

Extraction and Formation Dynamic of Oak-Related Volatile Compounds from Different Volume Barrels to Wine and Their Behavior during Bottle Storage

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The extraction rate of furfuryl aldehydes, guaiacol, and 4-methylguaiacol, *cis*- and *trans*-oak lactone, and vanillin and the formation rate of furfuryl alcohol and the volatile phenols 4-ethylguaiacol and 4-ethylphenol have been studied in wines matured in different capacity oak barrels (220, 500, and 1000 L). Also, the behavior of these compounds during 1 year of wine bottle storage was followed. The lactones were extracted at a linear rate with large differences that depended on barrel volume. Those compounds related to oak toasting (guaiacol, 4-methylguaiacol, furfuryl aldehydes, and vanillin) seemed to be extracted faster during the first days of oak maturation except for vanillin, which required at least 3 months to accumulate in the wine. The volatile phenols, 4-ethylphenol and 4-ethylguaiacol, were formed in large quantities after the first 90 days of oak maturation, coinciding with the end of spring and beginning of summer. Wines matured in 1000-L oak barrels resulted in the lowest levels of volatile compound accumulation. During bottle storage, some compounds decreased in their concentration (5-methylfurfural, vanillin), others experienced increases in their levels (lactones, furfural, 4-ethylguaiacol, 4-ethylphenol), and the concentration of other compounds hardly changed (guaiacol, furfuryl alcohol).

KEYWORDS: Oak; wine; volatile compounds; maturation; aging

INTRODUCTION

The maturation of wine in oak casks is a common practice that improves wine color, stability, and aroma. The aroma becomes more complex as a result of the extraction of volatile components from the wood matrix. Lactones, volatile phenols, furfuryl compounds, and vanillin are the most important group of aroma compounds of oak-aged wines. Consumers have come to appreciate the aroma and structure of such wines so it is important to know how different types of barrel influence wine quality.

The compounds extracted from the wood have different origins. The *cis*- and *trans*-isomers of 5-butyl-4-methyl-4,5-dihydro-2(3*H*)-furanone, i.e., *cis*- and *trans*- β -methyl- γ -octalactone (and most commonly called oak lactones), and vanillin (4-hydroxy-3-methoxy-benzaldehyde) are already present in natural wood, but their concentration increases with seasoning and toasting (1–4). Guaiacol (2-methoxyphenol), 4-methylguaiacol (2-methoxy-4-methylphenol), furfural (2-furancarboxalde-

hyde), and 5-methylfurfural (5-methyl-2-furancarboxaldehyde) have negligible quantities in green wood and are mainly formed during toasting as a result of lignin degradation (guaiacol and 4-methylguaiacol) or hemicellulose degradation, as is the case of furfuryl aldehydes (3, 5, 6).

The volatile phenols, 4-ethylguaiacol (4-ethyl-2-methoxyphenol) and 4-ethylphenol, have a microbiological origin. Some yeasts capable of contaminating wood (*Brettanomyces* and *Dekkera* genera) decarboxylate cinnamic acids and form these phenols in wines (7), which explains why their presence is commonly attributed to the maturation of wines in used barrels (8–11).

Thus, as Towey and Waterhouse (8, 12) stated, there are three main factors responsible for the pool of potential extractives found in barrels (oak origin, seasoning, and toasting), and the age of the barrel and its volume will affect their content in wines. Another principal factor determining the final quantities of these compounds in wine is the length of oak maturation and bottle storage, because during oak maturation the concentration of some of these compounds in wine will increase with oak contact time whereas others may be consumed by chemical or biochemical transformations, either in the barrel or in the bottle (13).

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The study presented here is focused on the rate of extraction or formation of oak-related volatile compounds in wines stored in barrels of different volume and their evolution in the bottle.

MATERIALS AND METHODS

Barrels. The nine barrels used in this experiment were made of American white oak and were obtained from the same cooperage firm in Spain with the same specifications. The barrels had been used once previously for the aging of Monastrell wine. The following barrels were used in the experiment: 220-L American oak barrels (3 barrels), 500-L American oak barrels (3 barrels), and 1000-L American oak barrels (3 barrels).

The wine used in this experiment was a Monastrell red wine vinified by Bodegas San Isidro in Jumilla, Murcia (13.85% alcohol v/v, pH 3.6). Wine samples were matured for 9 months in the barrels, with periodical topplings, and then stored 1 year in the bottle. Duplicated samples were taken from each barrel.

