

Accumulation of volatile compounds during ageing of two red wines with different composition

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Abstract

This work studied the influence of the composition of wine on the evolution of oak compounds and on esters in red wines aged in American oak barrels. For this purpose, two of the most utilized grape varieties in wine-making, *Merlot* and *Cabernet Sauvignon*, were used. The wines made with these two varieties showed differences in pH and alcoholic degree. The extraction of furanic aldehydes, oak lactones, phenolic aldehydes and phenolic alcohols was higher in the *Merlot* wine, with higher alcoholic degree, than in the *Cabernet Sauvignon* wine, with lower pH. The difference in the concentration of these compounds in both wines was important up to 10–12 months of ageing, as afterwards most of these compounds were degraded. The evolution of fermenting esters was not affected by the composition of wine.

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Keywords: Wine composition; Aging wine; Oak compounds; Esters; Oak wood

1. Introduction

Wine, during ageing in oak barrels, acquires aromatic complexity as a result of extractions of substances from wood. The quantity of compounds potentially extractable from oak barrels by wine depends mainly on the geographical origin and on the species of oak (Miller, Howell, Michaelis, & Dickmann, 1992), on the seasoning of the staves (Sefton, Francis, Pocock, & Williams, 1993), on the toasting of the barrel (Chatonnet, Boidron, & Pons, 1989) and on the age of the barrel (Chatonnet, 1991; Garde Cerdán, Rodríguez Mozaz, & Ancín Azpilicueta, 2002). Moreover, the composition of wine aged in the barrels also has an influence on the accumulation of aromatic compounds. Few studies exist on this latter aspect and these were made with synthetic wines with alcohol levels unreachable in wine. Maga (1989) investigated the extraction of *cis*- and *trans*-oak lactones, quantified together, from American oak wood (*Quercus alba*) macerated in model wines with different concentrations of ethanol (0%, 10%, 20%, 40% and 60%). In this study the highest concentration of these substances was

reached in the samples of 40% ethanol. Puech (1987) found that the extraction of phenolic aldehydes from Bulgarian oak shavings (*Quercus sessilis*) was higher in a hydro-alcoholic medium at 55% than in a medium with 10% of ethanol. Once extracted, the compounds from oak wood can undergo chemical or biochemical transformations in the wine (Boidron, Chatonnet, & Pons, 1988) and as a consequence, they can modify their concentration (Garde Cerdán, Torrea Goñi, & Ancín Azpilicueta, 2002; Spillman, Iland, & Sefton, 1998).

Other important compounds for the wine aroma are the ethyl esters, mainly synthesized by yeast during fermentation. At present, little is known about their evolution during ageing of wine in barrels. However, due to the fact that these compounds are in equilibrium with their corresponding acid and with ethanol, the alcohol level of wine could affect this equilibrium and as a consequence, their concentrations during ageing. It has also been observed that the evolution of esters during ageing can be influenced by the pH of wine (Garofolo & Piracci, 1994; Ramey & Ough, 1980).

The effect of factors related to oak barrels on the volatile composition of wine has been extensively studied. However, there are few studies about the influence of the composition of wine on the accumulation of oak compounds. The aim of this investigation was to study

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the accumulation of volatile compounds from oak wood in two wines with different alcoholic degree and pH, factors that can affect the extraction and subsequent evolution of the aroma compounds. The evolution of esters during ageing in both wines was also studied. In order to do so, wines made with *Cabernet Sauvignon* and *Merlot* varieties were used, as these are varieties broadly used for the elaboration of aged red wines. These wines were aged in American oak barrels of one-year use, during 18 and 17 months, respectively. The barrels remained in the winery under the same conditions of humidity and temperature.

