Evolution of the Phenolic Fraction and Aromatic Profile of Red Wines Aged in Oak Barrels

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ABSTRACT: Various changes occur in red wine during aging in oak barrels. Among these, the variation of the polyphenolic fraction and the transfer of aromatic compounds to the wine from oak wood are of great importance. The aim of the present work is to compare the chemical composition of wines aged in different new oak barrels with similar commercial denominations. During 8 months, the total polyphenol index (TPI), color parameters, anthocyanins and pyranoanthocyanins, and wood aromatic compounds were periodically evaluated. The measurement of the TPI and color parameters was similar in all wines, but significant differences were found in total anthocyanin and vitisin content and in certain aromatic compounds belonging to volatile phenols, furanic compounds, and phenolic aldehydes. The results obtained indicate the need for the winemaker to carry out preliminary tests in order to be able to choose the wood that best suits the sensorial profile of the wine.

1. INTRODUCTION

The aging of red wines in oak barrels is a practice traditionally used in different wine-making areas. The term “Crianza” is a traditional mention widely used in Spain to refer to this type of wines. During the aging in wood barrels, physical, chemical, and physicochemical processes occur.^{1,2} These processes include clarification, oxygen diffusion through stave pores, and extraction of compounds such as aromatic compounds and ellagitannins from oak wood.^{3,4} Among the major compounds released into the wine, ellagitannins are very important because they act as regulators of the oxidation phenomenon. In addition, they can accelerate condensation between tannins and anthocyanins.⁵ All the aforementioned processes can be influenced by the structural characteristics and the chemical composition of oak wood. In this way, the aromatic quality, the aromatic complexity, and the structure of the wine among other parameters are affected too.³

The chemical composition of oak wood is of great importance for obtaining quality aged red wines. This composition is determined by the oak species, their geographical origin,⁶ and the different processes applied in the cooperage.⁷ Seasoning and toasting applied in the cooperage and the ellagitannin content of oak wood determine the sensory impact of the aged wines by the contribution of different amounts of aromatic compounds. In the same way, the ellagitannin content will have an impact on the chromatic properties of wines.

Many studies have been carried out to compare the influence of wood types on the quality of aged red wine.

Such is the case of the volatile profile of wines aged in oak, chestnut, ash, cherry, and acacia wood barrels.³ Other authors studied the differences between aging techniques such as the addition of oak chips in stainless-steel tanks compared to used barrels.⁸ In the same way, research studies have evaluated the similarities or differences in the phenolic composition and the sensory characteristics between wines aged in new barrels (French and American) and wines aged with oak chips.⁹ The origin of the barrel used is a very important factor; in this sense, Spanish, French, and American oak wood have been compared.⁷ In addition, other authors investigated some quality characteristics of wines aged in French oak barrels from four different French forests.¹⁰

Normally, the winemaker chooses the style of barrel that he considers the most convenient for the aging of his red wine. However, he can only be guided by the commercial denominations and the treatments applied in the cooperage. Even though several investigations have been published about the transfer of compounds from the wood to the wine,^{11–15} it is interesting that the winemaker obtains information about the concrete number of compounds transferred from the barrels to

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Table 1. Characteristics of the Barrels Used in the Aging of Red Wines

barrel	oak specie	cooperage	abbreviation	origin	toast	age	volume (L)
Surtep Top Premium	<i>Quercus petraea</i>	Surtep	STP	French oak	medium	new use	225
Surtep Premium	<i>Quercus petraea</i>	Surtep	SP	French oak	medium	new use	225
Sylvain Réserve	<i>Quercus petraea</i>	Tonnellerie Sylvain	SR	French oak	medium	new use	225
Boutes Coeur	<i>Quercus petraea</i>	Boutes	BC	French oak	medium plus	new use	225
Alain Fouquet St. Emilion Style	<i>Quercus petraea</i>	Alain Fouquet	AF	French oak	medium low	new use	225
Treuil Tonnellerie de Brive	<i>Quercus petraea</i>	Tonnellerie de Brive	TB	French oak	medium low	new use	225

Figure 1. Evolution of the color intensity (a), tonality (b), and TPI (b) through the aging in oak barrels measured by spectrophotometry. Points in the same day with the same letter are not significantly different ($p < 0.05$). Mean $\pm$ standard deviation of three replicates.

the wine. The main objective of this experiment is to perform a comparative study about the evolution of the color parameters, total polyphenol index (TPI), anthocyanin and pyranoanthocyanin content, and wood volatile compounds of Tempranillo red wine using French oak barrels with a similar commercial denomination and similar toasting treatments.