To determine the volatile compounds two different methods were followed. Furfuryl compounds were determined after solvent extraction and gas chromatography analysis, as described by Cocito et al. (14) and Gómez-Plaza et al. (15). The determination of guaiacol, 4-methylguaiacol, *cis*- and *trans*-oak lactone, vanillin, 4-ethylphenol, and 4-ethylguaiacol in wine was made by gas chromatography–mass spectrometry using deuterated analogues of guaiacol, 4-methylguaiacol, *cis*- and *trans*-oak lactone, vanillin, and 4-ethylphenol as internal standards, following the method described by Pollnitz and Spillman (1, 16, 17). A solution of precisely known concentration of internal standards $^2\text{H}_3$ -guaiacol (5 $\mu\text{g}/\text{mL}$), $^2\text{H}_3$ -4-methylguaiacol (5 $\mu\text{g}/\text{mL}$), $^2\text{H}_4$ -*cis*-oak lactone (19 $\mu\text{g}/\text{mL}$), $^2\text{H}_4$ -*trans*-oak lactone (31 $\mu\text{g}/\text{mL}$), $^2\text{H}_3$ -vanillin (25 $\mu\text{g}/\text{mL}$), and $^2\text{H}_4$ -4-ethylphenol (25 $\mu\text{g}/\text{mL}$) in ethanol was prepared. The deuterated standards were donated by Dr. Pollnitz and Dr. Sefton from the Australian Wine Research Institute.

One hundred microliters of the latter solution was added to the sample (5 mL of wine) in a screw-cap vial using a glass syringe (100 μL , Hamilton). The organic solvent (2 mL of pentane/ether 2:1) was added, and the mixture was shaken briefly. A portion of the organic layer was then placed in a vial ready for instrumental analysis. The conditions for the gas chromatography–mass spectrometry analysis and quantification have been previously described (18).

Statistical Analysis. Significant differences among wines and for each variable were assessed by analysis of variance (ANOVA). These statistical analyses were performed using Statgraphics 2.0 Plus.

RESULTS AND DISCUSSION

***cis*- and *trans*- β -Methyl- γ -octalactones.** Both lactones increased linearly in concentration in wine during the oak maturation period (Figures 1 and 2), the rate of extraction increasing further from day 180 to 270, consistent with the results of Boidron et al. (10). Spillman et al. (13) stated that once a portion of wood is wetted, dissolution of lactones occurs rapidly. After 270 days, the wines matured in 220-L barrels showed the highest concentration of *cis*-lactone, followed by wines from 500- and 1000-L barrels. *trans*-Lactone showed the same dynamic of extraction, but at the end of the oak maturation period, the wines in 1000-L barrels had higher concentration of this compound than wines from 500-L barrels. Several factors could explain the higher levels of *trans*-lactone in wines from 1000-L barrels. For example, the content of lactones in oak wood and the *cis/trans* lactone ratio varies even among trees from the same forest, so barrels from the same geographical origin may well show differences (12). Seasoning is another possible source of variation. Chatonnet et al. (19) have stated that during seasoning, there is a change in the relative abundance of the two isomers in favor of the *cis* form, and even in the same cooperage firm, the position of the wood in the pile (in the inner part or outside) during seasoning could make some differences.

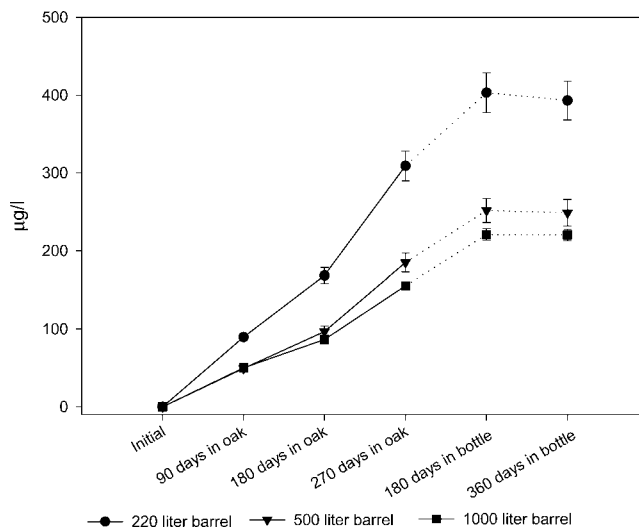


Figure 1. Evolution of the concentration of *cis*-oak lactone during barrel maturation (—) and bottle storage (···).