2. Materials and methods

2.1. Samples and vinification

Cabernet Sauvignon and *Merlot* wines from vineyards in the Navarra Region, in the North of Spain were used. The elaboration of the wines took place in the cellar and an identical process was used for both varieties. The grapes were destemmed and crushed before being put into stainless steel tanks. Maceration was carried out during fermentation with periodical pumping over. The alcoholic fermentation was made at 28 °C and brought to dryness. The wines underwent malolactic fermentation before being put into the barrels. The wines were aged in one-year-old barrels made of American oak (*Q. alba*). The wood was naturally seasoned for 36 months and during cooperage the wood was submitted to a medium toasting. All the barrels were fabricated at the Lafitte cooperage (Cognac, France). Samples of the two young wines were taken and in the case of the *Cabernet Sauvignon* variety, samples were taken after 3, 6, 9, 12, 15 and 18 months of ageing and for the *Merlot* variety after 2, 5, 8, 11, 14 and 17 months of ageing. Samples were taken from three barrels of the same lot and they were under the same conditions of humidity and temperature in the cellar. The general parameters for characterizing each wine are shown in Table 1. The main differences between these two wines were the alcoholic degree and pH. The alcoholic degree of the *Merlot* wine (13.6%, v/v) was considerably higher than that of the *Cabernet Sauvignon* wine (12.3%, v/v). The pH of the *Merlot* wine (3.70) was higher than the pH of the *Cabernet Sauvignon* wine (3.45).

2.2. Analysis of volatile compounds of wines by gas chromatography

The compounds studied were: 2-furancarboxaldehyde (furfural), 5-methyl-2-furancarboxaldehyde (5-methylfurfural), 5-hydroxymethyl-2-furancarboxaldehyde (5-hydroxymethylfurfural), 2-furanmethanol (furfuryl alcohol), *cis*- β -methyl- γ -octalactone (*cis*-oak lactone),

Table 1
Enological parameters^a of *Cabernet Sauvignon* and *Merlot* wines

	<i>Cabernet Sauvignon</i>	<i>Merlot</i>	
Alcohol level	12.3	13.6	% (v/v)
Total acidity ^b	7.34	6.04	g/l
pH	3.45	3.7	–
Volatile acidity ^c	0.31	0.33	g/l
Free SO ₂	32.2	30.0	mg/l
Total SO ₂	68	50	mg/l
Fe	4.0	3.5	mg/l
Sugar	2.0	1.9	g/l
Dry extract	29.5	30	g/l

^a Measured according to the methods described by the Office International de la Vigne et du Vin (1990).

^b Expressed as tartaric acid.

^c Expressed as acetic acid.

trans- β -methyl- γ -octalactone (*trans*-oak lactone), 4-hydroxynonanoic acid lactone (γ -nonalactone), 4-hydroxybutyric acid lactone (γ -butyrolactone), 4-hydroxy-3-methoxybenzaldehyde (vanillin), 3,5-dimethoxy-4-hydroxybenzaldehyde (syringaldehyde), 4-hydroxy-3-methoxycinnamaldehyde (coniferaldehyde), 4-allyl-2-methoxyphenol (eugenol), 2-methoxyphenol (guaiacol), 4-methyl-2-methoxyphenol (4-methylguaiacol), 4-ethylphenol, 4-ethyl-2-methoxyphenol (4-ethylguaiacol), ethyl butyrate, ethyl hexanoate, ethyl octanoate and ethyl decanoate. The extraction and analysis of these compounds were made following the method described in Garde Cerdán, Rodríguez Mozaz et al. (2002). To do so, a mixture of n-pentane-dichloromethane (3:2, v/v) was used as extracting solvents; both solvents were from Panreac (Montcada and Reixac, Barcelona, Spain). The identification and quantification of the volatile compounds was carried out with a GC-MS Finnigan (San Jose, CA, USA). A DB-WAX capillary column (30 m \times 0.25 mm i.d. and 0.25 μ m film thickness) with stationary phase of polyethylene glycol bonded and cross-linked (Cromlab, Barcelona, Spain) was used. The volume of sample injected was in all cases 1 μ l. The ionization was produced by electronic impact at 70 eV. The internal standards used were 2-octanol (Aldrich, Gillingham, England) and 3,4-dimethylphenol (Merck, Darmstadt, Germany). Standard solutions were prepared from Aldrich reagents dissolved in synthetic wine (alcohol 13%, v/v), whose pH was adjusted to 3.20 with NaOH 3 M. The synthetic wine was prepared by diluting 3 g of tartaric acid (Sigma Chemical Co., St Louis, MO, USA) and 65 ml of ethanol (Merck) in de-ionized water up to 500 ml. This resulting wine underwent the same process of extraction as the samples.

