustatory attributes of Tempranillo red wines after 12 months of aging in *Quercus alba* oak barrels from Missouri (M), Ohio (O), Kentucky (K) and Pennsylvania (P) forests. The results showed that the forest origin of *Quercus alba* affected the colour parameters of the wines, except for tonality, the total polyphenol index, and the phenolic composition of the aged red wines. Hence, barrel origin significantly affected the content of anthocyanins, stilbenes and ellagitannins but had little effect on the rest of phenolic

Table 7 Composition of phenolic families (mg/L) in the 3 groups of wines after 12 months of aging. Multivariate analysis of variance and percentage of variance attributable (%) to barrel origin, wine and the interaction of both (barrel origin × wine)

	Compound	K	M	O	P	Barrel origin (%)	Wine (%)	Barrelorigin × wine (%)
Group 1	Hydroxybenzoic acids	110.83 ± 7.14a	111.59 ± 7.19a	107.99 ± 6.96a	110.95 ± 7.15a	4.36	22.65	0.00
	Hydroxycinnamic acids	107.31 ± 7.05a	106.58 ± 7.00a	105.31 ± 6.92a	106.82 ± 7.02a	1.31	22.85	0.00
	Flavonols	65.21 ± 4.21a	64.07 ± 4.13a	63.73 ± 4.11a	63.11 ± 4.07a	3.94	22.75	0.00
	Flavan-3-ol	49.62 ± 3.26c	38.81 ± 2.55a	43.04 ± 2.83b	37.46 ± 2.46a	77.54	5.14	0.06
	Stilbenes	4.23 ± 0.27a	5.77 ± 0.37b	4.55 ± 0.29a	4.56 ± 0.22a	81.65	4.34	0.07
	Ellagitannins	12.89 ± 0.81a	17.66 ± 1.11c	15.00 ± 0.95b	15.40 ± 0.97b	78.55	5.11	0.06
	Anthocyanins	325.19 ± 21.84a	305.26 ± 20.50a	321.43 ± 21.59a	302.70 ± 20.33a	20.58	17.82	0.02
Group 2	Hydroxybenzoic acids	96.57 ± 19.38b	96.32 ± 20.96b	94.78 ± 21.47b	87.72 ± 20.41a	3.19	91.73	1.32
	Hydroxycinnamic acids	69.87 ± 7.01b	69.52 ± 6.45b	71.64 ± 5.00b	66.68 ± 7.08a	7.54	67.54	9.29
	Flavonols	39.76 ± 8.36b	39.68 ± 9.18b	39.76 ± 8.38b	35.94 ± 5.51a	4.34	86.39	5.07
	Flavan-3-ol	29.79 ± 1.85a	30.24 ± 2.18ab	31.12 ± 3.96b	30.55 ± 1.87ab	3.54	49.58	27.88
	Stilbenes	2.22 ± 1.15ab	2.26 ± 1.00b	2.15 ± 0.87a	2.18 ± 0.88ab	0.19	90.02	8.50
	Ellagitannins	10.34 ± 2.41c	9.89 ± 2.35b	10.57 ± 2.37c	8.16 ± 3.03a	12.69	68.47	15.36
	Anthocyanins	234.83 ± 77.14b	242.83 ± 88.20c	228.43 ± 79.33b	206.01 ± 90.56a	2.77	94.10	2.21
Group 3	Hydroxybenzoic acids	87.46 ± 13.76b	84.20 ± 16.50a	84.76 ± 15.38ab	81.97 ± 13.26a	1.82	83.64	8.71
	Hydroxycinnamic acids	91.30 ± 10.85a	90.03 ± 17.24a	91.52 ± 13.96a	90.13 ± 13.43a	0.24	89.23	4.46
	Flavonols	49.53 ± 12.55ab	47.41 ± 9.20a	48.79 ± 10.84ab	49.37 ± 9.12b	0.49	88.24	7.38
	Flavan-3-ol	28.78 ± 9.03a	28.33 ± 8.08a	28.27 ± 8.97a	30.75 ± 6.14b	1.60	82.81	13.67
	Stilbenes	3.09 ± 1.99bc	3.00 ± 1.85b	3.18 ± 1.95c	2.67 ± 1.67a	1.11	92.03	6.12
	Ellagitannins	9.50 ± 2.85c	7.77 ± 2.61a	9.39 ± 2.09c	8.61 ± 1.75b	8.42	62.54	25.57
	Anthocyanins	136.45 ± 74.94a	141.89 ± 68.10b	144.95 ± 64.64b	143.16 ± 63.97b	0.23	89.03	10.15