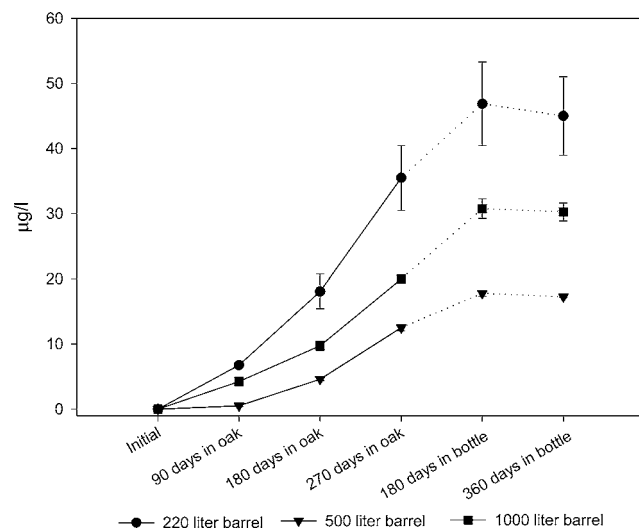


Figure 2. Evolution of the concentration of *trans*-oak lactone during barrel maturation (—) and bottle storage (···).

The levels of lactones found after 270 days of oak maturation were lower than those found previously, after 6 months of oak maturation in new barrels (18). This is coincident with the results of other authors who stated that the pool of these compounds in wood is finite and the progressive use of a barrel will result in a decrease of their extraction levels (8, 20–22).

At the end of the maturation period, the concentration of *cis*-lactone in the wines from the 500- and 1000-L barrels represented 60% and 50% of that found in wines matured in 220-L barrels. We also found that during oak maturation the *cis/trans* ratio decreased in all the wines. Since Pollnitz (17) found that *cis/trans* isomerization hardly occurred in the weakly acidic medium of wine, this may indicate that *cis*-lactone is more easily extracted than *trans*-lactone at the beginning of the oak maturation period and as the maturation progresses the rate of *trans*-lactone extraction increases.

Spillman et al. (13) found that 30–40% of the final concentration of oak lactones was extracted by a hydroalcoholic medium during the first 6 weeks in new barrels. In our study, wines needed almost 180 days in oak to reach these percentages, difference probably being due to the fact that the barrel had been used once previously.

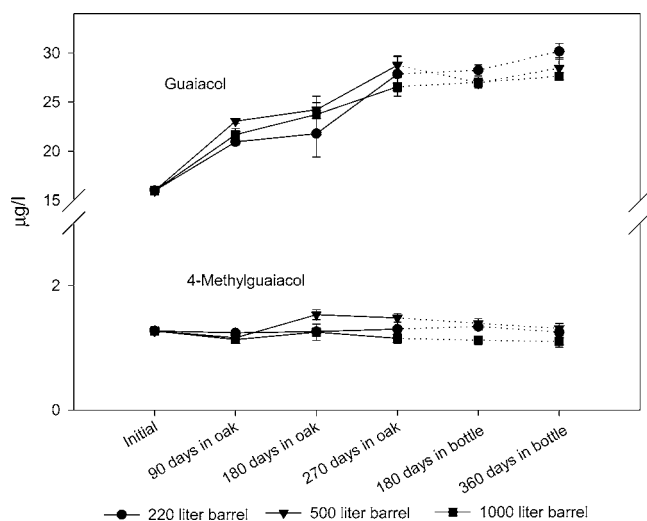


Figure 3. Evolution of the concentration of guaiacol and 4-methylguaiacol during barrel maturation (—) and bottle storage (· · ·).

We also observed an increase in lactone concentration during wine bottle storage, although the *cis/trans* ratio was unaffected. The increase of lactones during bottle storage could be due to the formation of oak lactone from soluble precursors. Otsuka et al. (23) found that a precursor of oak lactone (3-methyl-4-(3',4'-dihydroxy-5'-methoxybenzoyloxy)-octanoic acid) was present in the wood, this bound lactone representing the amount of extra oak lactone released from oak after strong acidic reflux. Also, Masson et al. (24) mentioned the presence of another lactone precursor, the 6'-*O*-gallate derivative of (3*S*,4*S*)-4- β -D-glucopyranosyloxy-3-methyloctanoic acid, but the formation of oak lactone from these precursors during barrel maturation or wine conservation has never been demonstrated. The studies of Pollnitz (17) might also indicate the presence of a bound lactone soluble in hydroalcoholic media since he observed large increases in the lactone concentration when a hydroalcoholic solution that had previously been in contact with oak wood was stored 10 days at 50 °C.