The results shown in figures are the arithmetic average of 6 analyses, since a representative sample was taken from three barrels of the same lot and from each sample two extractions were made of the volatile compounds to be analyzed. All the results of the figures are shown with their standard deviation (Miller & Miller, 1988).

3. Results and discussion

3.1. Evolution of furanic compounds

Furanic compounds, with the exception of furfuryl alcohol, form during the toasting of the wood through degradation of carbohydrates. Furfural, 5-methylfurfural and 5-hydroxymethylfurfural showed a similar evolution in both wines up to 6–8 months of ageing; afterwards the accumulation of these compounds was much higher in the *Merlot* wine, with higher alcoholic degree and higher pH, than in the *Cabernet Sauvignon* wine (Fig. 1a, c and d). According to Maga (1989) and Puech (1981), high alcoholic degrees and low pHs increase the extraction of oak volatile compounds. In this study it was observed that the alcoholic degree was more important for the extraction of these compounds than the pH. After 10 months the three furanic aldehydes were degraded to a large extent in the *Merlot* wine; after 14 months of ageing the concentrations were similar for both wines except for 5-methylfurfural, which showed a higher concentration in the *Merlot*. The degradation of these compounds is due to their reduction to the corresponding alcohols by means of biological mechanisms (Boidron et al., 1988), although the possibility of a chemical reduction cannot be excluded. The furfuryl alcohol was formed in the two wines although in a greater quantity in the *Merlot* wine where the extraction of furfural was higher (Fig. 1b). None of the three aldehydes did reach their threshold level reported by Boidron et al. (1988), although these compounds, even at low concentrations, can increase the aroma of oak lactones (Reazin, 1981).

3.2. Evolution of lactone compounds

The two isomers of β -methyl- γ -octalactone are thought to be formed during the toasting of the barrels through dehydration of 2-methyl-3-(3,4-dihydroxy-5-methoxybenzo)-octanoic acid present in oak wood; γ -nonalactone is present in oak wood and γ -butyrolactone is formed through dehydration of γ -hydroxybutyric acid during the toasting of the wood and also comes from alcoholic fermentation. The different composition of both wines had an effect on the accumulation of *cis*- and *trans*-oak lactones, which were extracted in higher concentration in the *Merlot* wine than in the *Cabernet Sauvignon* wine (Fig. 2a and b). In both wines the concentration of *cis*-oak lactone was above 92 $\mu\text{g/l}$, which is taken as the threshold level (Chatonnet, Boidron, & Pons, 1990), during the whole ageing period. The concentration of *trans*-oak lactone was in both wines below its threshold level (460 $\mu\text{g/l}$) (Chatonnet et al., 1990). Unlike the two previous compounds, the accumulation of γ -nonalactone was not affected by the different composition of the wines as its concentration was similar for both of them over the whole period of ageing (Fig. 2c). This compound was extracted in the two first months and its concentration remained constant up to the end of the studied period. The concentration of γ -nonalactone was around 30 $\mu\text{g/l}$, considered its threshold level (Singleton, 1995), so that it is likely to make a positive contribution to the quality of the wines since it presents a pleasant fruity aroma. γ -Butyrolactone was produced in a similar concentration during alcoholic fermentation in both wines (Fig. 2d). This compound did not accumulate during ageing;

