AGRICULTURAL AND FOOD CHEMISTRY

Influence of Wine Turbidity on the Accumulation of Volatile Compounds from the Oak Barrel

NEREA JIMÉNEZ MORENO, ANA GONZÁLEZ MARCO, AND CARMEN ANCÍN AZPILICUETA*

Department of Applied Chemistry, Universidad Pública de Navarra, Campus de Arrosadía s/n, E-31006 Pamplona, Spain

In this work the influence of the turbidity of a red wine aged in oak barrels on the accumulation of volatile compounds has been studied. From the results obtained, it may be concluded that the wine turbidity had an effect on the concentration of many of the oak volatile compounds, especially furanic and phenolic compounds. In all cases the concentrations of furanic aldehydes and vanillin were significantly higher (p < 0.01) in the filtered wine than in the unfiltered wine. However, in the unfiltered wine the concentrations of γ -butyrolactone and eugenol were higher. This difference in the concentrations would be due to the different compositions of the wines and the binding of compounds to some components of the lees. Likewise, it has been observed that the composition of the lees is fundamental in the binding of volatile compounds to the lees as the fermentative lees behave differently from the lees from the wine aging in barrels.

KEYWORDS: Wine turbidity; wine aging; volatile compounds; lees

INTRODUCTION

Oak barrels have been traditionally used for the aging of wine and other alcoholic beverages because of the positive effects they have on the quality of these beverages. Among these positive effects can be included color stabilization, spontaneous clarification, which takes place inside the barrel, and greater aromatic complexity attained by the product. The increase in wine aromatic complexity is mainly due to the accumulation of certain volatile compounds from oak wood, which are extracted by wine during the aging process in the barrel.

During wine storage in barrels, the accumulation of oak volatile compounds in wine depends mainly on the quantity of volatile compounds and the concentration of their precursors present in the wood. The species and the geographical origin of the oak wood used to make the barrels have an influence on the concentration of these compounds in the wood (1, 2). In addition, during the making of barrels, some of the processes such as the drying of the oak wood (3-5) and the toasting of the barrels (6, 7) modify the initial content of these volatile compounds. The number of uses of the barrels and the duration of the aging process also have an influence on the quantity of compounds present in the wood (8-10). Besides this, the accumulation of volatile compounds during wine aging depends on the composition of this product, although few studies have been made on the influence of the composition of the wine on the extraction of oak aromatic compounds. Garde-Cerdán et al. (11) found that the extraction of these compounds depended more on the alcoholic degree than on the pH of the wine, as its accumulation was greater in a Merlot wine, with a higher level of alcohol, than in a Cabernet Sauvignon with a lower pH.

Wine aging in barrels may be carried out by previously filtering the product to reduce the number of rackings or by maintaining the initial turbidity of the wine that is going to be aged. Filtration modifies the composition of wine because the product loses particles from the grape, yeast, bacteria, salts, colloids, and amorphous substances (12). On the other hand, several authors using model wine have found that certain nonvolatile compounds such as mannoproteins or yeast walls, normally present in the wine, can bind volatile compounds (13–15). Consequently, it would seem that the application of a filtration treatment before storing the wine in barrels could have an influence on the accumulation of the oak volatile compounds during aging.

For these reasons, the aim of this work was to study the influence of the turbidity level of a red wine on the accumulation of volatile compounds from oak wood. To do so, a Merlot wine was used, and part of this wine was aged in barrels after a diatomaceous earth filtration and the remainder was aged without filtering. The wine remained for 18 months in the barrels, given that, in Spain, this is considered to be the minimum storage time in barrels required to produce high-quality wines, known as "Gran Reserva", according to regulatory specifications on aging.

MATERIALS AND METHODS

Samples and Vinification. The grape used was *Vitis vinifera* var. Merlot (2001 vintage) grown in the vineyards of the Navarra region, in northern Spain. The vinification process was carried out in a cellar.

^{*} Corresponding author [telephone (34) 948-168909; fax (34) 948-169606; e-mail ancin@unavarra.es].