Guaiacol and 4-Methylguaiacol. Very low quantities of these compounds were detected in our study (Figure 3). The maximum extraction of guaiacol occurred during the first 90 days of oak maturation, and the concentration of 4-methylguaiacol was negligible throughout the period studied. Their concentrations were at all times below their thresholds in wine, which Boidron et al. (10) gave as 75 and 65 $\mu\text{g/L}$ for guaiacol and 4-methylguaiacol, respectively, in red wine. It has been stated that those compounds related with toasting decrease very fast after two uses of a barrel, whereas the compounds existing naturally in the wood, such as the lactones, can be extracted over a longer period of time (21). At the end of the oak period the highest values of these compounds were found in wines stored in the 220-L barrels but with no significant differences with 500- and 1000-L barrels. Only small variations were detected during bottle storage. Their low concentration could result in lower smoky and spicy aromas, although some authors (22) have stated that threshold values may not be reliable data in a complex medium such as wine and some compounds may influence wine aroma at lower levels than their individual threshold values by means of additive, synergistic, or suppressive effects.

Vanillin. The evolution of vanillin extraction is shown in Figure 4. Very low quantities of vanillin were extracted during the first 90 days of oak maturation. However, the rate of extraction increased from day 90 to 270 and at the end of the

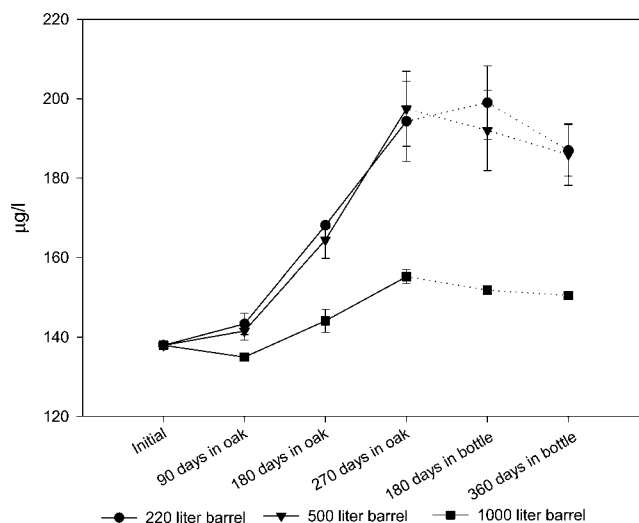


Figure 4. Evolution of the concentration of vanillin during barrel maturation (—) and bottle storage (· · ·).

oak period, as also occurred with the lactones and guaiacol, the wines from 220- and 500-L barrels had significantly higher concentration than wines from 1000-L barrels, the levels of vanillin in wines from 220- and 500-L barrels being very similar. This finding differs from that of Puech (25), who found that the concentration of vanillin observed in wines matured 9 months in 220-L barrels was only reached after 21 months in wines matured in 500-L barrels.

Also, contrary to our results, Spillman et al. (13) found that the rate of vanillin extraction was very fast during the first 20 weeks of oak storage of a hydroalcoholic medium. Vanillin is present in green wood and is also associated to the heating process of the wood, so higher concentration are expected to be found in the first millimeters of the toasted staves. These would be extracted very quickly into the wine, although this was not the case in our study probably because most of the vanillin in the first millimeters had been extracted in the first fill of the barrel. After the first 90 days, wine can impregnate the staves more deeply and extract the vanillin. Also, the profile of extraction could be consistent with a partial generation of vanillin by hydrolytic mechanism, by acid hydrolysis of lignin or lignin-like precursors (13). Since wine is a weakly acidic hydroalcoholic solution, a small portion of the lignin in contact with wine may undergo slow hydrolysis (26). To check if differences in the alcohol content and pH value between wines from different volume barrels could affect the extraction of oak volatile compounds, these two variables were measured in the wines at the end of the oak maturation period. The wines had a mean alcohol content of 14% (v/v) and the pH was 3.45, with no significant differences between wines.

During the first 6 months in the bottle there was little variation in the vanillin concentration, whereas a decrease was observed after the second 6 months, probably because of its transformation into vanillic alcohol and its ethyl ester (1, 17).

Furfuryl Compounds. Furfural was extracted faster in wine matured in the smallest barrel (Figure 5). Maximum values were obtained at 180 days for wines stored in 220- and 500-L barrels, whereas the extraction continued until day 270 for wines in the 1000-L barrel, although the concentration in wines from these barrels was significantly lower than that in wines from 220- and 500-L barrels.