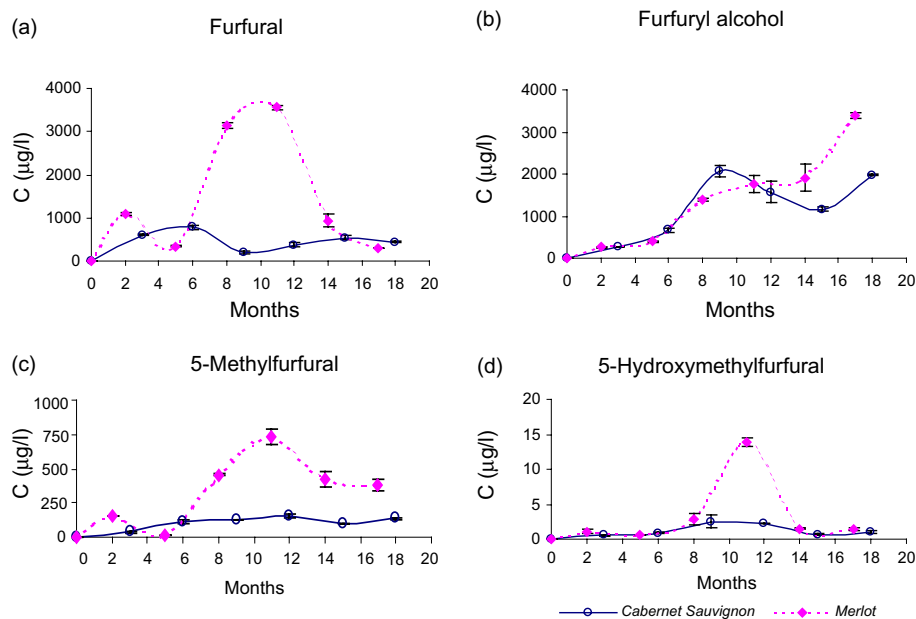


Fig. 1. Evolution of the concentration ($\mu\text{g/l}$) of furfural, furfuryl alcohol, 5-methylfurfural and 5-hydroxymethylfurfural for *Cabernet Sauvignon* and *Merlot* wines during maturation in American oak barrels. Each point is the mean \pm standard deviation of six replicates.

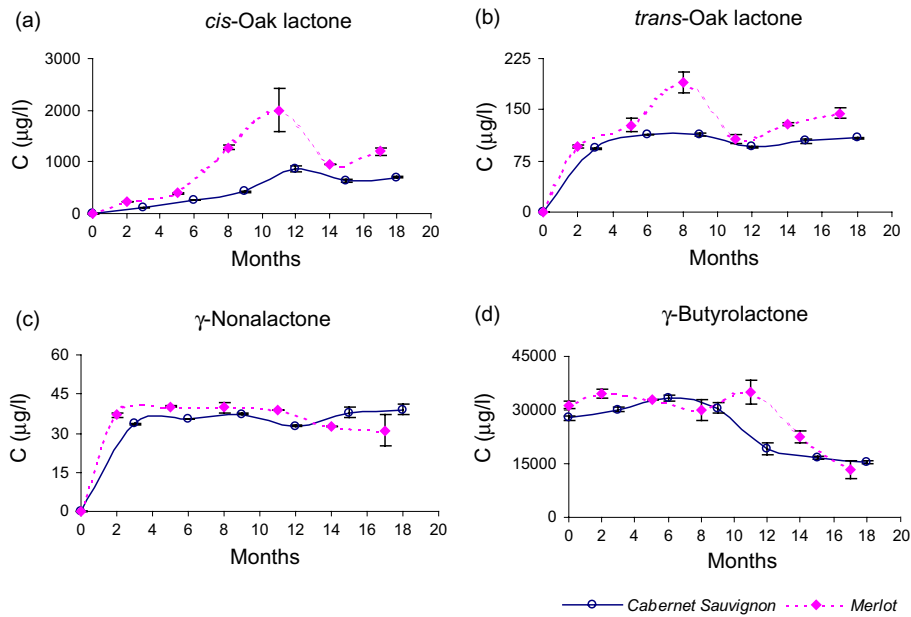


Fig. 2. Evolution of the concentration ($\mu\text{g/l}$) of *cis*-oak lactone, *trans*-oak lactone, γ -nonalactone and γ -butyrolactone for *Cabernet Sauvignon* and *Merlot* wines during ageing in American oak barrels. Each point is the mean \pm standard deviation of six replicates.