Table 1. Enological Parameters in Wine after Malolactic Fermentation (Young Wine), at the Halfway Stage (8 Months), and at the End of Aging (18 Months) in American and French Oak Barrels

		pН	alcohol (v/v %)	total acidity (g/L) ^a	volatile acidity (g/L) ^b	free SO ₂ (mg/L)
young wine		3.30 ± 0.02	14.8 ± 0.4	5.7 ± 0.1	0.28 ± 0.00	12.0 ± 1.4
filtered wine						
American oak	8 months	3.46 ± 0.01	14.6 ± 0.4	6.1 ± 0.1	0.58 ± 0.01	11.0 ± 1.4
	18 months	3.44 ± 0.01	14.3 ± 0.3	5.5 ± 0.1	0.59 ± 0.05	11.0 ± 1.4
Allier oak	8 months	3.45 ± 0.01	14.6 ± 0.3	6.1 ± 0.1	0.57 ± 0.04	9.0 ± 0.0
	18 months	3.38 ± 0.01	13.9 ± 0.3	6.1 ± 0.1	0.54 ± 0.04	6.5 ± 0.7
Nevers oak	8 months	3.44 ± 0.01	14.6 ± 0.4	6.0 ± 0.1	0.56 ± 0.01	10.0 ± 1.4
	18 months	3.34 ± 0.01	14.1 ± 0.3	6.0 ± 0.1	0.58 ± 0.06	9.5 ± 0.7
unfiltered wine						
American oak	8 months	3.46 ± 0.01	14.6 ± 0.3	6.2 ± 0.1	0.60 ± 0.02	10.5 ± 0.7
	18 months	3.38 ± 0.01	14.5 ± 0.3	6.1 ± 0.1	0.57 ± 0.03	15.0 ± 1.4
Allier oak	8 months	3.46 ± 0.01	14.6 ± 0.4	5.9 ± 0.1	0.41 ± 0.03	13.0 ± 1.4
	18 months	3.40 ± 0.01	14.3 ± 0.3	5.6 ± 0.1	0.52 ± 0.04	12.5 ± 2.1
Nevers oak	8 months	3.46 ± 0.01	14.6 ± 0.4	6.0 ± 0.1	0.54 ± 0.01	12.0 ± 0.0
	18 months	3.31 ± 0.01	14.5 ± 0.3	5.9 ± 0.1	0.51 ± 0.04	12.5 ± 0.7

^a Expressed as tartaric acid. ^b Expressed as acetic acid.

Grapes were destemmed and crushed, and alcoholic fermentation was achieved in stainless steel tanks at 27 °C and taken to dryness; the duration of fermentation was 6 days. Maceration was carried out during fermentation, with periodic pumping over. The wine underwent malolactic fermentation during 54 days before aging in barrels. Half of the wine was filtered in a horizontal tray filter (Della Toffola, Signoressa di Trevignano, Italy) using diatomaceous earth precoats. Fritted diatomaceous earth (Celite 545) was used (World Minerals, Santa Barbara, CA) activated by calcination at 1100-1200 °C. The permeability of the earth was 3.2 darcys, so that a rough filtration of the wine was obtained. Wine turbidity values before and after the filtration treatment were 3420 and 1519 NTU, respectively. The filtered and unfiltered wines were subsequently aged over a period of 18 months in new barrels of American oak (Quercus alba) and French oak (Quercus sessilis) from the Allier and Nevers regions of France. In all cases, wine was put into two barrels of each type of oak. Each barrel had a capacity of 225 L, and all of them were made at the Intona cooperage (Murchante, Navarra, Spain). For the manufacture of the barrels, the wood was naturally seasoned for 36 months and all of the barrels were submitted to a medium toasting. Storage of barrels was carried out in a cellar at controlled temperature and humidity (18 \pm 2 °C and 80-85%, respectively). Samples to be analyzed were taken from the barrels after 1, 2, 3, 4, 6, 8, 10, 12, 14, 16, and 18 months of aging. One representative sample of 750 mL was taken from each of the barrels, and the volatile compounds from each sample were analyzed in duplicate.

The organoleptic characteristics of the wines obtained were evaluated by experts with wide knowledge of the wines produced in Navarra.