2. EXPERIMENTAL SECTION OR COMPUTATIONAL METHODS

2.1. Red Wine. The red wine used in the present work was made by Bodegas Comenge (Ribera del Duero, Spain) using *Vitis vinifera* L. cv. Tempranillo 2017 vintage grapes. The red wine was made using the typical process of Ribera del Duero wines. The making process includes destemming and crushing of red grapes. During the fermentation process, the liquid has been macerated on the seeds and skins trying to extract enough phenolic compound concentration to get an optimal wine for aging. These processes have been performed in stainless-steel tanks. After alcoholic fermentation, the pressing procedure was carried out in a vertical press. The final wine (after malolactic fermentation) was analyzed, and the basic enological

parameters were obtained. Ethanol content = 14% vol; pH = 3.7; volatile acidity = 0.35 g/L of acetic acid. The wine was not filtered before the aging in the oak barrel.

2.2. Oak Barrels and the Aging Process. The characteristics of the French barrels used are shown in Table 1. They are high-quality new oak barrels from the following cooperages: Surtep (Bordeaux, France); Tonnellerie Sylvain (Saint-Denis-de-Pile, France); Boutes (Bordeaux, France); Alain Fouquet (AF; Angers, France); and Brive Tonneliers (Brive-la-Gaillarde, France). All wines were aged in Bodegas Comenge per triplicate for 8 months. Sampling was performed one time every 2 months.

2.3. Analysis of the Starting Wine. The analysis of ethanol content, pH, and volatile acidity was determined before the aging process by Fourier transform infrared spectroscopy using the OenoFossequipment (FOSS Iberia, Barcelona, Spain).

2.4. Analysis of Color Parameters and the TPI. The color parameters determined were the color intensity and the tonality following the Glories procedure.¹⁶ The absorbance at 420, 520, and 620 nm with a 1 mm path length glass cell was

Figure 2. Evolution of total anthocyanins (mg/L) (a), vitisins (mg/L) (b), and vinylphenolic pyranoanthocyanins (mg/L) (b) through the aging in oak barrels measured by HPLC-DAD. Points in the same day with the same letter are not significantly different ($p < 0.05$). Mean $\pm$ standard deviation of three replicates.

measured using the UV/visible 8453 spectrophotometer from Agilent Technologies (Palo Alto, CA, USA). The same spectrophotometer was used to obtain the absorbance at 280 nm with a 1 cm path length quartz cell. The TPI was determined after diluting the wine sample 100 times.¹⁷

2.5. Analysis of Anthocyanins and Pyranoanthocyanins. The anthocyanin and pyranoanthocyanin compounds were analyzed by high-resolution liquid chromatography with a diode array detector (HPLC-DAD). The Agilent Technologies 1100 (Palo Alto, CA, USA) chromatograph equipment was used for this purpose. A Kinetex C18 (100 $\times$ 4.6 mm; 2.6 μ m)

(Phenomenex, Torrance, CA, USA) reverse phase column was used for separation.

The mobile phase has been formed by two eluents used in a 0.8 mL/min work flow: solvent A (water/formic acid, 95:5, v/v) and B (methanol/formic acid, 95:5). Detection was performed by scanning in the 500–600 nm range, and calibration was performed using an external standard at 525 nm and expressed as mg/L of malvidin-3-glucoside ($r^2 = 0.9999$).¹⁸

2.6. Analysis of Wood Aromatic Compounds. Volatile phenols, furanic compounds, lactones, phenolic aldehydes, and

phenolic alcohols were analyzed by gas chromatography–mass spectrometry (GC–MS). The equipment used was an Agilent Technologies 6890N-MSD-5973N gas chromatography–mass spectrometer. Chromatographic separation¹⁹ was performed with the DB-WAX column (30 m × 0.25 mm internal diameter × 0.25 μ m film thickness) (J&W Scientific, Folsom, CA, USA). The method was calibrated using the following external standards: guaiacol, eugenol, furfural, furfuryl alcohol, 5-methyl furfural, oak lactone, vanillin, syringaldehyde, acetovanillone, ethyl vanillin, and vanillin alcohol (Merck, Hohenbrunn, Germany).

