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CHESTNUT WOODEN BARRELS FOR THE AGEING OF WINE SPIRITS

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Background

This results of the study on the suitability of chestnut barrels for ageing of wine spirits was presented by the delegation of Portugal at the meeting of "Vitivinicultural Spirituous Beverages" experts group in April 2017.
According to the results of this 20-years long study, chestnut barrels show among others: improvement of the brandy's quality and complexity, higher contents of wood extractable compounds, more evolved colour (analytical and sensory), higher antioxidant activity, faster evolution of spirit, economic advantages.
Taking into account the importance of results presented, the experts group "Vitivinicultural Spirituous Beverages" decided to share them with the vitivinicultural community by publishing them as a collective expertise document on the website of the OIV.

Abstract

The production of high quality wine spirits implies an ageing period in wooden barrels, during which several phenomena occur and contribute to the acquisition of the desired chemical characteristics and sensory properties. These changes are mainly determined by the release of compounds from the wood into the distillate, which is closely dependent on the wood botanical species. The oak wood, especially from the French region of Limousin, is traditionally used in the ageing of wine spirits. Moreover, several studies made to evaluate the oak wood potential for cooperage and ageing of wine spirits reinforced its importance. In Mediterranean countries other species have been exploited for this purpose, especially the chestnut (Castanea sativa Mill.) that is used since the early Christian era, although no scientific study on its suitability for cooperage and ageing of wine spirits was known until 1995. The lack of information led our team to investigate these issues under a comprehensive approach, including the study of the chemical composition, chromatic characteristics, sensory properties and antioxidant activities of the wine spirits aged in chestnut and in oak wooden barrels, and their relationship with the chemical composition of the corresponding kinds of wood. The trials were performed to compare the effects of the botanical species, together with other relevant ageing factors: the toasting of the wood and the barrel size, during four and three years of ageing, respectively. The results obtained showed that wine spirits aged in chestnut wood presented a significantly higher content of total phenolics and of low molecular weight compounds analyzed by HPLC and GC (some of which are chemical markers), as well as higher antioxidant activities, relatively to the wine spirits aged in Limousin oak wood. The wine spirits aged in chestnut wood also exhibited more evolved chromatic characteristics, more complex sensory profile and higher overall quality that made them look older. Therefore, the use of chestnut wood can contribute to improve the sustainability of the ageing process, enhancing the quality of the aged wine spirits in a shorter period and with lower cost.

CHESTNUT WOODEN BARRELS FOR THE AGEING OF WINE SPIRITS

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1. Aim

Provide scientific evidence that the chestnut wooden barrels are suitable for the ageing of wine spirits.

2. Background

The production of high quality wine spirits implies an ageing period in wooden barrels, which corresponds to a minimum of six months for those produced out of delimited regions (Reg. EC nº 110/2008), one year for Armagnac (Déc. nº 2009 -1285), and two years for Cognac (Déc. nº 2009 -1146) and Lourinhã[1] wine spirits (Dec.-Lei nº 323/94).
During the ageing process in wooden barrels the wine spirit undergoes remarkable chemical and sensory modifications that determine its final quality. These changes are the consequence of several phenomena, namely the release of wood extractable compounds into the wine spirit (Puech *et al.*, 1984; Puech *et al.*, 1985). Among the wood extractable compounds, those of low molecular weight are of great importance due to their influence on the colour, aroma and flavour of the aged wine spirit (Léauté *et al.*, 1998). The release of wood compounds into the wine spirit is mainly determined by the wood botanical species (Miller *et al.*, 1992; Viriot *et al.*, 1993; Prida and Puech, 2006), together with the cooperage technology (Rabier and Moutounet, 1991), the barrel dimension (Guymon and Crowell, 1970; Singleton, 1995) and the cellar conditions (Cantagrel *et al.*, 1992).
Regarding the wood botanical species, several studies have been made to evaluate the oak wood potential for cooperage, especially the French and the American oaks, focusing their physical and mechanical properties as well as their chemical composition (Chatonnet and Dubourdieu, 1998; Zahri *et al.*, 2007). Therefore, the importance of these kinds of oak species was reinforced, and many authors (Singleton, 1995; Haluk and Irmouli, 1998) have support that only these species enhance the quality of the aged wine spirit. In addition, the oak wood, especially from the French region of Limousin (mostly *Quercus robur* L.), is traditionally used in the ageing of wine spirits, including the world's most recognized ones: Cognac and Armagnac.
However, in European countries bordering the Mediterranean Sea, the wood of other botanical species is also traditionally used for this purpose, namely the chestnut wood (*Castanea sativa* Mill.). This species has a great importance owing to historical, economical and social aspects of its cultivation (Conedera *et al.*, 2004). The widespread cultivation of chestnut in coppices for wood production starting in Western Europe from the early Middle Ages to 16th century (Pitte, 1986).