Figure 6 shows the evolution of the content of 5-methylfurfural and furfuryl alcohol during the studied period. Very low

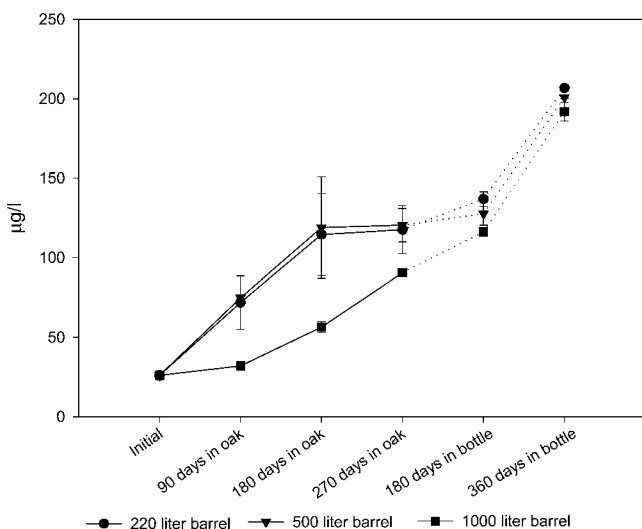


Figure 5. Evolution of the concentration of furfural during barrel maturation (—) and bottle storage (· · ·).

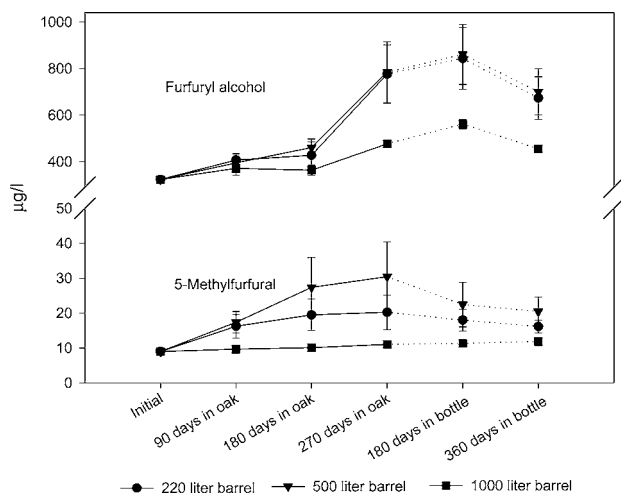


Figure 6. Evolution of the concentration of 5-methyl furfural and furfuryl alcohol during barrel maturation (—) and bottle storage (· · ·).

values of 5-methylfurfural were detected during the oak and bottle period, with no significant differences among the wines.

Furfuryl alcohol (2-furanmethanol) is not extracted from the wood, but it is formed by degradation of the furfuryl aldehydes. Only the furan aldehydes are formed during toasting and the alcohol by enzymatic reaction (27), so factors affecting enzymatic reaction (temperature, pH, and microorganism viability) affect its formation (8). The appearance of furfuryl alcohol, a biological reduction product of furfural, indicates that there was microbial activity in these barrels for at least part of the storage period, although the degradation of furfural to furfuryl alcohol by microbiological enzymatic action may also occur in the absence of bioflora (10). Its concentration in all of the wines was low during the first 180 days of oak maturation but increased rapidly from day 180 to 270; however, its organoleptic importance is not very significant.

Recent studies have shown that another compound, which does have some sensory significance, could be formed from furfural, either by microbiological activity or by chemical pathways. This compound is the 2-furanmethanethiol, described as having a "toasty" aroma (28, 29).

During the last 6 months of bottle storage, furfural increased in concentration. Schreier (30) postulated that furfural in aged wines does not originate exclusively from oak but can also be