therefore it seems that the contribution of oak wood was negligible. After 9–11 months of maturation γ -butyrolactone decreased in a similar way in both wines. This could be due to the opening of the cycle by hydrolysis to form the corresponding acid (Muller, Kepner, & Webb, 1973).

3.3. Evolution of phenolic aldehydes

These compounds arise from thermal degradation of lignin. Fig. 3 shows the evolution of the concentration of vanillin, syringaldehyde and coniferaldehyde. These compounds were extracted in a larger quantity in the

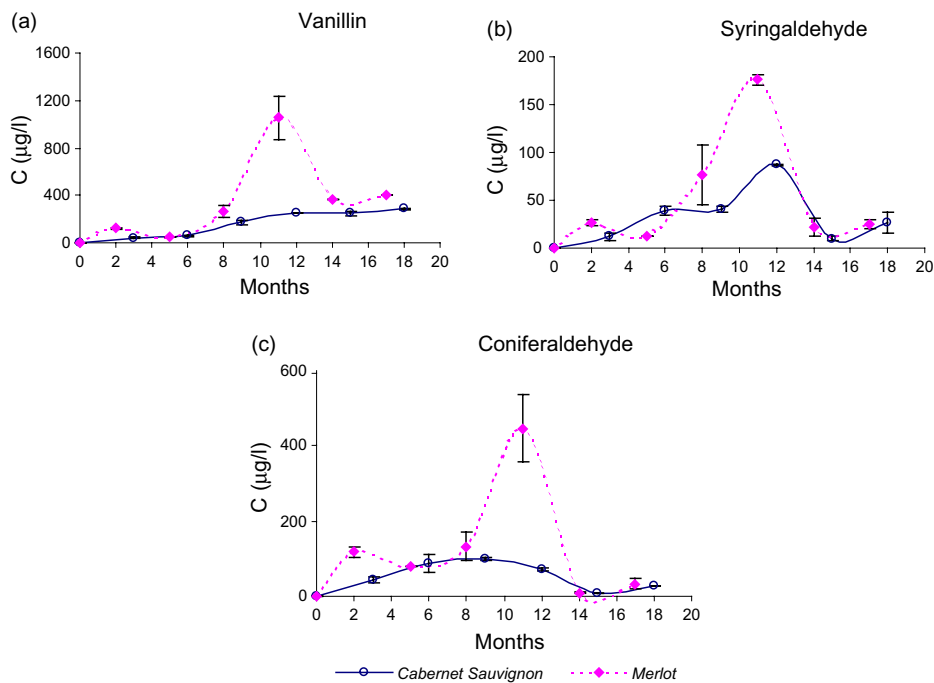


Fig. 3. Evolution of the concentration ($\mu\text{g/l}$) of vanillin, syringaldehyde and coniferaldehyde for *Cabernet Sauvignon* and *Merlot* wines during ageing in American oak barrels. Each point is the mean \pm standard deviation of six replicates.