[1] Lourinhã is the unique Portuguese Geographical Denomination that exclusively produces aged wine brandies (DOC).

Some other references indicated that from the 18th century onwards the chestnut wood was very appreciated to make barrels for the storage and transportation of European wines (Taransaud, 1976), such as Porto wine (Filipe *et al.*, 1998).
To the best of our knowledge no scientific study on the suitability of chestnut for the ageing of wine spirits was made until 1995.

3. Research performed

The lack of scientific information led our team to investigate this issue under a broader approach (Belchior, 1995) in the last nineteen years. It was studied the influence of the wood botanical species used in cooperage together with other relevant ageing factors, such as the toasting level of the barrel, the barrel dimension, the cellar conditions and the ageing time, in the chemical composition, chromatic characteristics, sensory properties and antioxidant activities of the aged wine spirits, and their relationship with the chemical composition of the corresponding kinds of wood.

3.1. Characteristics of wine spirits aged in chestnut wood *versus* wine spirits aged in oak wood

3.1.1. Influence of the wood botanical species and the toasting level

For this purpose it was performed a study based on a three factorial design (five wood botanical species x three toasting levels x four years of ageing) with two replications. The heartwood staves of the different kinds of wood (chestnut from the Northern Portugal, oak from the French regions of Limousin and Allier, American oak and Portuguese oak) were seasoned in the open air during three years at a cooperage industry (J.M. Gonçalves Cooperage), in the Northern Portugal. Their anatomical study (Carvalho, 1998) allowed identifying the botanical species: the chestnut was *Castanea sativa* Mill.; the Limousin oak was *Quercus robur* L.; the Allier oak was *Quercus sessiliflora* Salisb.; the American oak was a mixture of *Quercus Alba* L./*Quercus stellata* Wangenh. and *Quercus lyrata* Walt./*Quercus bicolor* Willd.; the Portuguese oak was *Quercus pyrenaica* Willd. Staves of each botanical species were used to make six 250-L barrels. The barrels of each kind of wood were divided in three groups. The barrels of each group were then submitted to heat treatment, with one of the following toasting levels: light (QL), medium (QM) and heavy (QF).
The barrels were placed at Adega Cooperativa da Lourinhã in 1996 in similar cellar conditions and filled with the same Lourinhã freshly distilled wine spirit. The composition of the wine distillate was as follows: ethanol content - 76.9% v/v; total acidity - 44.9 g acetic acid/hL A.E. (absolute ethanol); acetaldehyde - 28.4 g/hL A.E.; ethyl acetate - 96.9 g/hL A.E.; methanol – 102.5 g/hL A.E.; higher alcohols – 363.2 g/hL A.E.
The total phenolic content (IFC) of the wine spirits was determined by the Folin–Ciocalteau method (Singleton and Rossi, 1965; Brun, 1979). The low molecular weight compounds were analyzed according to the methods developed and validated in our laboratory for HPLC (Canas *et al.*, 2003) and for GC (Caldeira *et al.*, 2004). The analytical colour parameters were determined by the CIELab method (Bakker *et al.*, 1986). The measurement of the *in vitro* antioxidant activity of the wine spirits was performed by the DPPH$^\bullet$ method (Canas *et al.*, 2008a).
A two-way analysis of variance (ANOVA) was performed to evaluate the statistical effect of the wood botanical species and the toasting level on the chemical composition of the wine spirits. Calculation of