Analysis of Volatile Compound of Wine by Gas Chromatography. The compounds identified were furfural, 5-methylfurfural, 5-hydroxymethylfurfural, furfuryl alcohol, vanillin, syringaldehyde, cis-βmethyl- γ -octalactone, *trans-\beta*-methyl- γ -octalactone, γ -nonalactone, γ -butyrolactone, guaiacol, 4-methylguaiacol, 4-propylguaiacol, phenol, *m*-cresol, *p*-cresol, and eugenol. The extraction and analysis of these compounds were made using the method described in Garde-Cerdán et al. (8). A mixture of n-pentane/dichloromethane (3:2 v/v) was used as extracting solvent; both chemicals were from Panreac (Montcada y Reixac, Barcelona, Spain). The identification and quantification of the volatile compounds were done by using a GC-MS Shimadzu QP-5000 (Kyoto, Japan). A DB-Wax capillary column (30 m × 0.25 mm i.d. and 0.25 μ m film thickness) with a stationary phase of bonded and cross-linked polyethylene glycol (Cromlab, Barcelona, Spain) was used. The chromatographic conditions were as follows: He as carrier gas (40 cm/s); injector temperature, 230 °C; temperature of the transfer line, 240 °C. The components were separated after splitless injection of 1 μ L of extract at an initial column temperature of 40 °C for 5 min, with a two-step program of 2 °C/min to 150 °C and then 4 °C/min to 240 °C, and held at that temperature for 10 min. The ionization was produced by electronic impact at 70 eV and selected ion monitoring

(SIM). The internal standards used were 2-octanol (Aldrich, Gillingham, U.K.) and 3,4-dimethylphenol (Merck, Darmstadt, Germany).

The precision of the method was calculated from the coefficient of variation (CV), and the results fluctuated between 4 and 15% for the different quantified compounds. To examine the accuracy of the method, the recovery index was used. The recovery index varied between 70 and 104% depending on the different compounds analyzed.

Enological Parameters and Statistical Analysis. The enological parameters used were those published by the Office International de la Vigne et du Vin (16).

The data were analyzed statistically using the analysis of variance. The software used was SPSS v 12 (SPSS Inc., Chicago, IL).

RESULTS AND DISCUSSION

Enological Parameters. Table 1 shows the enological parameters of the filtered and unfiltered wines during aging in barrels as well as those of the young wine. This table shows that the free SO_2 concentration was somewhat higher in the unfiltered wine than in the filtered one. This could have been due to the fact that lees in the unfiltered wine consume oxygen, creating a more reductive environment than that of the filtered wine aged without lees. The pH and the alcoholic degree were similar in all samples.

Sensory analysis showed that the best olfactory assessment corresponded to the filtered wines, whereas the unfiltered ones presented the best in-mouth sensations.

Furanic Compounds. To determine if the wine filtration had an influence on the accumulation of volatile compounds during wine aging in barrel, an analysis of variance was applied to the data obtained throughout the aging period (Table 2). In this table, it can be observed that the filtered wine showed a higher concentration of furanic aldehydes than the unfiltered wine. Furfuryl alcohol, however, was found in a higher concentration in the unfiltered sample than in the filtered one (Table 2). The biological reduction of furanic aldehydes to their corresponding alcohols was probably lower in the filtered wine than in the unfiltered one because the enzymes, which are responsible for this microbial reduction, are partially excreted to the medium, and so the reduction would be more important in the presence of microbial biomass (17). Diatomaceous earth filtration is not a sterilizing filtration, yet this treatment diminishes the wine microbial population responsible for the reduction of these aldehydes to alcohols. Another factor that could have had an influence on the lower concentration of these aldehydes in the unfiltered samples could be the binding of these compounds to