Liquid extraction with dichloromethane was performed before the chromatographic separation. A 2.5 mL volume of red wine was mixed with 250 μ L of dichloromethane and 25 μ L of 3,4-dimethylphenol solution (10 mg/L) (Merck, Hohenbrunn, Germany) as the internal standard; 0.37 g of NaCl was added and stirred in a vortex for 5 min. After centrifugation at 7500 rpm for 15 min at 4 °C, the dichloromethane phase was extracted and injected into the chromatograph (1 μ L).

2.7. Statistical Analysis. PC Statgraphics v.5 software (Graphics Software Systems, Rockville, MD, USA) was used to calculate means, standard deviations, analysis of variance, the least significant difference test, and the principal component analysis (PCA). The significance was set at $P < 0.05$.

3. RESULTS AND DISCUSSION

3.1. Evolution of the Color Intensity, Tonality, and TPI. In general, a decrease in the color intensity was observed in all the wines during the aging period in oak barrels (Figure 1a). These results were confirmed by other authors too^{20,21} but in contrast with the published results.²² Our results are related to the loss of total anthocyanin content during the aging period (Figure 2a). This is possibly due to the reaction of the pigments with other wine compounds. The difference in the results reported by ref 22 could be due to a major content of stable pigments such as vitisin in these wines before the aging period. Higher concentrations of these pigments can lead to greater color stabilization, which can also be seen in absorbance parameters such as color intensity. Nevertheless, after 240 days, the values of these wines did not show significant differences compared with the other aged red wines. Therefore, no relationship was found between the color intensity parameter and the type of toasting applied in the studied barrels.

The tonality measures the ratio of the yellow (Abs420nm) to the red component (Abs520nm). All wines slightly increased the tonality after 240 days of aging (Figure 1b). This typical increase of tonality after an aging stage is due to an increase of 420 nm absorbance and a decrease of 520 nm absorbance during the aging period. At the first analysis (60 days of aging), significant differences could be appreciated between Boutes Coeur (BC), Treuil Tonnellerie de Brive (TB), Surtep Top Premium (STP), and Surtep Premium (SP) wines. However, when the aging period was finished (240 days), nonsignificant differences were found between the red wines.

Regarding the TPI, there was a progressive increase throughout the aging for all wines (Figure 1b). This is due to the transfer of ellagitannins²³ from oak wood, and these tannins are derived from galloyl units esterified to a sugar core.²⁴ Once the aging period was over, significant differences were found between AF, TB, Sylvain Réserve (SR), and STP

barrels. Therefore, the type of toasting applied in the barrel does not seem to be a significant parameter on the TPI. Possibly, due to the fact that all barrels with “medium toast” denomination experienced a similar process in the cooperage, other authors observed a decrease in TPI in heartwood extracts from *Castanea sativa* Mill., when a medium toast is applied compared to a light toast.²⁵

3.2. Evolution of Anthocyanins and Pyranoanthocyanins. The typical color of red wines is due to the presence of anthocyanins and their derivatives. Figure 2a shows the total anthocyanin content from grapes. This fraction includes the monomeric anthocyanins such as the acylated anthocyanins from free anthocyanins with acetic, *p*-coumaric, or caffeic acid. In general, the total anthocyanins have decreased significantly through the aging period. This decrease has been detected in all studied wines, and it confirms and explains the color tonality increase, as shown in Figure 1, and also the decrease of color intensity. These results are according to those obtained by other authors who observed a decrease of anthocyanins through 15 months of aging in a bottle.²⁴ The reason for this progressive decrease in monomeric pigments is that the anthocyanins extracted from grapes react with other wine compounds such as pyruvic acid, acetaldehyde, vinylphenol, vinylcatechol, vinylguaiacol, or monomeric and dimeric procyanidins.²⁶ In addition, the anthocyanins can react with ellagitannins from the oak wood.^{27–29} It is interesting that the wines aged in BC barrels (medium plus toast) showed total anthocyanin values significantly higher than the rest of the wines in all the aging periods. Regarding the end of aging, the wines AF and SR showed significantly higher values than STP wines.