least significant difference (LSD) was applied for comparison of the different averages. All the calculations were carried out using Statistica vs ’98 edition (Statsoft Inc., Tulsa, USA).
Considering the aim of the present manuscript, the results shown are related to the most important findings on the characteristics of the wine spirits aged in chestnut wooden barrels (CT) and in Limousin wooden barrels (LO), in the fourth year of ageing.
Interesting and coherent outcomes were obtained on the influence of these botanical species and toasting levels on the contents of the low molecular weight compounds - kinetics of extraction/oxidation (Canas, 2003; Caldeira, 2004) and on the sensory properties (Caldeira *et al.*, 2006a) of the wine spirits during the four years of ageing, as well as the effect of the toasting level on the low molecular weight compounds of the wine spirits (Canas *et al.*, 1999; Canas, 2003; Caldeira *et al.*, 2006b).
The results of ANOVA show significant differences of the chemical composition between wine spirits aged in chestnut and in oak barrels. The first ones can be clearly differentiated from the last ones by their very significantly higher phenolic content (IFC): 35.33 ± 6.62 and 12.12 ± 2.36, respectively (Belchior *et al.*, 2001).
Results of detailed analysis by HPLC (Table 1) and by GC (Table 2) demonstrates the richness of the wine spirits aged in chestnut barrels in the majority of the low molecular weight phenolic compounds. Hence, they present higher contents of gallic acid, vanillic acid, syringic acid and in ellagic acid - Table 1 - as well as in eugenol, 4-ethylphenol, and 4-allyl-syringol – Table 2.

Table 1. Content of low molecular weight compounds quantified by HPLC in four-years aged wine spirits in chestnut barrels and in Limousin oak barrels (concentrations in mg/L A.E., except for coumarins which are in μg/L A.E.).

	Effect	CT	LO	Interaction wood x toasting
5-Hydroxymethyl-2-furaldehyde (HMF)	*ns*	7.10 ± 4.71	5.98 ± 3.68	*ns*
Furfural	*ns*	17.32 ±15.47	18.25 ± 10.97	*ns*
5-Methyl-furfural	***	6.58 ± 1.01 *b*	1.94 ± 1.52 *a*	*
Gallic acid	***	222.33 ± 36.46 *b*	14.98 ± 1.74 *a*	**
Vanillic acid	***	7.97 ± 2.82 *b*	3.85 ± 1.53 *a*	*
Syringic acid	***	31.47 ± 9.14 *b*	5.70 ± 3.03 *a*	*
Vanillin	*ns*	10.27 ± 6.58	8.92 ± 4.65	*ns*
Syringaldehyde	*ns*	17.99 ± 13.14	19.79 ± 12.60	*ns*
Coniferaldehyde	*ns*	8.01 ± 4.95	9.98 ± 5.19	*ns*
Sinapaldehyde	**	9.08 ± 5.31 *a*	17.24 ± 10.83 *b*	*
Ellagic acid	**	85.47 ± 34.72 *b*	63.80 ± 17.28 *a*	*ns*
Umbelliferone	*ns*	0.011 ± 0.007	0.012 ± 0.01	*ns*
Scopoletin	***	0.11 ± 0.02 *a*	0.51 ± 0.14 *b*	*ns*

x ± s – mean ± standard deviation of six values; Means followed by the same letter in a row are not significantly different at 0.05*, 0.01** or 0.001*** level of significance; ns - without significant difference; CT – chestnut; LO – Limousin oak. Adapted from Canas *et al.* (2011)

Scopoletin is higher in the wine spirits aged in Limousin oak (Table 1), being recognized as a phenolic marker of oak wood (Tricard *et al.*, 1987).
Benzaldehyde is also higher in the wine spirits aged in Limousin oak but its content is below the olfactive detection threshold referred in the literature - 2 mg/L (Maga, 1985), which indicates that it does not contribute directly to wine spirit aroma, either in those aged in chestnut or in Limousin oak barrels (Table 2). Furthermore, the wine spirits aged in chestnut barrels can be differentiated from those aged in Limousin oak barrels by significantly higher contents of many aroma compounds (Table 2).

Table 2. Content of low molecular weight compounds quantified by GC in four-years aged wine spirits in chestnut barrels and Limousin oak barrels (concentrations in mg/L A.E.).