For each parameter values with different letters are significantly different between the samples ($p < 0.05$)

K wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels

compounds. Wines aged in P barrels showed the lowest colour intensity and content of most phenolic families and ellagitannins, but the highest luminosity. Wines aged in K and O barrels showed the highest content of ellagitannins. The forest origin of wood did not affect the visual attributes of the wines but produced important effects on their gustatory profile. Wines aged in K and O barrels showed the highest content of tannin level and ripe tannins, and were the most balanced. K wines were the best valued in the overall perception of the gustatory sensory analysis, followed by M wines. P wines were characterized by acidity and astringent tannins and obtained the lowest score in the overall perception of the gustatory phase. Finally, the influence of the barrel origin on the phenolic composition of the wines was evaluated according to their initial phenolic composition. Wines with the highest initial CI and phenol content had the highest content of ellagitannins after barrel aging. The results of the present paper indicated that K barrels were the most appropriate for the aging of Tempranillo wines, as was previously observed by our work group in terms of aromatic quality.

Appendix

See Table 8.

Table 8 Composition of individual phenolics (mg/L) of all the wines after 12 months of aging

Compounds	Wine 1				Wine 2				Wine 3				Wine 4			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Hydroxybenzoic acids																
Gallic acid	83.20c	62.67a	75.39b	75.01b	109.5ab	115.5b	111.4b	100.0a	69.70b	68.10b	67.70b	57.40a	102.5	103.3	99.38	102.3
Syringic acid	7.60c	4.73ab	4.54a	5.17b	5.21d	3.65b	2.01a	4.16c	4.28b	4.03b	4.14b	3.33a	1.68b	1.83b	1.47a	1.71b
Ellagic acid	1.57a	2.20b	1.61a	1.50a	2.30a	2.57b	3.15c	2.10a	1.61c	1.27b	1.14a	1.04a	3.51ab	3.24a	4.05c	3.74bc
Total	92.37c	69.60a	81.54b	81.68b	117.0ab	121.7b	116.5ab	106.3a	75.59b	73.40b	72.98b	61.77a	107.7	108.4	104.9	107.8
Hydroxycinnamic acids																
cis-Caftaric acid	7.22c	5.23a	6.42b	6.19b	4.43a	4.85b	4.97b	4.24a	5.96b	5.87b	5.70b	4.95a	4.78b	4.68ab	4.35a	4.84b
trans-Caftaric acid	31.34c	23.57a	27.51b	28.4b	29.99ab	31.39b	31.44b	28.30a	18.30b	18.00b	19.10b	15.80a	42.44	42.21	41.75	41.79
cis-Coutaric acid	7.07c	5.00a	5.95b	6.07b	4.89a	5.31a	7.05b	5.17a	4.58b	4.47b	4.42b	3.97a	5.97ab	6.04ab	6.39b	5.55a