Table 2. /	Analysis of	Variance	Depending	on Win	e Filtration	Treatment ^a
------------	-------------	----------	-----------	--------	--------------	------------------------

	filtered wine ($n = 132$)				unfiltered wine ($n = 132$)				
	mean	Cl ^b	min	max	mean	CI	min	max	F ^c
furanic compounds									
furfural	368.9	321-417	72	1032	149.8	134-165	40	345	75.2**
5-methylfurfural	143.4	126-160	28	394	68.3	62-74	12	130	69.6**
5-hydroxymethylfurfural	1.6	1.4-1.7	0.7	4.6	1.3	1.1-1.4	0.5	3.6	8.6**
furfuryl alcohol	476.8	398-556	38	1497	645.2	526-764	59	2225	5.4*
phenolic aldehydes									
vanillin	222.6	210-234	73	395	171.8	156-187	34	374	26.2**
syringaldehyde	177.1	164-190	51	352	191.3	170-213	17	500	1.3
lactones									
γ -butyrolactone	12.8	12.4–13.1	8.0	17.8	13.7	13.2-14.2	8.6	20.0	9.2**
γ -nonalactone	51.0	46.6-55.6	24.1	120.9	54.3	50.3-58.2	32.4	113.2	1.1
<i>cis</i> -oak lactone	824.8	736.0-913.7	114.2	1883.6	850.0	748.3-951.7	163.9	2116.7	0.1
trans-oak lactone	403.6	358.1-449.0	85.4	887.8	463.9	421.8-506.0	144.1	901.25	3.7
phenolic compounds									
guaiacol	12.6	11.8-13.5	5.3	22.1	12.7	11.9-13.6	6.7	27.5	0.1
4-methylguaiacol	0.56	0.49-0.62	0.16	2.40	0.38	0.34-0.42	0.11	0.97	22.1**
4-propylguaiacol	0.53	0.48-0.57	0.00	1.14	0.82	0.72-0.92	0.13	2.10	26.2**
phenol	8.8	8.2-9.4	3.1	15.0	7.3	6.9-7.8	2.2	12.6	14.2**
p-cresol	0.095	0.089-0.101	0.00	0.19	0.069	0.065-0.074	0.03	0.13	51.5**
<i>m</i> -cresol	0.092	0.083-0.010	0.00	0.25	0.078	0.073-0.082	0.04	0.15	8.8**
eugenol	48.1	44.4–51.9	9.9	88.1	59.7	55.1-64.3	18.3	105.2	15.0**

^a Means are expressed in $\mu g/L$, except γ -butyrolactone mean, which is expressed in mg/L. ^b Ninety-five percent confidence interval. ^c*, significantly different (p < 0.05); **, significantly different (p < 0.01).

the lees formed in barrel during wine aging. These furanic aldehydes also showed an affinity for the fermentation lees added to a commercial red wine aged in oak barrels, and they remained bound to them; furfural and 5-methylfurfural showed a high affinity for the fermentative lees, whereas the affinity of 5-hydroxymethylfurfural for these lees was inferior (*18*).

The concentrations of furfural and 5-methylfurfural increased to an important extent, especially in the filtered samples during the first month of aging, reaching its highest concentration quickly in all of the wines (Figure 1a,b). Several authors have found that furanic aldehydes are the compounds that most increase during the toasting of the barrel (7, 19). These compounds are found in high concentration in the outer layer of the wood so that the diffusion gradient is very high and its extraction by the wine very fast. Likewise, in the filtered wines, which are more limpid, the extraction of furfural and 5-methylfurfural was faster than in the unfiltered ones. The concentrations of both compounds were higher in the filtered wines practically throughout the whole aging period, although at the end of the process, the samples showed similar concentrations of furfural and 5-methylfurfural. In the filtered wines, 5-hydroxymethylfurfural reached its highest content after 3-4 months of aging in barrel, whereas in the unfiltered wines the highest concentration of this compound was reached after 14 months of aging (Figure 1c). The reduction of 5-hydroxymethylfurfural to its corresponding alcohol has still not been observed in wine, but certain biochemical transformations or chemical reactions with other components of wine could be responsible for the decrease observed in the concentration of this compound (20). Thus, Es-Safi et al. (21) showed that 5-hydroxymethylfurfural played a major role in the flavanol polymerization process. The concentration of furfuryl alcohol progressively increased up to the end of the aging period in all samples (Figure 1d). The highest concentration of this alcohol is produced at the end of aging period, because enzymatic reactions are slow and require a prolonged period of time.