	Effect	CT	LO	Interaction wood x toasting
1-Butanol	*ns*	30.76 ± 3.80	28.17 ± 2.10	*ns*
2-Methyl-1-butanol + 3-Methyl-1-butanol	*ns*	3649.86 ± 326.93	3369.55 ± 214.59	*ns*
1-Hexanol	*ns*	30.58 ± 2.44	28.30 ± 1.82	*ns*
cis-3-Hexen-1-ol	*ns*	2.96 ± 0.25	2.72 ± 0.18	*ns*
trans-2-Hexen-1-ol	*ns*	0.72 ± 0.06	0.67 ± 0.04	*ns*
cis-2-Hexen-1-ol	*ns*	0.90 ± 0.38	0.95 ± 0.36	*
Benzyl alcohol	*	0.94 ± 0.14 *b*	0.79 ± 0.07 *a*	*ns*
2-Phenylethanol	**	26.74 ± 3.28 *b*	23.44 ± 1.36 *a*	*ns*
2-Methyl-1-propil acetate	**	1.65 ± 0.27 *b*	1.30 ± 0.14 *a*	*ns*
Ethyl butirate	*ns*	11.43 ± 1.33	11.54 ± 0.80	*ns*
Ethyl- 2-methylbutirate	*ns*	1.72 ± 0.18	1.65 ± 0.14	*ns*
3-Methyl-1-butyl acetate	***	10.50 ± 1.82 *b*	7.51 ± 0.65 *a*	*
Ethyl pentanoate	*ns*	0.27 ± 0.03	0.27 ± 0.01	*ns*
Ethyl hexanoate	*ns*	5.00 ± 0.49	4.86 ± 0.35	*ns*
Ethyl L-lactate	*ns*	162.67 ± 16.63	156.92 ± 7.83	*ns*
Ethyl octanoate	*ns*	12.43 ± 1.43	12.25 ± 0.94	*ns*
Ethyl decanoate	*ns*	2.61 ± 0.36	2.65 ± 0.30	*ns*
Diethyl succinate	*	9.35 ± 1.63 *b*	8.25 ± 1.19 *a*	*ns*
Ethyl dodecanoate	*ns*	0.32 ± 0.06	0.32 ± 0.05	*ns*
DL-Malic acid diethyl ester	*	4.49 ± 0.52 *a*	6.60 ± 1.30 *b*	*ns*
Acetic acid	*ns*	936.58 ± 117.13	841.83 ± 71.65	*ns*
Propanoic acid	*ns*	5.33 ± 0.48	5.61 ± 0.49	*ns*
Hexanoic acid	**	2.23 ± 0.29 *b*	1.85 ± 0.22 *a*	*ns*
Octanoic acid	*	6.99 ± 0.81 *b*	5.82 ± 0.44 *a*	*ns*
Decanoic acid	*	3.26 ± 0.39 *b*	2.77 ± 0.20 *a*	*ns*
Dodecanoic acid	*ns*	0.19 ± 0.04	0.19 ± 0.02	*ns*
Linalol	*ns*	1.35 ± 0.71	0.97 ± 0.34	*
α-Terpineol	**	0.34 ± 0.05 *b*	0.27 ± 0.04 *a*	*
4-Ethyl-guaiacol	*ns*	1.06 ± 0.15	0.96 ± 0.06	*ns*
Eugenol	***	0.90 ± 0.21 *b*	0.53 ± 0.12 *a*	*
4-Ethyl-phenol	*	1.12 ± 0.19 *b*	0.98 ± 0.08 *a*	*
Syringol	*ns*	0.33 ± 0.07	0.39 ± 0.13	*ns*
4-Allyl-syringol	**	0.48 ± 0.13 *b*	0.29 ± 0.07 *a*	*ns*
Benzaldehyde	*	0.44 ± 0.21 *a*	0.66 ± 0.04 *b*	*ns*
Acetoin	*ns*	7.95 ± 1.04	7.47 ± 0.43	*ns*
trans-β-Methyl-γ-octalactone	***	0.01 ± 0.00 *a*	1.12 ± 0.34 *b*	*ns*
cis-β-Methyl-γ-octalactone	***	0.00 ± 0.00 *a*	2.80 ± 0.49 *b*	*ns*

x ± s – mean ± standard deviation of six values; Means followed by the same letter in a row are not significantly different at 0.05*, 0.01** or 0.001*** level of significance; ns - without significant difference; CT – chestnut; LO – Limousin oak. Adapted from Canas *et al.* (2011)