trans-Coutaric acid	19.86b	15.45a	18.91b	18.33b	24.52b	22.33a	22.32a	22.41a	12.60b	12.40b	11.80ab	10.90a	41.88	41.75	41.27	41.60
Caffeic acid	3.33c	2.04a	2.56b	2.24a	6.08b	4.62a	5.66b	5.62b	5.74a	5.60a	16.10c	13.70b	3.57a	3.61a	3.61a	4.43b
trans-Fertaric acid	3.83	3.80	3.76	3.51	2.08ab	2.18b	1.91a	2.54c	2.20a	3.81b	3.78b	2.34a	5.00bc	4.61ab	4.57a	5.06c
p-Coumaric acid	1.90b	1.65a	1.97b	1.91b	4.80ab	5.15bc	5.25c	4.53a	9.58c	9.62c	7.24b	6.00a	0.57c	0.61c	0.35a	0.47b
Total	74.55c	56.74a	67.08b	66.65b	76.79	75.83	78.60	72.81	58.96a	59.77a	68.14b	57.66a	104.2	103.5	102.3	103.7
Flavonols																
Myricetin-3-gal	3.10	2.81	2.97	2.87	1.33b	2.21d	0.62a	1.55c	4.16d	3.60c	2.70a	3.17b	2.06b	1.80a	1.80a	1.96ab
Myricetin-3-glc	16.17c	12.72a	14.22b	14.22b	14.83b	15.30b	16.08b	13.17a	13.30b	14.00b	13.90b	10.60a	9.77	9.41	9.83	9.11
Quercetin-3-gal	7.64b	6.23a	7.62b	7.30b	4.06bc	3.43a	4.43c	3.90b	3.24b	2.85a	3.20b	2.62a	6.94	6.71	6.54	6.60
Quercetin-3-glc	5.01c	3.88a	4.31b	4.45b	3.55	3.53	3.63	3.51	6.69b	6.59b	6.46b	5.48a	4.26	4.31	4.10	4.03
Quercetin-3-glcU	3.74b	2.78a	3.57b	3.59b	2.66ab	2.53a	3.00c	2.83bc	5.01bc	4.71ab	5.34c	4.43a	3.18	3.09	3.39	3.25
Isorhamnetin-3-glc	2.06c	1.62a	1.87b	1.94bc	1.36	1.41	1.41	1.29	2.40b	2.41b	2.39b	2.02a	1.38a	1.41a	1.74b	1.65b
Myricetin	17.31b	14.43a	17.17b	17.34b	8.11ab	8.79b	8.11ab	7.79a	9.17b	9.56b	9.30b	7.91a	19.99	19.96	19.82	20.11
Quercetin	8.59b	6.53a	8.01b	6.98a	6.81	7.04	7.07	6.42	7.25b	7.98c	7.22b	4.63a	12.69	12.51	11.67	11.62
Kaempferol	0.62c	0.41a	0.56b	0.58bc	1.23	1.19	1.24	1.13	0.71b	1.01c	0.73b	0.26a	2.32	2.29	2.25	2.24
Isorhamnetin	0.66c	0.49a	0.64bc	0.60b	0.36a	0.36a	0.42b	0.34a	0.59c	0.63c	0.53b	0.40a	0.74	0.75	0.76	0.73
Total	64.90b	51.90a	60.94b	59.87b	44.30	45.79	46.01	41.93	52.52b	53.34b	51.77b	41.52a	63.34	62.23	61.90	61.30
Flavan-3-ol																
Catechin	30.82	31.14	30.59	29.91	31.71	29.03	31.62	28.51	30.13	31.07	28.93	29.56	48.19c	36.38a	41.79b	37.69a
Stilbenes																
trans-Piceid	0.55b	0.45a	0.76c	0.50ab	0.31a	0.39b	0.52c	0.30a	0.50a	0.50a	0.50a	1.38b	2.15a	3.55c	2.42ab	2.46b
trans-Resveratrol	0.61b	0.40a	0.65b	0.92c	1.11a	1.14a	1.47b	1.04a	0.36a	0.40ab	0.47c	0.42b	1.96	2.06	2.01	1.96
Total	1.16b	0.85a	1.41c	1.42c	1.42ab	1.53b	1.99c	1.34a	0.86a	0.90a	0.97a	1.80b	4.11a	5.61b	4.43a	4.43a
Ellagitannins	10.93c	9.67b	11.70c	7.53a	13.28	13.07	13.78	13.69	6.61a	7.42a	9.85b	7.09a	12.53a	17.16c	14.59b	14.96b
Anthocyanins																
Delphinidin-3-glc	40.61c	36.06b	31.38a	31.86a	66.49bc	71.02c	64.38b	58.96a	27.30b	32.10c	26.10b	19.60a	57.97b	53.61ab	57.66ab	53.40a