Phenolic Aldehydes. The filtered wine showed a greater concentration of vanillin than the unfiltered one, but no significant difference was found in the concentration of syrin-

galdehyde between both types of wine (**Table 2**). These results suggest that in the filtered wine a lower biological reduction of vanillin to its corresponding alcohol was produced, whereas the lees and/or macromolecules in suspension, present in a higher concentration in the unfiltered wine, probably would have been retaining part of the vanillin extracted from the wood. In an earlier study (18) it was observed that vanillin showed scant affinity for the fermentative lees. However, the fact that the binding of this compound to the lees formed in the barrel took place during wine aging is likely to have been due to several factors. In the first place, it has to be taken into account that the lees formed in the barrel during wine aging are different from the lees from alcoholic fermentation. Thus, Chassagne et al. (22) found that the lees from Chardonnay and Pinot Noir wines showed greater affinity for 4-ethylguaiacol and 4-ethylphenol than the lees obtained after fermentation of a synthetic must. Likewise, during wine aging in barrels, autolysis of yeast leads to the enrichment of the medium in macromolecules, mainly from the yeast walls, and these macromolecules also show capacity for binding volatile compounds (13, 14, 23). In addition, Chassagne et al. (22) showed that autolyzed yeast biomass retained a greater quantity of 4-ethylguaiacol and 4-ethylphenol than non-autolyzed yeast. Several authors have described the influence of pH, temperature, or ionic strength on the interactions between aromatic substances and food components, especially proteins, in model systems (22, 24-26). The concentration of ethanol in the medium as well as the presence of certain compounds, such as esters, also has an influence on the adsorption of aromatic compounds to a substrate (22, 27, 28). Consequently, due to the great physical-chemical complexity of wine, the affinity between the aromatic compounds and the lees will change depending on the wine composition and the lees used.

Figure 2 shows the evolution of the concentrations of vanillin and syringaldehyde during wine aging in Allier, Nevers, and American oak barrels for both filtered wines and unfiltered wines. In the filtered wines the concentration of vanillin increased during the first months of aging and later diminished due to its biological reduction to produce the corresponding















Figure 1. Evolution of the concentration of furanic compounds in filtered and unfiltered wines aged in barrels of different oak woods.





alcohol. In the second half of aging, the vanillin content of these wines remained practically constant. These data would seem to indicate that the maximum reduction of vanillin, in the filtered wine, took place between 6 and 12 months of wine storage in barrel. In the unfiltered wines this compound accumulated more slowly, keeping a practically constant concentration over the first 12 months of aging; from then on, there was an important increase in its content in these wines. This coincides with the results of Fernandez de Simón et al. (29), who found that vanillin is extracted from the wood above all after the first 12 months

of aging. Likewise, the maximum concentration of syringaldehyde was also reached earlier in the filtered wines compared to the unfiltered ones (**Figure 2b**). Only the unfiltered wine aged in Allier oak barrels showed a vanillin concentration higher than its threshold level (320 μ g/L) at the end of the studied period.

Lactones. The filtration treatment had no influence on the content of *cis*- and *trans*-oak lactone of the wines (**Table 2**). Therefore, it would seem that the wine turbidity had no influence on the accumulation of oak lactones in the product and that these compounds did not show any affinity for the lees formed



b. trans -OAK LACTONE



c. y-NONALACTONE

d. y-BUTYROLACTONE



Figure 3. Evolution of the concentration of lactones in filtered and unfiltered wines aged in barrels of different oak woods.

during wine aging in barrel. These compounds also showed scant affinity for the fermentative lees (18). The concentration of these lactones increased progressively in all of the samples during wine aging in barrel, reaching its maximum content in all of the wines after approximately 1 year of aging (Figure 3a,b). Garde-Cerdán and Ancín-Azpilicueta (10) found that the concentration of the two isomers of oak lactone diminished after 9 months of aging both in new barrels and in barrels that had been used once before. However, Jarauta et al. (30) found that the concentration of cis-oak lactone increased during 24 months of a red wine aging in barrels of American and French oak. From the first month of aging all of the wines showed concentrations of cis-oak lactone higher than its perception threshold (74 μ g/L) for red wines. In the case of the *trans* isomer, only the wines aged in French oak barrels showed concentrations higher than its perception threshold in red wines, 320 µg/L.