These discriminating compounds belong to various chemical families, including higher alcohols (benzyl alcohol and 2-phenylethanol), esters (2-methyl-1-propil acetate or isobutyl acetate, 3-methyl-1-butyl acetate or isoamyl acetate, diethyl succinate), acids (hexanoic acid, octanoic acid and decanoic acid) and terpenes (α-terpeniol). Although many of these compounds do not possess intensive aroma features and probably do not contribute directly to wine spirit aroma, they may impact the perception of the aroma and flavour complexity (Ebeler *et al.*, 2000).

The DL-malic acid diethyl ester and the two isomers of β-methyl-γ-octalactone are chemical markers of the Limousin oak wine spirits.

The aforementioned higher alcohols, esters, acids and terpenes (Nikanen and Nikanen, 1991; Ebeler *et al.*, 2000) are already present in the freshly distilled wine spirit proceeding from wine as the result of yeasts metabolism, although their contents can change during the ageing period (Caldeira, 2004). Therefore, the highest contents found in the wine spirits aged in chestnut barrels should be related to more favourable ageing conditions promoted by this kind of wood inside the barrel, determining the extraction of wood compounds and other reactions in which the higher alcohols, esters, acids and terpenes are eventually involved. Despite these compounds are not derived from wood, their solubility and volatility in the wine spirit can be both influenced by the contents of wood-related non-volatile compounds and the ethanol/water structure of the wine spirit (Aishima *et al.*, 1992; Piggot *et al.*, 1992; Conner *et al.*, 1999).

Despite a short period of ageing, the wine spirits aged in chestnut wood stand out by their more evolved colour (lower lightness, higher saturation and higher coordinates a* and b*) than that of the wine spirits aged in Limousin oak wood (Table 3). These results confirm the trend observed during the first three years of ageing (Canas *et al.*, 2000a), and are in accordance with the positive correlations found between the phenolic content and the chromatic characteristics of the aged wine spirits (Escolar *et al.*, 1993; Canas *et al.*, 2000a).

Table 3. Chromatic characteristics of the wine spirits aged four years in chestnut barrels and in Limousin oak barrels.

	Effect	CT	LO	Interaction wood x toasting
L*	***	84.79 ± 5.66 *a*	92.14 ± 2.50 *b*	*
C*	***	58.10 ± 13.80 *b*	34.67 ± 8.72 *a*	*ns*
a*	**	3.20 ± 5.72 *b*	-1.75 ± 1.39 *a*	*
b*	***	57.82 ± 13.61 *b*	34.58 ± 8.81 *a*	*ns*

x ± s – mean ± standard deviation of six values; Means followed by the same letter in a row are not significantly different at 0.01** or 0.001*** level of significance; ns - without significant difference; CT – chestnut; LO – Limousin oak; L*- luminosity; C* - saturation; a* and b* - chromaticity coordinates. Adapted from Canas *et al.* (2011)

It is important to point out that the study of the antioxidant activity is of great importance owing to its relation to the nutraceutical quality of the wine spirit. To our knowledge, the research made by our team was the first that established the relationship between the wood botanical species or the toasting level and the antioxidant activity of the aged wine spirits over the time (Canas *et al.*, 2008a).
Regardless the toasting level, there is a highly significant effect of the wood botanical species on the antioxidant activity of the studied wine spirits. The wine spirits aged in chestnut wood presents higher antioxidant activity (% DPPH inhibition = 93.5 $\pm$ 0.91) than those aged in Limousin oak wood (% DPPH inhibition = 45.7 $\pm$ 2.38), which is positively correlated with their total phenolic content (r = 0.9990). Significant correlations were also found between antioxidant activity and gallic acid (r=0.9555) and ellagic acid (r=0.6138) contents, indicating that these compounds can greatly contribute to the overall antioxidant power of the wine spirits (Canas *et al.*, 2008a). Hence, the higher antioxidant activity exhibited by the wine spirits aged in chestnut wood can be partially due to the richness in gallic acid and ellagic acid. This hypothesis is supported by some studies that proved the strong antiradical activity of these phenolic acids even at very low concentrations in different aged wine spirits (Duriez *et al.*, 2001; Umar *et al.*, 2003).