Table 8 (continued)

Compounds	Wine 1				Wine 2				Wine 3				Wine 4			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Cyanidin-3-glc	1.77b	1.59a	3.31c	1.87b	3.80a	5.10b	5.71c	3.48a	2.20b	2.25b	2.16b	1.30a	5.02b	4.66a	4.50a	4.45a
Petunidin-3-glc	34.85c	32.03b	29.85ab	29.45a	50.57ab	54.88c	52.65bc	48.09a	28.70c	31.80d	26.10b	21.20a	52.23	49.37	52.02	48.85
Peonidin-3-glc	6.61b	6.44b	5.86a	5.69a	11.78ab	12.84c	12.22bc	11.17a	7.02c	8.08d	6.39b	5.13a	10.40bc	9.73ab	10.90c	9.51a
Malvidin-3-glc	107.0b	99.83ab	92.94a	92.31a	151.3ab	164.6c	157.3bc	143.7a	103.0c	115.0d	93.70b	77.10a	143.7	134.9	141.4	133.3
Delphinidin-3-acglc	1.18ab	1.10a	2.83c	1.26b	1.80ab	1.98c	1.94bc	1.76a	1.66b	1.60b	1.40a	1.35a	2.80c	2.56ab	2.73bc	2.47a
Cyanidin-3-acglc	1.95b	1.31a	2.39c	1.82b	1.75c	1.45b	1.81c	1.03a	1.99c	1.64b	1.56b	0.99a	0.46a	0.55c	0.50b	0.43a
Petunidin-3-acglc	2.39b	1.70a	2.39b	2.48b	2.10c	1.91b	2.79d	1.70a	2.34d	1.75c	1.56b	1.28a	2.47	2.46	2.32	2.40
Peonidin-3-acglc	0.95c	0.73a	0.87b	1.09d	0.77c	0.59b	0.48a	1.04d	1.37b	1.62c	1.43b	0.61a	0.89c	0.32a	0.53b	0.84c
Malvidin-3-acglc	7.22c	6.18b	5.50a	6.10b	8.29	8.38	8.07	8.28	9.04c	9.18c	8.06b	6.57a	8.39	8.40	8.36	8.36
Delphinidin-3-cmglc	3.47b	3.30b	2.84a	2.91a	5.72a	6.20b	5.82ab	5.50a	2.89c	3.33d	2.61b	2.09a	4.94a	5.30ab	5.48b	5.17ab
Cyanidin-3-cmglc	0.60a	0.73b	0.70b	0.68b	1.39b	1.20a	1.41b	1.19a	1.34c	0.70b	0.55a	0.57a	0.71a	0.92b	0.69a	0.89b
Petunidin-3-cmglc	2.86b	2.93b	2.49a	2.39a	5.34ab	5.75b	5.48b	5.05a	3.03c	3.40d	2.56b	2.17a	5.23b	4.60a	5.03b	4.62a
Peonidin-3-cmglc	1.46b	1.84d	1.15a	1.58c	2.10b	2.36c	1.51a	2.15b	1.56b	2.20c	1.48b	0.99a	1.78b	1.64a	1.94c	1.61a
Malvidin-3-cmglc	9.72b	9.34b	8.42a	8.12a	17.85ab	20.01c	19.12bc	16.91a	10.60c	11.70d	9.18b	7.67a	18.77	17.34	18.00	17.56
Total	222.6b	205.1a	192.9a	189.6a	331.0ab	358.3c	340.7bc	310.0a	204.0c	226.4d	184.8b	148.6a	315.7	296.4	312.1	293.9
Compounds	Wine 5				Wine 6				Wine 7				Wine 8			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Hydroxybenzoicacids																
Gallicacid	83.04	80.71	78.88	79.68	90.40	93.00	93.20	90.60	70.37	70.06	65.46	66.61	56.50	55.60	54.20	53.30
Syringic acid	3.52c	3.16b	2.78a	3.17b	4.21a	5.11b	4.53a	7.84c	3.44b	3.15ab	3.07a	3.05a	3.36a	4.21b	3.21a	3.48a
Ellagic acid	2.53d	1.58b	2.15c	1.26a	1.60a	1.55a	2.43b	1.68a	1.46b	1.31a	1.67c	1.62c	1.26	1.37	1.27	1.26
Total	89.08	85.46	83.81	84.10	96.21	99.66	100.2	100.1	75.28	74.52	70.19	71.27	61.12	61.18	58.68	58.04
Hydroxycinnamic acids																
cis-Caftaric acid	6.16	6.32	6.39	6.37	2.97	3.13	3.07	2.99	7.17	7.18	7.39	7.20	4.06a	4.42ab	4.50b	4.41ab
trans-Caftaric acid	38.96	38.79	39.29	39.03	43.00	43.70	43.00	43.30	22.19	22.23	21.99	21.99	45.50	45.20	45.00	45.10
cis-Coutaric acid	3.17b	2.86a	2.87a	2.79a	6.07b	5.42a	5.28a	6.25b	2.97	2.95	2.82	2.74	6.46ab	7.62c	6.95b	6.17a
trans-Coutaric acid	27.61	27.66	27.70	26.26	27.50a	34.80c	34.30bc	31.70b	21.77	22.13	21.24	21.83	36.30	36.50	36.70	35.70
Caffeic acid	15.96a	15.86a	15.95a	17.40b	4.41a	5.99b	5.90b	4.56a	10.90	10.39	10.76	10.01	4.98	4.74	4.95	4.72
trans-Fertaric acid	9.87	10.08	10.44	9.95	1.94b	1.48a	1.97b	1.88b	9.74	9.27	9.19	9.51	1.31a	1.58b	2.14c	2.17c
p-Coumaric acid	1.01b	1.26c	1.35c	0.90a	6.85	6.48	6.59	6.73	0.27a	0.38b	0.47c	0.50c	2.87	2.76	2.82	2.62
Total	102.7	102.8	104.0	102.7	92.74	101.0	100.1	97.41	75.01	74.52	73.85	73.78	101.5	102.8	103.1	100.9
Flavonols																
Myricetin-3-gal	1.17b	1.50c	1.04a	1.47c	1.26a	1.95c	1.71b	1.29a	2.20b	1.88a	2.14b	2.14b	2.54a	2.31a	2.79b	2.34a
Myricetin-3-glc	8.22	8.12	8.02	7.74	4.03a	5.03b	4.73b	3.85a	12.79	12.78	12.50	12.74	16.40	15.90	16.60	16.20