With regard to γ -nonalactone, wine turbidity did not have an influence on its concentration (**Table 2**). No retention was found of this lactone by the lees from alcoholic fermentation of a red wine (18). Consequently, it would seem that this compound shows no affinity for fermentative lees or for the lees formed in barrel during wine aging. γ -Nonalactone was extracted to an important extent at the beginning of aging in all samples and later diminished both in the filtered wines and in the unfiltered ones (**Figure 3c**). All of the wines exceeded the perception threshold of this compound (30 μ g/L) during the aging process in barrel, although only the unfiltered wines kept a concentration higher than 30 μ g/L after 18 months of aging in barrel. Unlike the rest of the lactones, the filtration treatment had an influence on the content of γ -butyrolactone, which was found in higher concentration in the unfiltered wine than in the filtered one (**Table 2**). This lactone, already present in young wine, is formed by dehydration of the γ -hydroxybutyric acid during the wood toasting, so its extraction becomes possible during the aging of the wine in barrel. The concentration of γ -butyrolactone showed small variations throughout the aging in all of the wines studied (**Figure 3d**). The aperture of the cycle through a hydrolysis reaction to produce the corresponding acid could be responsible for the slight decreases of γ -butyrolactone observed throughout the studied period (*31*).

Phenolic Compounds. The filtration treatment had an influence on the concentration of all the volatile phenols studied in the wine, except in the concentration of guaiacol (**Table 2**). 4-Methylguaiacol, phenol, *p*-cresol, and *m*-cresol were found in higher concentrations in the filtered wines, whereas the concentrations of 4-propylguaiacol and eugenol were higher in the unfiltered wines. These results would seem to indicate that, in the unfiltered wines, 4-methylguaiacol, phenol, *p*-cresol, and *m*-cresol could be retained in the lees and/or macromolecules present in higher concentration in these samples than in the filtered samples. However, 4-propylguaiacol and eugenol did not bind to the lees formed in barrel during wine aging. Just as was observed in other volatile compounds from wood, the lees formed in the barrel showed a behavior different from that of the fermentative lees (*18*).

The concentration of guaiacol increased progressively in all wine samples during aging in barrels, reaching its maximum concentration at the end of the studied period (18 months) in





c. 4-PROPYLGUAIACOL



-8 High

··*····



Months

g. EUGENOL

e. p-CRESOL





J. Agric. Food Chem., Vol. 55, No. 13

b. 4-METHYLGUAIACOL

2,5 1,5 0,5 Months

d. PHENOL



Figure 4. Evolution of the concentration of volatile phenols in filtered and unfiltered wines aged in barrels of different oak woods.

all samples (Figure 4a). 4-Methylguaiacol was extracted earlier in the filtered wines than in the unfiltered ones (Figure 4b).

During a great part of the aging process, the filtered wines showed higher concentration of this compound than the unfiltered ones, although at the end of the studied period the concentrations of 4-methylguaiacol in both the filtered and unfiltered wines were similar. 4-Propylguaiacol was also extracted earlier in the filtered wines than in the unfiltered ones, although later the extraction of this compound was more important in the unfiltered wines, so that during the second half of the studied period, the concentration of 4-propylguaiacol was greater in the unfiltered samples than in the filtered ones (**Figure 4c**). The concentrations of guaiacol, 4-propylguaiacol, and 4-methylguaiacol found both in the filtered and in the unfiltered wines were much lower than their perception thresholds, which are, respectively, 75, 65, and 10 μ g/L in red wine.

In the case of phenol, a progressive increase in its concentration was observed throughout the aging process in barrel, so that the maximum concentration of this compound was reached at the end of the studied period in all cases (**Figure 4d**). *p*-Cresol and *m*-cresol were extracted earlier in the filtered wines, just as occurred with 4-propylguaiacol and 4-methylguaiacol, although all of the wines showed very low concentrations of these compounds (**Figure 4e,f**). None of the wines showed concentrations higher than their perception thresholds.

Eugenol was the most abundant volatile phenol in all of the wines studied. Its concentration increased in all samples up to approximately the end of the first year of aging. Afterward, it remained practically constant up to the end of the studied period (**Figure 4g**). Several authors have observed that the concentration of this compound increases in a constant way during wine aging in barrel (11, 20, 32), whereas Fernández de Simón et al. (29) found that eugenol was mainly extracted during the first year of wine storage in barrel, which coincides with the results of this study. The concentration of eugenol in none of the wines exceeded 500 μ g/L, its perception threshold in red wine.