Pyranoanthocyanins are pigments only present in wines and not in the grape as these compounds are formed by different condensation reactions during the vinification process.³⁰ Malvidin-3-*O*-glucoside pyruvate (vitisin A) and malvidin-3-*O*-glucoside-4-vinyl (vitisin B) are stable pigments formed by condensation between anthocyanins and secondary metabolites released by the yeast (pyruvic acid and acetaldehyde).^{31,32} The results obtained indicate that the vitisin levels remained constant through the aging period with values around 3–4 mg/L (Figure 2b). The BC wines showed the highest content of vitisins during the 240 days of aging, and these wines showed the highest content in anthocyanins too (Figure 2a); therefore, these barrels were able to maintain both pigments.

The vinylphenolic pyranoanthocyanins are molecules with great color stability. They are formed by condensation between vinylphenols (formed enzymatically by yeast with hydroxycinnamate decarboxylase activity) and grape anthocyanins.³³ Figure 2b shows the vinylphenolic pyranoanthocyanin content through aging. All studied wines showed a progressive decrease of these compounds. The wines aged in AF and TB barrels (medium low toast) obtained lower concentrations of vinylphenolic pyranoanthocyanins during the first 180 days. However, at the end of aging, no significant differences were found between wines. It is possible that a relationship occurs between the light toasting and this loss of vinylphenolic pyranoanthocyanin compounds.

3.3. Evolution of Volatile Phenols and Furanic Compounds. The volatile phenols analyzed have been the guaiacol (*o*-methoxyphenol) and eugenol [2-methoxy-4-(2-propenyl)phenol]. Both compounds are formed to high temperature by lignin degradation during the toasting of the barrels.³⁴ The sensory descriptor of guaiacol is the smoky,⁷ and

Figure 3. Evolution of volatile phenols ($\mu\text{g/L}$) (a) and furanic compounds ($\mu\text{g/L}$) (b) through the aging in oak barrels measured by GC–MS. Points in the same day with the same letter are not significantly different ($p < 0.05$). Mean $\pm$ standard deviation of three replicates.

eugenol is described by its spices, cloves, and smoke aroma.³⁴ At the first 120 days of aging, significant higher values of volatile phenols were detected in wines aged in STP, BC, and SP than in the rest of the wines (Figure 3a). These high values (around $80 \mu\text{g/L}$) could be due to the toasting of the barrels, medium and medium plus (see Table 1). At the end of aging, the lowest values were shown in wines aged in AF and TB barrels with medium low toast; therefore, the intensity of toasting seems to be related to the transfer of volatile phenols. These results were similar to those obtained by authors who aged wines in French oak barrels during the same aging time.¹

The furanic derivatives found in the wines were furfural, 5-methylfurfural, and furfuryl alcohol. These compounds have aromas related to almonds, toasted almonds, and moldy hay, respectively.³⁵ The furanic derivatives, with the exception of furfuryl alcohol, are formed during the toast by hemicelluloses degradation.³⁶ 5-Methylfurfural comes from rhamnose, and furfural comes from xylose.³⁷ Furfuryl alcohol is formed by the reduction of furfural.³⁵ The wines aged in BC barrels have shown the lowest content of furanic compounds through aging (Figure 3b), always below $2000 \mu\text{g/L}$. These results could be due to the degradation of furanic compounds with high temperatures during toasting, reducing their contents in the oak wood. After 120 days of aging, a significant increase of furanic compounds was observed, especially in STP wines with the highest content through all the aging. In all the wines analyzed, furfuryl alcohol was the compound found in the

greatest quantity. These results agree with those obtained by other authors for the same aging time,³⁷ but in our assays, we found more concentrations of these compounds, possibly because of the usage of new barrels. In the first use of the barrel, a higher concentration of aroma compounds is released compared to reused barrels.^{38,39}

3.4. Evolution of Lactones and Phenolic Aldehydes.

The evolution of lactones (Figure 4a) include cis- and trans-whiskey-lactone isomers (β -methyl- γ -octalactone) that are formed by dehydration of 2-methyl-3-(3,4-dihydroxy-5-methoxybenzo)-octanoic acid present in oak wood during the toasting of the barrels.⁴¹ Some authors found lactones in untoasted oak wood (however, in lower values).⁴⁰ The cis isomer is a stronger odorant than the trans isomer, and its presence is associated to the “coconut” descriptor.⁴¹ The highest extractions of lactones from the wood could be appreciated after 120 days of aging. The different contents of these compounds were significantly different in all the wines studied and at all times. In addition, the highest content was found in STP wines and the lowest content was found in SP wines. Because both barrels have the same toasting commercial denomination (Table 1), this result is because the lactone content is determined by many factors such as wood species and the geographical origin.⁴² It is interesting to note that in most of the measurements, the threshold of perception of the cis isomer exceeded ($92 \mu\text{g/L}$), whereas the contents in the