Table 8 (continued)

Compounds	Wine 5				Wine 6				Wine 7				Wine 8			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Quercetin-3-gal	3.10b	2.93ab	2.77a	2.70a	4.50	4.61	4.62	4.38	2.52b	2.31ab	2.36ab	2.14a	3.06a	3.38b	3.50b	3.40b
Quercetin-3-glc	5.14	5.25	4.92	4.96	3.12	3.19	2.93	2.94	2.99	3.05	2.92	2.91	6.01	6.35	6.02	5.98
Quercetin-3-glcU	3.84	3.75	4.08	4.00	2.34	2.28	2.43	2.37	2.23ab	2.18a	2.41b	2.35ab	4.49a	4.55ab	4.98b	4.82ab
Isorhamnetin-3-glc	3.65	3.94	3.61	3.72	1.58	1.65	1.62	1.61	1.52	1.44	1.54	1.52	1.85	1.87	1.84	1.84
Myricetin	10.31	10.53	10.36	10.32	10.50	10.70	10.10	10.50	9.54	9.27	9.70	9.93	14.10	14.20	14.10	13.70
Quercetin	7.32	7.51	7.11	7.10	9.97a	11.40b	10.30a	10.40a	0.79b	0.66a	0.98c	0.92c	11.60	11.60	11.60	11.20
Kaempferol	0.83ab	0.83ab	0.75a	0.86b	1.20ab	1.30b	1.21ab	1.10a	0.11b	0.06a	0.07a	0.06a	2.33	2.36	2.36	2.27
Isorhamnetin	1.60	1.63	1.59	1.65	0.69	0.76	0.71	0.70	0.20c	0.12a	0.15b	0.12a	0.76	0.75	0.76	0.72
Total	45.17	46.00	44.23	44.53	39.19	42.87	40.36	39.14	34.90	33.75	34.76	34.82	63.14	63.27	64.55	62.47
Flavan-3-ol																
Catechin	42.27	40.07	42.63	41.36	34.62	35.17	29.33	30.48	27.23	27.75	26.23	28.02	29.82	28.42	34.02	32.56
Stilbenes																
trans-Piceid	4.19	4.23	4.21	4.24	0.96b	0.81a	0.81a	0.84a	2.25	2.19	2.08	2.21	0.67a	0.76b	0.71ab	0.70ab
trans-Resveratrol	2.21b	1.85a	2.03ab	1.88a	1.04b	1.23c	0.45a	0.42a	0.85b	0.67a	0.66a	0.70a	0.77a	1.51c	1.43bc	1.33b
Total	6.40	6.08	6.24	6.12	2.00b	2.04b	1.26a	1.26a	3.10b	2.86ab	2.73a	2.91ab	1.44a	2.27c	2.14bc	2.03b
Ellagitannins	8.58c	4.02a	6.22b	8.37c	7.75a	9.67b	8.52a	8.56a	9.43c	7.82b	6.98b	5.51a	6.71a	6.74a	9.33b	6.46a
Anthocyanins																
Delphinidin-3-glc	20.32b	19.13ab	19.22ab	17.97a	17.70b	22.40c	21.40c	15.70a	49.89	50.46	48.18	50.53	40.00ab	42.90bc	44.30c	39.10a