Concerning the sensory analysis, it was carried out by a panel of 16 tasters selected and trained, and also assessed by its reliability (Caldeira *et al.*, 1999). The tasters previously generated colour, olfactory and gustatory attributes. The reliability of the attributes was evaluated (Caldeira *et al.*, 2002). The tasters were asked to score these attributes on a structured scale (0-no perception to 5–highest perception) and the overall quality of the wine spirits between 0 (without quality) and 20 (maximum quality). A two-way analysis of variance (ANOVA) was performed to evaluate the statistic effects of the wood botanical species and the toasting level on the sensory properties under study. Calculation of least significant difference (LSD) was applied for comparison of the different averages. All the calculations were carried out using Statgraphics vs 5.0 (STCT Inc., Rockville, USA).

The results show that the wood botanical species had a significant effect on the intensities of 14 attributes and overall quality. The discriminating attributes topaz, greenish, vanilla, woody, toasted, body, astringency, flavour evolution, flavour complexity, retronasal aroma and flavour persistence are positively correlated with the overall quality and the age of the wine spirits, while yellow-straw, golden and glue are negatively correlated with these parameters (Canas *et al.,* 2000a; Caldeira *et al.*, 2006a).

The wine spirits aged in chestnut barrels present more evolved sensory colour, characterized by higher intensity of topaz and lower intensities of golden and yellow-straw, while the wine spirits aged in Limousin oak barrels exhibit higher intensities of golden and yellow-straw hues (Figure 1).

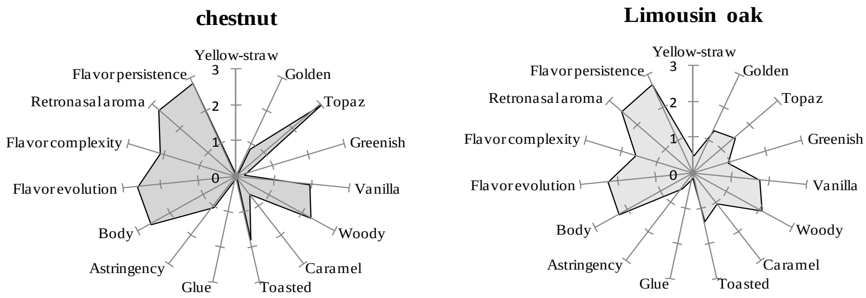

Figure 1. Sensory profiles with the average LSD attributes of wine spirits aged four years in chestnut barrels and in Limousin oak barrels. Adapted from Canas *et al.* (2011)

Interestingly, the colour perceived by the tasters (Figure 1) is coherent with the analytical colour of these wine spirits (Table 3), since the highest intensity of topaz found in the wine spirits aged in chestnut wood and the predominance of golden in those aged in Limousin wood, which correspond to the combination of a* (reddish colour) and b* (yellowish colour) highest values and lowest values, respectively.

As for the analytical colour, the sensory colour of the wine spirits reflect the differences in their chemical composition and a faster evolution induced by the chestnut wood during the first four years of ageing, as already verified in the first three years (Canas *et al.*, 2000a).

The wine spirits aged in chestnut barrels present the highest intensity of vanilla, which probably result from the slightly higher content of vanillin found in them (Table 1). This feature is noteworthy due to the remarkable role of this phenolic aldehyde as a key-odorant compound of the aged wine spirits (Caldeira et al., 2008). The woody attribute is also more pronounced in the wine spirits aged in chestnut barrels. This sensory property can be related with the highest content of vanillin in these wine spirits as well as the presence of other compounds like syringol (Table 2), as reported by Caldeira *et al.* (2008). The wine spirits aged in chestnut barrels can also be differentiated by the highest intensity of toasted aroma. In the above mentioned study, Caldeira *et al.* reported the relationship found by GC-O between this attribute and several compounds, namely 4-propylguaiacol, syringol, 4-methylsyringol and 4-allylsyringol. The same authors also found a strong correlation between the syringol and 4-allylsyringol contents and the toasted attribute. Indeed, the highest level of the last volatile phenol in the wine spirits aged in chestnut wooden barrels (Table 2) can partially explain the highest intensity of toasted aroma of these wine spirits.