The results obtained in this work have shown that wine turbidity had an effect on the accumulation of oak volatile compounds during wine aging in barrels. However, due to the complexity of this subject, it would be important to carry out more studies to determine the influence of the different nonvolatile components of wine on the volatile composition of this beverage.

LITERATURE CITED

- Marco, J.; Artajona, J.; Larrechi, M. S.; Rius, F. X. Relationship between geographical origin and chemical composition of wood for oak barrels. *Am. J. Enol. Vitic.* **1994**, *45*, 192–200.
- (2) Ancín, C.; Garde, T.; Torrea, D.; Jiménez, N. Extraction of volatile compounds in model wine from different oak woods: effect of SO₂. *Food Res. Int.* **2004**, *37*, 375–383.
- (3) Sefton, M. A.; Francis, I. L.; Pocock, K. F.; Williams, P. J. The influence of natural seasoning on the concentrations of eugenol, vanillin and *cis-* and *trans-β-*methyl-γ-octalactone extracted from French and American oakwood. *Sci. Aliments* **1993**, *13*, 629– 643.
- (4) Chatonnet, P.; Boidron, J. N; Dubourdieu, D.; Pons, M. Évolution de certains composés volatils du bois de chêne au cours de son séchage. Premiers résultats. J. Int. Sci. Vigne Vin 1994, 28, 359– 380.
- (5) Doussot, F.; De Jéso, B.; Quideau, S.; Pardon, P. Extractives content in cooperage oak wood during natural seasoning and toasting; influence of tree species, geographic location and singletree effects. J. Agric. Food Chem. 2002, 50, 5955–5961.
- (6) Chatonnet, P.; Boidron, J. N.; Pons, M. Incidence du traitment thermique du bois de chêne sur sa composition chimique, 2^e partie: évolution de certains composés en fonction de l'intensité de brulage. *Connaiss. Vigne Vin* **1989**, *23*, 223–250.