Cyanidin-3-glc	1.13b	1.08ab	1.09ab	1.04a	1.31a	2.31c	1.73b	1.27a	3.91b	3.91b	4.00b	3.58a	3.74c	2.41a	2.67b	2.26a
Petunidin-3-glc	22.17	20.87	20.84	20.64	14.20b	19.10d	17.20c	12.60a	47.23	48.41	45.71	48.22	31.90ab	34.00bc	35.40c	30.80a
Peonidin-3-glc	18.84b	17.98ab	17.84ab	16.81a	4.54a	6.39c	5.87b	4.24a	10.09a	10.41a	10.41a	11.46b	7.96ab	8.39bc	8.62c	7.46a
Malvidin-3-glc	103.9b	97.68ab	97.71ab	93.37a	39.77a	53.40c	48.80b	36.40a	148.9	152.5	143.6	151.3	89.50ab	95.10bc	99.00c	85.60a
Delphinidin-3-acglc	0.89	0.88	0.88	0.85	1.17ab	1.15ab	1.24b	1.14a	1.81a	1.79a	2.10b	1.74a	1.34b	1.51c	1.46c	1.17a
Cyanidin-3-acglc	0.64b	0.64b	0.64b	0.35a	1.37b	1.60c	1.67c	1.06a	1.15b	1.43c	0.80a	1.84d	1.40a	1.90c	2.16d	1.71b
Petunidin-3-acglc	0.82b	0.74a	0.90c	0.95c	1.10b	1.51c	1.92d	0.71a	1.86b	1.70a	1.77ab	2.09c	1.09a	1.82b	2.04c	1.81b
Peonidin-3-acglc	1.14	1.09	1.16	1.07	0.70b	1.22d	0.92c	0.63a	0.46a	0.52b	0.52b	0.74c	1.22c	0.64a	1.68d	0.77b
Malvidin-3-acglc	7.11	6.76	6.89	6.66	2.94a	3.90c	3.54b	3.02a	8.10b	8.33b	7.41a	8.10b	5.49b	5.18b	5.41b	4.49a
Delphinidin-3-cmglc	1.85bc	1.74ab	1.66a	1.96c	1.34b	1.72c	1.35b	1.03a	4.50ab	4.73b	4.34a	4.35a	3.50a	3.52a	4.02b	3.29a
Cyanidin-3-cmglc	0.70c	0.41a	0.39a	0.52b	0.32a	0.81c	0.58b	0.29a	0.61a	0.60a	0.60a	0.91b	0.61a	1.15c	1.17c	0.89b
Petunidin-3-cmglc	2.40ab	2.37a	2.23a	2.55b	1.06b	1.59d	1.28c	0.91a	4.63b	4.70b	4.14a	5.32c	2.73ab	2.93b	3.15c	2.63a
Peonidin-3-cmglc	3.89b	3.70ab	3.61ab	3.47a	0.93b	0.82a	1.08c	0.86a	1.66a	2.18c	1.92b	1.70 a	1.24b	1.50c	1.47c	1.12a
Malvidin-3-cmglc	13.37b	12.35a	12.34a	11.85a	4.45b	5.90c	5.62c	3.60a	18.18ab	18.73b	17.02a	18.02ab	8.47a	9.31b	9.68b	7.95a
Total	199.2b	187.4ab	187.4ab	180.0a	92.90b	123.8d	114.2c	83.46a	303.0	310.4	292.6	309.9	200.2ab	212.3bc	222.2c	191.1a