Merlot wine, with higher alcoholic degree and higher pH, than in the *Cabernet Sauvignon* wine. Puech (1981) found that the compounds coming from lignin are soluble in hydro-alcoholic media, especially if these media are acid. Nishimura, Ohnishi, Masuda, Koga, and Matsuyama (1983) found that ethanol favours the extraction of vanillin, syringaldehyde and coniferaldehyde from the lignin, through a process of ethanolysis, although the study that these authors carried out, was with a synthetic wine of 60% ethanol content. In our study the extraction was more affected by the alcoholic degree than by the pH. In the wine with higher ethanol level (*Merlot* wine) these compounds presented a maximum concentration at 11 months of ageing (Fig. 3a–c). Afterwards their concentration decreased probably due to their microbiological reduction to the corresponding alcohols (Spillman, Pollnitz, Liacopoulos, Pardon, & Sefton, 1998). In the *Cabernet Sauvignon* wine, vanillin

and coniferaldehyde hardly changed their concentrations during ageing.

The most important compound of this group, from the point of view of its contribution to the wine aroma, is vanillin. In the *Merlot* wine, this compound reached higher concentrations than its threshold level, which is 320 $\mu\text{g/l}$ for red wines (Boidron et al., 1988), after 11 months of permanence of wine in the barrels. In the *Cabernet Sauvignon* wine the threshold level was never reached, although after 12 months of ageing the concentrations of this compound were near to this level. The final concentrations of vanillin found in the two wines (285 $\mu\text{g/l}$ for the *Cabernet Sauvignon* wine and 401 $\mu\text{g/l}$ for the *Merlot* wine) were somewhat higher than those found by Spillman, Pollnitz, Liacopoulos, Skouroumounis, and Sefton (1997) for red wine aged in new American oak barrels (200 $\mu\text{g/l}$).