The glue attribute is higher in the wine spirits aged in Limousin oak barrels, which can be related to the richness in some esters such as ethyl acetate, which tends to increase over the ageing time (Puech *et al.*, 1984).

Concerning the gustatory attributes, the wine spirits aged in chestnut are also more evolved, complex and persistent than those aged in Limousin oak. These sensory properties can derive from higher dry extract (Belchior *et al.*, 2001), total phenolic and low molecular weight extractable compounds of the wine spirits aged in chestnut wood (Tables 1 and 2).

As expected, the average overall quality is significantly higher in the wine spirits aged in chestnut barrels (14.0) than in those aged in Limousin oak barrels (13.3) owing to the highest scores of the attributes positively correlated with the overall quality and with the age. Few interactions between the wood and the toasting level were verified, which indicate the independence of these factors' effects on the sensory properties of the aged wine spirits, and confirm that the wood botanical species has a decisive role in the ageing process.

Based on the ANOVA results that contribute to the significant discrimination between the wine spirits aged in chestnut wood and those aged in Limousin oak wood, a principal component analysis was performed to achieve a global evaluation of the data. The three principal components, explaining 85.0 % of the total variance, make the splitting between the wine spirits aged in barrels of different kind of wood (Figure 2).

The first component (PC1), accounting for 61.9 % of the total variance, shows the separation of two main clusters: one formed by the wine spirits aged in chestnut wood that are located at positive values of PC1 and another constituted by those aged in Limousin oak wood that are placed at negative values of PC1.

PC1 has strong positive vectors loading for 5-methyl-furfural (Mfurf), gallic acid (Gall), vanillic acid (Van), isobutyl acetate (IsobAc), diethyl succinate (Diethsuc), hexanoic acid (HexAc), α-terpeniol (Terpen), eugenol (Eugen), 4-ethylphenol (EthPhen), 4-allylsyringol (AllylSyr), saturation (C*), chromaticity coordinates (a* and b*), total phenolic content (IFC), topaz (Topaz), greenish (Green), woody (Woody), toasted (Toast), body (Body), flavour evolution (Evol), flavour complexity (Complex) and flavour persistence (Persist), that are the most determining characteristics to distinguish the wine spirits aged in chestnut barrels from those aged in Limousin oak barrels.