Table 8 (continued)

Compounds	Wine 9				Wine 10				Wine 11				Wine 12			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Hydroxybenzoic acids																
Gallic acid	78.58	77.56	76.16	75.84	97.30b	101.5b	98.17b	80.84a	91.70b	90.60b	89.20ab	81.00a	110.7	109.1	112.2	107.4
Syringic acid	3.26c	2.85ab	2.73a	3.08bc	2.09a	2.61b	2.53b	3.81c	6.27c	5.48b	5.63b	4.62a	2.57a	3.04b	3.11b	2.35a
Ellagic acid	2.87b	2.65ab	2.61ab	2.47a	1.89a	2.16b	2.15b	1.87a	1.74ab	1.63a	1.87b	1.67a	2.05	2.20	2.18	2.20
Total	84.71	83.06	81.50	81.39	101.3b	106.2b	102.9b	86.52a	99.71b	97.71b	96.70b	87.29a	115.3	114.3	117.5	112.0
Hydroxycinnamic acids																
cis-Caftaric acid	4.38a	4.78b	4.36a	4.33a	2.59a	2.54a	2.75a	4.91b	4.08b	4.02b	3.99b	3.61a	4.69	4.76	4.72	4.83
trans-Caftaric acid	36.09	35.82	35.16	34.71	33.50	33.62	33.58	31.13	16.00b	15.70b	15.60b	14.20a	29.82	29.77	29.97	29.83
cis-Coutaric acid	4.31	4.21	4.08	4.21	3.77a	4.01a	3.70a	4.52b	4.02b	2.90a	2.91a	2.72a	4.14ab	4.47b	3.90a	4.07a
trans-Coutaric acid	40.34	40.90	39.97	39.40	32.04b	32.36b	32.24b	26.08a	16.00b	16.00b	17.00b	14.20a	24.23	23.89	25.16	24.57
Caffeic acid	3.39b	2.62a	3.93c	3.91c	5.88a	5.88a	5.80a	7.41b	15.60b	15.00b	14.40b	12.70a	2.36b	2.66c	2.09a	2.22ab
trans-Fertaric acid	4.77b	5.04b	4.33a	4.82b	4.19	4.26	4.25	4.25	2.93a	3.47b	3.39b	2.99a	2.23b	2.20b	1.91a	1.97a
p-Coumaric acid	0.40c	0.29b	0.24a	0.23a	0.62b	0.47a	0.50ab	3.19c	11.80b	11.40ab	12.00b	10.60a	0.68b	0.75c	0.58a	0.64b
Total	93.68	93.67	92.07	91.61	82.59	83.15	82.83	81.49	70.43b	68.49b	69.29b	61.02a	68.15	68.51	68.33	68.13
Flavonols																
Myricetin-3-gal	0.56a	1.19c	0.98b	1.15c	1.15a	1.56b	1.90c	1.92c	2.43c	2.25bc	2.10b	1.89a	1.38	1.33	1.38	1.29
Myricetin-3-glc	14.12	14.76	14.34	14.22	6.51a	6.99ab	7.65b	10.68c	13.80b	13.50b	13.20b	11.80a	4.12ab	4.12ab	4.24b	3.82a
Quercetin-3-gal	4.20	4.01	4.21	3.99	2.96a	3.08a	3.18a	3.97b	3.11a	3.48b	3.46b	3.39ab	3.38	3.39	3.30	3.23
Quercetin-3-glc	4.31	4.24	4.01	4.05	3.11a	3.29a	3.17a	4.04b	4.04bc	4.15c	3.73ab	3.49a	2.76	2.77	2.66	2.64
Quercetin-3-glcU	3.23	3.04	3.32	3.27	2.33a	2.36a	2.63b	3.26c	3.02	2.97	3.08	2.81	2.06ab	1.98a	2.20b	2.13ab
Isorhamnetin-3-glc	1.56	1.45	1.55	1.54	1.79a	2.03b	2.01b	1.81a	2.02c	1.78b	1.77b	1.60a	1.11	1.09	1.16	1.13
Myricetin	9.58b	8.56a	8.63a	8.65a	10.77a	12.16b	12.63b	10.99a	6.99	6.54	6.99	6.40	8.49	8.29	8.64	8.24
Quercetin	7.60b	6.66a	7.30ab	6.84a	2.63a	2.96ab	3.17b	6.58c	1.60c	1.41b	1.37b	1.13a	5.39	5.16	5.42	5.08
Kaempferol	1.65c	1.41ab	1.30a	1.47b	0.14a	0.11a	0.11a	0.99b	0.13b	0.11a	0.21c	0.12ab	0.70a	0.72a	0.81b	0.76ab
Isorhamnetin	0.34b	0.31ab	0.31a	0.29a	0.25a	0.24a	0.30b	0.55c	0.13b	0.10a	0.20c	0.09a	0.42b	0.40b	0.35a	0.42b
Total	47.17	45.64	45.95	45.47	31.62a	34.78ab	36.74b	44.77c	37.27b	36.29b	36.11b	32.72a	29.81	29.24	30.17	28.73
Flavan-3-ol																
Catechin	24.83c	19.88b	17.37a	23.21c	13.13a	31.14d	17.91c	15.70b	27.22	32.01	29.05	28.44	29.26a	31.62ab	37.60c	33.48b
Stilbenes																
trans-Piceid	1.57b	1.20a	1.94c	1.94c	2.97b	2.89b	2.80b	1.56a	0.64b	0.39a	0.36a	0.36a	2.44ab	2.58b	2.25a	2.25a
trans-Resveratrol	1.09c	0.86b	0.91b	0.67a	1.97b	1.82b	2.41c	1.06a	1.23ab	2.01c	1.34b	1.08a	1.44c	1.05a	1.13ab	1.20b
Total	2.66b	2.07a	2.84b	2.60b	4.94bc	4.71b	5.21c	2.62a	1.87b	2.40c	1.70b	1.44a	3.88b	3.63ab	3.37a	3.45a
Ellagitannins	14.90b	10.98a	11.97a	11.83a	8.15b	5.52a	8.62b	8.96b	11.86c	11.84c	10.57b	8.18a	10.50c	9.28b	11.67d	6.37a
Anthocyanins																