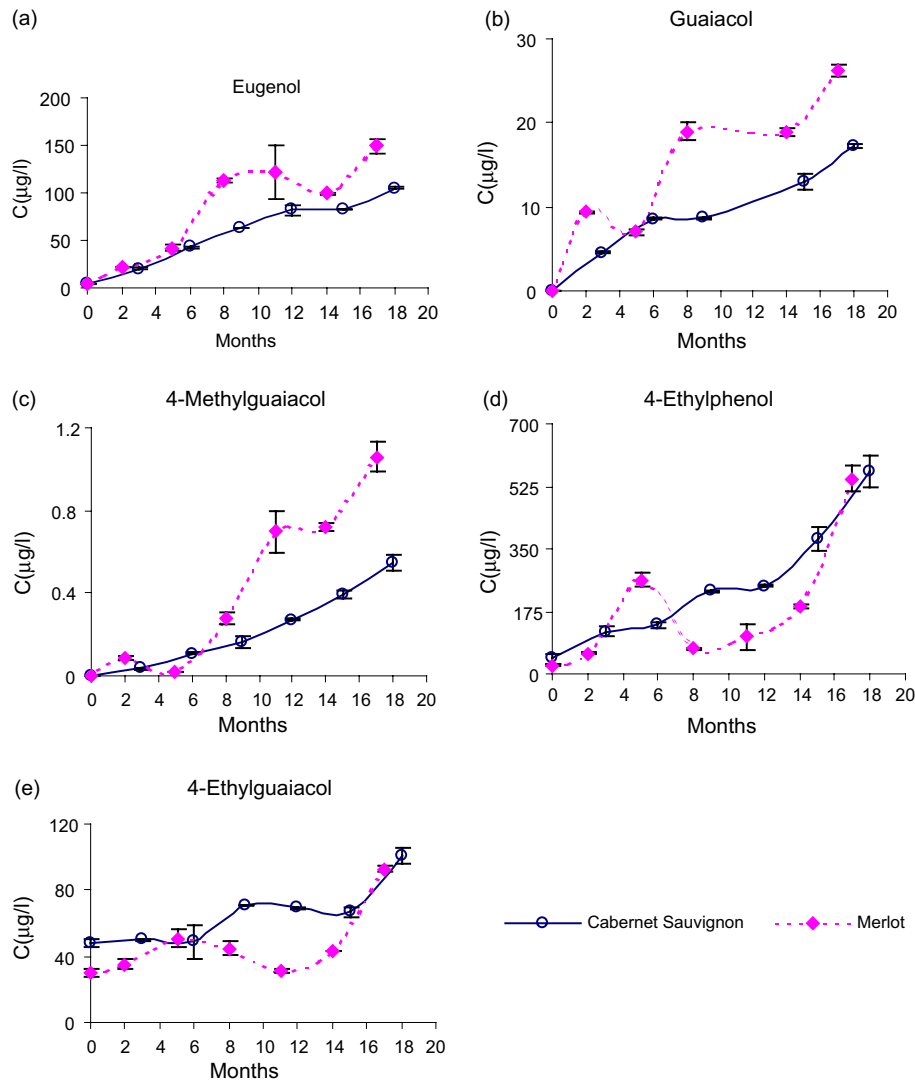


Fig. 4. Evolution of the concentration ($\mu\text{g/l}$) of eugenol, guaiacol, 4-methylguaiacol, 4-ethylphenol and 4-ethylguaiacol for *Cabernet Sauvignon* and *Merlot* wines during ageing in American oak barrels. Each point is the mean \pm standard deviation of six replicates.

3.4. Evolution of phenolic alcohols and ethylphenols

The phenolic alcohols form from the thermal degradation of lignin at high temperature. Ethylphenols form from yeast species (*Brettanomyces* and *Dekkera* genus) during the maturation of wine in barrels. As happened with most of the previous compounds, the extraction of eugenol, guaiacol and 4-methylguaiacol was higher in the *Merlot* wine than in the *Cabernet Sauvignon* wine probably due to the higher ethanol level of the former (Fig. 4a–c). These phenolic alcohols were accumulated over the whole period of ageing in oak barrels. Towey and Waterhouse (1996), and Spillman, Iland et al. (1998) also found that eugenol was extracted in a constant way during ageing of wine. The threshold level of eugenol, which provides the wine with a clove aroma, is 500 µg/l in red wines (Boidron et al., 1988); this level was never reached in our samples.

The concentrations of 4-ethylphenol and 4-ethylguaiacol increased during ageing, although the accumulation was faster in the *Cabernet Sauvignon* wine, with lower alcoholic degree, than in the *Merlot* wine (Fig. 4d and e). Dias, Pereira-da-Silva, Tavares, Malfeito-Ferreira, and Loureiro (2003) found that the greater concentration of ethanol reduced the microbial activity of yeast and made the synthesis of ethylphenols difficult. After 16 months of ageing, the concentrations of these compounds were similar up to the end of ageing. These wines did not reach the limit beyond which the ethyl-

phenols can have a negative influence on the aroma of wines (620 µg/l for 4-ethylphenol and 140 µg/l for 4-ethylguaiacol) (Chatonnet, Dubourdieu, Boidron, & Pons, 1992).

3.5. Evolution of ethyl esters

The initial concentration of ethyl butyrate was higher in the *Cabernet Sauvignon* than in the *Merlot* wine (Fig. 5a). During ageing of the wine, the concentration of this compound showed variations but at the end of ageing it diminished in both wines and they showed similar concentrations. The concentration of ethyl hexanoate did not alter during the first six months and subsequently it increased until it reached a maximum at 12 months (Fig. 5b). After one year the levels of this compound diminished. The concentration of ethyl octanoate (Fig. 5c) initially increased and then remained practically constant until the compound became hydrolyzed in both wines. Contrary to what occurred with ethyl octanoate, ethyl decanoate was formed after one year of ageing (Fig. 5d).