- (7) Cadahía, E.; Fernández de Simón, B.; Jalocha, J. Volatile compounds in Spanish, French, and American oak woods after natural seasoning and toasting. *J. Agric. Food Chem.* 2003, *51*, 5923–5932.
- (8) Garde-Cerdán, T.; Rodríguez-Mozaz, S.; Ancín-Azpilicueta, C. Volatile composition of aged wine in used barrels of French and of American oak. *Food Res. Int.* 2002, *35*, 603–610.
- (9) Gómez-Plaza, E.; Pérez-Prieto, L. J.; Fernández-Fernández, J. I.; López-Roca, J. M. The effect of successive uses of oak barrels on the extraction of oak-related volatile compounds from wine. *Int. J. Food Sci. Technol.* **2004**, *39*, 1069–1078.
- (10) Garde-Cerdán, T.; Ancín-Azpilicueta, C. Effect of oak barrel type on the volatile composition of wine: storage time optimization. *LWT–Food Sci. Technol.* **2006**, *39*, 199–205.
- (11) Garde-Cerdán, T.; Torrea-Goñi, D.; Ancín-Azpilicueta, C. Accumulation of volatile compounds during ageing of two red wines with different composition. J. Food Eng. 2004, 65, 349–356.
- (12) Ribéreau-Gayon, P.; Glories, Y.; Maujean, A.; Dubourdieu, D. Clarification and stabilization treatments: fining wines. In *Handbook of Enology: The Chemistry of Wine, Stabilization and Treatments*, 1st ed.; Wiley: Chichester, U.K., 2000; Vol. 1, pp 271–299.
- (13) Voilley, A.; Beghin, C.; Charpentier, C.; Peyron, D. Interactions between aroma substances and macromolecules in a model wine. *Lebensm. Wiss. Technol.* **1991**, *24*, 469–472.
- (14) Lubbers, S.; Voilley, A.; Feuillat, M.; Charpentier, C. Influence of mannoproteins from yeast on the aroma intensity of a model wine. *Lebensm. Wiss. Technol.* **1994**, *27*, 108–114.
- (15) Lubbers, S.; Charpentier, C.; Feuillat, M.; Voilley, A. Influence of yeast walls on the behavior of aroma compounds in a model wine. *Am. J. Enol. Vitic.* **1994**, *45*, 29–33.
- (16) Office International de la Vigne et du Vin. Recueil des Méthodes Internationales d'analyse des Vins et des Moûts, édition officielle; OIV: Paris, France, 1990.
- (17) Chatonnet, P.; Dubourdieu, D.; Boidron, J. N. Incidence des conditions de fermentation et d'élevage des vins blancs secs en barriques sur leur composition en substances cédées par le bois de chêne. *Sci. Aliments* **1992**, *12*, 665–685.
- (18) Jiménez-Moreno, N.; Ancín-Azpilicueta, C. Binding of oak volatile compounds by wine lees during simulation of wine ageing. *LWT–Food Sci. Technol.* **2007**, *40*, 619–624.
- (19) Caldeira, I.; Clímaco, M. C.; Bruno de Sousa, R.; Belchior, A. P. Volatile composition of oak and chestnut woods used in brandy ageing: modification induced by heat treatment. *J. Food Eng.* 2006, *76*, 202–211.
- (20) Spillman, P. J.; Pollnitz, A. P.; Liacopoulos, D.; Pardon, K. H.; Sefton, M. A. Formation and degradation of furfuryl alcohol, 5-methylfurfuryl alcohol, vanillyl alcohol and their ethyl esters in barrel-aged wines. *J. Agric. Food Chem.* **1998**, *46*, 657–663.
- (21) Es-Safi, N. E.; Cheynier, V.; Moutounet, M. Study of reactions between (+)-catechin and furfural derivatives in the presence or absence of anthocyanins and their implication in food color change. J. Agric. Food Chem. 2000, 48, 5946–5954.
- (22) Chassagne, D.; Guilloux-Benatier, M.; Alexandre, H.; Voilley, A. Sorption of wine volatile phenols by yeast lees. *Food Chem.* 2005, *91*, 39–44.
- (23) Dufour, C.; Bayonove, C. L. Influence of wine structurally different polysaccharides on the volatility of aroma substances in a model system. J. Agric. Food Chem. 1999, 47, 671–677.
- (24) Damodaran, S.; Kinsella, J. E. Flavor protein interactions. Binding of carbonyls to bovine serum albumin: thermodynamic and conformational effects. *J. Agric. Food Chem.* **1980**, *28*, 567– 571.
- (25) O'Neil, T. E.; Kinsella, J. E. Binding of alkanone flavors to β-lactoglubolin: effects of conformational and chemical modification. J. Agric. Food Chem. 1987, 35, 770–774.
- (26) Druaux, C.; Lubbers, S.; Charpentier, C.; Voilley, A. Effects of physico-chemical parameters of a model wine on the binding of γ-decalactone on bovine serum albumin. *Food Chem.* **1995**, *53*, 203–207.

- (28) Ramirez-Ramirez, G.; Chassagne, D.; Feuillat, M.; Voilley, A.; Charpentier, C. Effect of wine constituents on aroma compound sorption by oak wood in a model system. *Am. J. Enol. Vitic.* 2004, *55*, 22–26.
- (29) Fernández de Simón, B.; Cadahía, E.; Jalocha, J. Volatile compounds in a Spanish red wine aged in barrels made of Spanish, French, and American oak wood. J. Agric. Food Chem. 2003, 51, 7671–7678.
- (30) Jarauta, I.; Cacho, J.; Ferreira, V. Concurrent phenomena contributing to the formation of the aroma of wine during aging

in oak wood: an analytical study. J. Agric. Food Chem. 2005, 53, 4166-4177.

- (31) Muller, C. J.; Kepner, R. E.; Webb, A. D. Lactones in wine: a review. *Am. J. Enol. Vitic.* **1973**, *24*, 5–9.
- (32) Towey, J. P.; Waterhouse, A. L. The extraction of volatile compounds from French and American oak barrels in Chardonnay during three successive vintages. *Am. J. Enol. Vitic.* **1996**, 47, 163–172.

Received for review February 21, 2007. Revised manuscript received May 25, 2007. Accepted May 29, 2007.

JF0705163