Figure 4. Lactones ($\mu\text{g/L}$) (a), phenolic aldehydes, and phenolic alcohols (b) through the aging in oak barrels measured by GC–MS. Points in the same day with the same letter are not significantly different ($p < 0.05$). Mean $\pm$ standard deviation of three replicates.

trans isomer only exceeded after 120 days of aging (460 $\mu\text{g/L}$).⁴³

Figure 4b shows the phenolic aldehydes vanillin and syringaldehyde and their derivatives ethyl vanillin, acetovanillone, and vanillin alcohol. Vanillin (4-hydroxy-3-methoxybenzaldehyde) and syringaldehyde (hydroxy-3,4-dimethoxybenzaldehyde) are formed by lignin degradation during the toasting of oak wood^{34,44} or even byproducts of lignin biosynthesis.⁴⁵ Vanillin is characterized by its vanilla, coffee, and smoked aroma. It shows a low perception threshold (0.2–0.32 mg/L) according to ref 46, while syringaldehyde is not usually considered in the aromatic profile of wine because of its high perception threshold (50 mg/L).⁴⁷ In general, a progressive increase of these compounds was appreciated through aging. The BC barrels released the lowest content of phenolic aldehydes, which means that medium plus toasting denomination did not cause a higher concentration than the other types of toasting. In this respect, several authors have reported that the high toasting temperature degrades vanillin, and this results in low values found in wines aged in heavy

toast oak barrels.⁴⁴ The wines aged in STP and SR showed the highest concentrations in phenolic aldehydes and its derivatives at the end of aging. In all analyses, syringaldehyde was the compound found in the highest concentration with values up to 9000 $\mu\text{g/L}$.

3.5. Wood Aromatic Compounds at the End of the Aging Period. Figure 5 shows the PCA of the same four groups of aromatic compounds that were monitored during the aging in oak barrels.

85.17% of the variability distribution is explained by the first two components. PC1 is positively contributed by all the wood aromatic compounds studied, whereas PC2 is positively contributed by phenolic aldehydes and furanic compound contents and negatively contributed by lactones and volatile phenols.

The PCA allowed us to identify five different clusters. Wines aged in SR and AF barrels form a cluster with medium and medium low toast, respectively. In the positive distribution of the PC1, the wines aged in STP (medium toast) form a cluster with high levels in all wood aromatic compounds studied.⁴⁸

Figure 5. Projection of the first two components of the wood aromatic compounds (PCA) at the end of the aging period.

In the negative values of PC2, three clusters were identified. The TB barrels (medium low toast) were grouped in the positive values of PC2 showing high contents in phenolic aldehydes and furanic compounds but low values in lactones and volatile phenols.

The BC barrels formed a cluster in the negative values of both principal components; in addition, they showed low content of phenolic aldehydes and furanic compounds.

These results clearly indicate that barrels with similar commercial denomination produce wines with different volatile profiles. This could be because the information declared by the cooperage is not enough to know a volatile wood profile in the aged wine. Other parameters such as temperature and the time of toasting in conjunction with the origin of oak wood or the type of drying can give more information about the future volatile profile of the wine.

4. CONCLUSIONS

The aging of the same red wine in French oak barrels with the same capacity and a similar type of toast has resulted in significant differences in some studied enological parameters. The measurements of color intensity, tonality, and TPI were similar in all the wines. Nevertheless, the wines aged in BC barrels were able to maintain higher contents in total anthocyanins and in vitisin stable pigments. Regarding the release of aromatic compounds from oak wood, the barrels with slightly high toasting resulted in higher values of volatile phenols but lower values in furanic compounds and phenolic aldehydes. Other compounds such as lactones seem to be unrelated to the type of toasting applied.

The correct choice of the type of barrel is a very important factor to obtain high-quality red wines. Barrels with the same origin and the same commercial designation were possibly subjected to receiving different toasting temperatures, and this can produce different wine profiles. The wine maker's choice should be based on previous assays because similar commercial denomination can give rise to different wines.

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Notes

The authors declare no competing financial interest.

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