Table 8 (continued)

Compounds	Wine 9				Wine 10				Wine 11				Wine 12			
	K	M	O	P	K	M	O	P	K	M	O	P	K	M	O	P
Delphinidin-3-glc	8.01c	5.33a	6.23b	5.38a	9.84a	15.46b	19.86c	30.97d	35.00b	38.50c	33.30b	28.60a	24.12c	22.09b	25.58c	19.53a
Cyanidin-3-glc	0.32d	0.18a	0.23c	0.20b	0.72a	1.65b	1.91c	2.19d	2.93c	1.69a	2.41b	2.26b	2.72c	2.71c	2.08a	2.34b
Petunidin-3-glc	8.12c	5.50a	6.55b	5.92a	8.61a	13.99b	17.83c	27.83d	32.30c	30.90bc	28.80b	24.20a	19.81b	18.66b	21.66c	16.12a
Peonidin-3-glc	1.16c	0.83ab	0.90b	0.81a	2.01a	3.19b	4.14c	7.45d	7.10c	6.15b	5.80b	4.80a	6.25c	5.59b	6.45c	4.87a
Malvidin-3-glc	22.18c	15.00a	17.25b	15.51a	26.44a	42.57b	54.36c	86.66d	104.0c	99.50bc	93.50b	78.50a	57.74c	52.26b	62.70d	46.23a
Delphinidin-3-acglc	0.23b	0.19a	0.23b	0.25b	1.20a	1.42b	1.55c	1.28a	2.03c	1.54b	1.44ab	1.36a	0.74a	0.95c	0.82b	0.74a
Cyanidin-3-acglc	0.23c	0.15a	0.45d	0.20b	0.85c	0.64b	0.49a	1.07d	1.40a	1.46a	1.67b	1.70b	0.59a	0.68b	0.71b	0.61a
Petunidin-3-acglc	0.77d	0.31a	0.51b	0.68c	0.48a	0.80b	0.91c	1.45d	1.55b	1.36a	1.60b	1.26a	0.66b	0.63b	0.77c	0.55a
Peonidin-3-acglc	0.15b	0.13a	0.14ab	0.24c	0.29a	0.30a	0.57b	0.73c	0.82b	0.90c	1.03d	0.72a	0.20a	0.25bc	0.26c	0.24b
Malvidin-3-acglc	2.18d	1.63c	0.97a	1.16b	2.46a	3.86b	4.22c	5.48d	6.53b	5.46a	6.18b	5.13a	2.78b	2.86b	3.20c	2.43a
delphinidin-3-cmglc	0.63d	0.28a	0.42b	0.47c	0.52a	1.10b	1.60c	2.72d	2.62bc	2.70c	2.45b	1.94a	1.53c	1.28b	1.73d	1.16a
Cyanidin-3-cmglc	0.15a	0.13a	0.21b	0.32c	0.14a	0.16a	0.38b	0.64c	0.62c	0.55b	0.43a	0.52b	0.45d	0.35b	0.38c	0.20a
Petunidin-3-cmglc	0.49b	0.20a	0.51b	0.51b	0.61a	1.21b	1.25b	2.63c	2.50c	2.62c	2.18b	1.82a	1.50c	1.14b	1.44c	0.94a
Peonidin-3-cmglc	0.13a	0.13a	0.18b	0.18b	0.29a	0.43b	0.66c	1.37d	1.75d	1.15c	1.02b	0.90a	0.76c	0.78c	0.46a	0.56b
Malvidin-3-cmglc	1.76c	1.31b	2.19d	1.06a	2.84a	4.61b	6.26c	9.46d	9.79c	9.52c	8.74b	7.08a	5.31b	4.98b	5.28b	4.29a
Total	46.51c	31.31a	36.96b	32.91a	57.30a	91.40b	116.0c	181.9d	210.9c	204.0bc	190.6b	160.8a	125.2c	115.2b	133.5c	100.8a

Different letters in the same row and in the same wine indicate statistically significant differences ($p < 0.05$). Data from different wines were not statistically compared

glc glucoside, *acglc* acetyl-glucoside, *cmglc* coumaroyl-glucoside, *gal* galactoside, *glcU* glucuronide, *K* wines aged in Kentucky barrels, *M* wines aged in Missouri barrels, *O* wines aged in Ohio barrels, *P* wines aged in barrels

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Compliance with ethics requirements All procedures performed in studies involving human participants were in accordance with the ethical standards of the institutional and/or national research Committee and with the 1964 Helsinki declaration and its later amendments or comparable ethical standards.

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