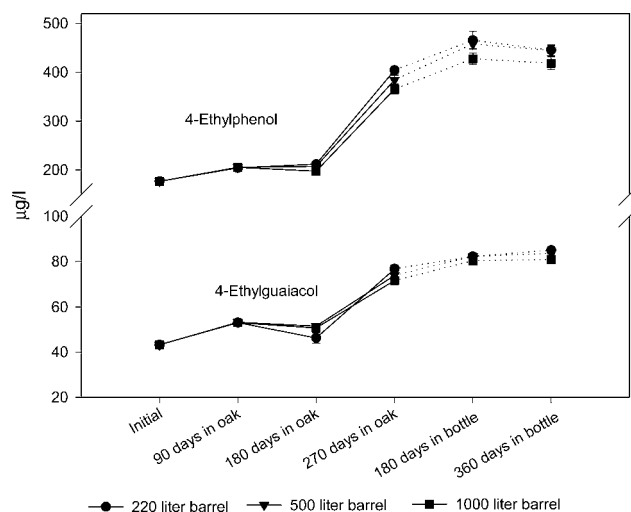


Figure 7. Evolution of the concentration of 4-ethylguaiacol and 4-ethylphenol during barrel maturation (—) and bottle storage (· · ·).

formed from wines during bottle aging from other hexoses and pentoses in the wine. In a study by Perez-Coello et al. (31) related to the evolution of volatile compounds during bottle storage, it was found that furfural increased its concentration from 60 $\mu\text{g/L}$ in young wines to 160 $\mu\text{g/L}$ in wine stored 1 year and to 1230 $\mu\text{g/L}$ after 4 years of bottle storage.

The concentration of 5-methylfurfural decreased during bottle storage as did the concentration of furfuryl alcohol, especially during the last 6 months of bottle storage. Furfuryl alcohol may be subjected to slow chemical degradation in wine (13).

4-Ethylphenol and 4-Ethylguaiacol. Both compounds showed a stable and low concentration until day 180, as reflected in **Figure 7**, increasing from day 180 to 270 (between the months of May to August), with no significant differences among wines at the end of the oak maturation period. The measured concentrations are below their thresholds values of 140 $\mu\text{g/L}$ for 4-ethylguaiacol and 620 $\mu\text{g/L}$ for 4-ethylphenol (20), although Pollnitz (17) stated that the sensory impact of both volatile phenols seems to be additive. As the barrel becomes more impregnated with wine, it can become populated with undesirable microorganisms that can produce significant concentrations of 4-ethylphenol and 4-ethylguaiacol with medicinal and horsey aromas (8). The analysis of used wood reveals an increase of concentration of these compounds (10). Chatonnet et al. (9) found 496 $\mu\text{g/L}$ in new barrels and 1285 $\mu\text{g/L}$ in 5-year-old barrels. Their concentration is sometimes high enough to become an organoleptic defect (32). The maturation of wines in oak barrels demands great care, especially at the end of spring and until autumn because of the increase in temperature and evaporation and the risk of microbial alteration. The phenomenon can also occur in anaerobic conditions, SO_2 being the only tool to prevent this (9). These observations reflect our results; 4-ethylguaiacol experienced little increases during bottle storage and 4-ethylphenol increased its concentration the first 6 months in bottle. When assessors were asked to rank the wines in order by aroma preference there was a negative correlation with 4-ethylphenol (33).

However, it must be remembered that although wine maturation conditions are very important in determining the concentration of 4-ethylphenol and 4-ethylguaiacol, genetic or cultural factors may also be influential. Different grape varieties and different practices of vinification and maturation that affect phenolic fraction could indirectly affect the volatile fraction (34, 35). Monastrell grapes and wines have large quantities of

phenolic acids, and this may also have contributed to the large quantities of volatile phenols found in our wines (36).

The results described here have shown that different rates of extraction have been observed, depending mainly on the origin of the compounds in the wood (toasting or present in natural wood).

Those compounds that are not extracted from wood but whose formation is related to oak maturation and microbiological activity (furfuryl alcohol, 4-ethylguaiacol, and 4-ethylphenol) increased in concentration during the period comprising the end of spring and summer (from day 180 to 270) when high temperature favors microflora growth and enzyme activity.

During bottle storage the concentration of some compounds extracted from the wood increased, as is the case of the lactones and furfural, which is an interesting finding for enologists when planning wine maturation and bottle storage.

The results have also shown that the highest concentration of oak-related volatile compounds was obtained in the smallest barrels, but the differences with wines from 500-L barrels were smaller than expected, especially as regards toasting-related compounds. Two reasons could explain this fact. The first one is that larger barrels usually can be used more times than smaller barrels, imparting oak volatile compounds to wines for longer times (18, 37), but also another reason that could explain the finding is that the toast level of 500-L barrels could be higher than that of 220-L barrels. Barrels of the same toast level made by the same cooper can vary considerably in their actual toast levels because no objective definition of toast level exists (12). Also, the structure of wood such as the proportion of late to early wood and the abundance of fibers may influence heat conduction and reactions to heating (38).

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