As such, the alcohol level and pH had no effect on the evolution of the ethyl esters during ageing of the wines in oak barrels. In the literature it is reported that pH affects acetate esters more than ethyl esters during ageing (Flanzy, 2000). In our work, when the initial concentration of any ester was different in each wine, this equalled out during ageing. It seems that the esters

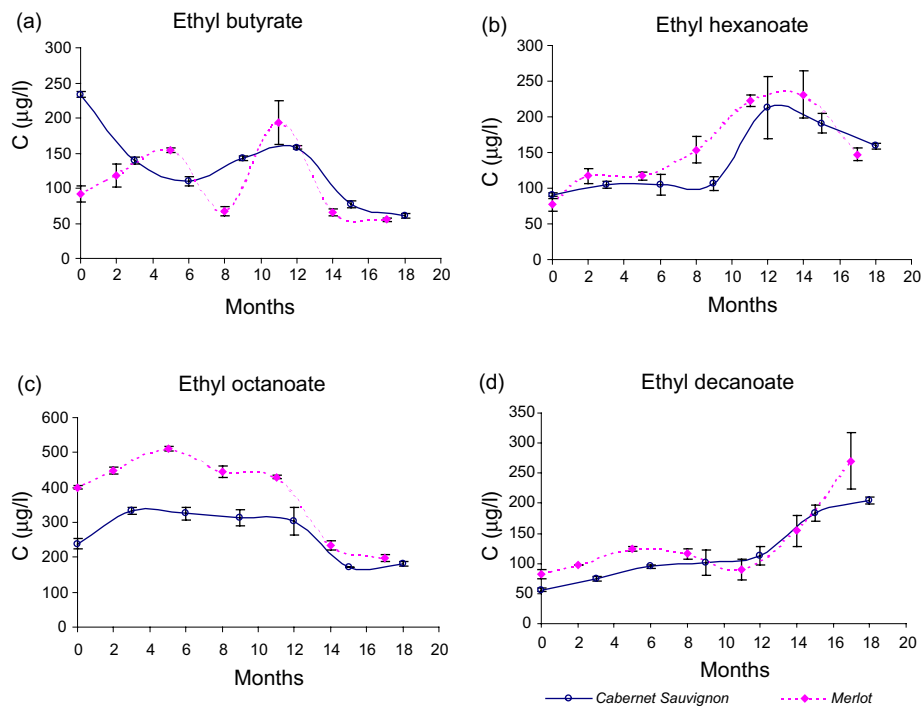


Fig. 5. Evolution of the concentration of ethyl butyrate, ethyl hexanoate, ethyl octanoate and ethyl decanoate (µg/l) for *Cabernet Sauvignon* and *Merlot* wines during maturation in American oak barrels. Each point is the mean \pm standard deviation of six replicates.

tended to reach their concentration equilibrium unaffected by the alcohol level or the pH of the wines. Ramey and Ough (1980) also observed that the hydrolysis rates of the esters in model wines that remain bottled, was hardly affected by the concentration of ethanol. From other studies carried out on bottled wines, miscellaneous results have been obtained. Thus, Simpson (1978), observed that the concentrations of ethyl hexanoate, ethyl octanoate and ethyl decanoate increased over the period that white wine was in the bottle. On the other hand Marais (1978), with South African wine, found that the concentration of these same esters diminished over time.

4. Conclusions

The accumulation of oak compounds was different depending on the wine aged in the barrels. Furanic aldehydes (furfural, 5-methylfurfural and 5-hydroxymethylfurfural), oak lactones, phenolic aldehydes (vanillin, syringaldehyde and coniferaldehyde) and phenolic alcohols (eugenol, guaiacol and 4-methylguaiacol) were accumulated in higher concentration in the *Merlot* wine than in the *Cabernet Sauvignon* wine. Therefore the extraction of these compounds was more favoured in the wine with higher alcoholic degree (*Merlot*) than in the wine with lower pH (*Cabernet Sauvignon*). The difference in the concentration of oak volatiles between both wines was important up to 10–12 months of ageing; afterwards most of them were degraded. The evolution of fermenting esters was unaffected by the composition of wine. In both samples, the concentrations of ethyl butyrate and ethyl octanoate decreased during ageing, while ethyl hexanoate and ethyl decanoate were accumulated. From this study it can be concluded that for the aroma of aged red wines it is not only important to consider the seasoning, toasting, age and origin of oak barrels, but also the composition of the young wine used for ageing.

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