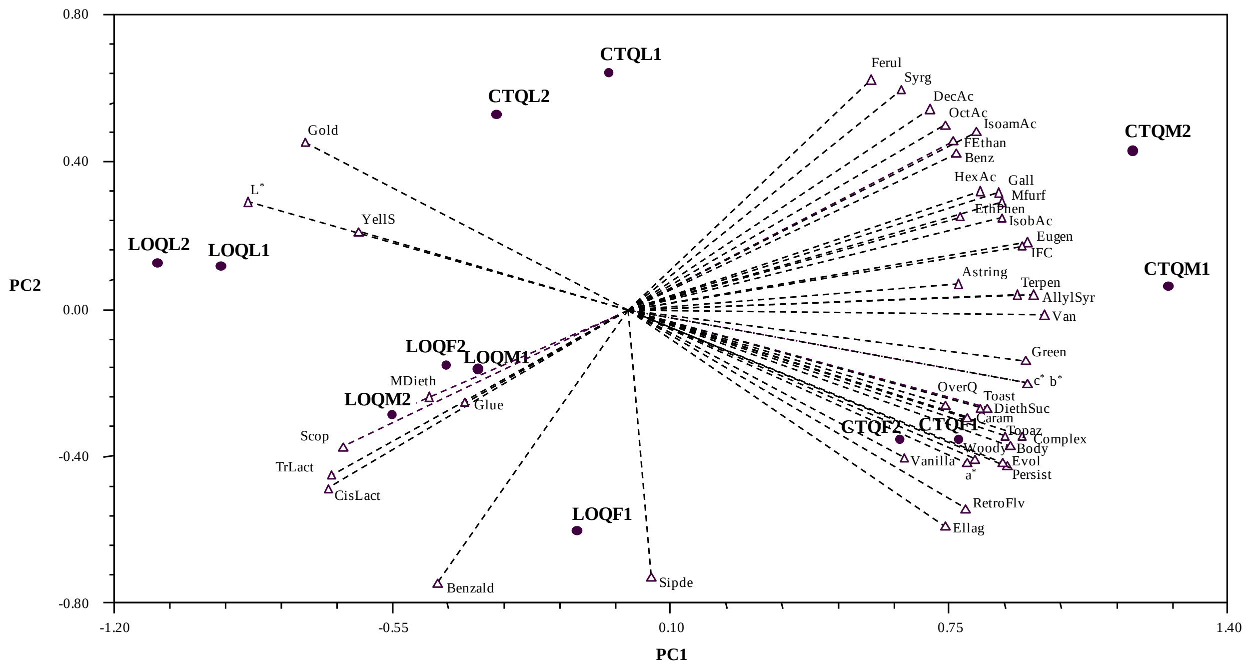

Figure 2. Projection of four-years wine spirits aged in chestnut barrels (CT) and Limousin barrels (LO) and total phenolic content, low molecular weight compounds contents, chromatic characteristics and sensory attributes in the space defined by the 1st and 2nd principal components: (●) wine spirits. Adapted from Canas *et al.* (2011)

PC1 also exhibits strong negative vector loading for luminosity (L*), scopoletin (Scop), *cis* and *trans* isomers of β-methyl-γ-octalactone (CisLact and TrLact), yellow-straw (YellS) and golden (Gold), which are closely related with the position of wine spirits aged in Limousin oak barrels.

3.1.2. Influence of the wood botanical species and the barrel dimension

For this purpose it was performed a study based on a two factorial design (two barrel dimensions x two wood botanical species) with three replications. The heartwood staves of chestnut (*Castanea sativa* Mill.) from the Northern Portugal and oak from the French region of Limousin (*Quercus robur* L.) were seasoned in the open air during three years at J.M. Gonçalves Cooperage (Palaçoulo, Portugal). The staves of each botanical species were used to make six barrels of 250-L and another six barrels of 650-L, all with heavy toasting. The barrels were placed at Adega Cooperativa de Lourinhã in similar cellar conditions and were filled with the same Lourinhã freshly distilled wine spirit. The composition of the wine distillate was as follows: ethanol content – 75.9% v/v; total acidity – 22.6 g acetic acid/hL A.E.; acetaldehyde - 5.3 g/hL A.E.; ethyl acetate - 62.6 g/hL A.E.; methanol – 86.8 g/hL A.E.; higher alcohols – 432.2 g/hL A.E.. Wine spirits sampling was made in the third year of ageing.

The chemical composition was assessed by the total polyphenol index - Ipt (Ribéreau-Gayon, 1970) and by the low molecular weight compounds quantified by HPLC (Canas *et al.*, 2003). The chromatic characteristics were determined by the CIELab method (Bakker *et al.*, 1986).

A two-way analysis of variance (ANOVA) was done to evaluate the wood botanical species and the barrel dimension effects on the chemical characteristics of the wine spirits. Calculation of least significant difference (LSD) was applied for comparison of the different averages. All the calculations were carried out using Statistica vs ’98 edition (Statsoft Inc., Tulsa, USA).

Regardless the barrel dimension, the contents of low molecular weight compounds found in the three-years aged wine spirits are similar to those observed in the first study (see 3.1.1.), except for furfural, 5-methyl-furfural, vanillic acid, syringic acid, sinapaldehyde and scopoletin (Table 4).

Table 4. Content of low molecular weight compounds quantified by HPLC in three-years aged wine spirits in chestnut barrels and in Limousin oak barrels (concentrations in mg/L A.E., except for coumarins which are in µg/L A.E.).

	Effect	CT	LO	Interaction wood x barrel dimension
5-Hydroxymethyl-2-furfural (HMF)	*ns*	7.48 ± 1.64	6.29 ± 1.45	*
Furfural	*ns*	22.60 ± 8.49	29.79 ± 10.85	*
5-Methyl-furfural	*ns*	1.87 ± 0.65	2.12 ± 0.72	**
Gallic acid	***	235.17 ± 28.74 *b*	19.26 ± 3.21 *a*	*ns*
Vanillic acid	*ns*	3.18 ± 1.12	4.52 ± 1.91	*ns*
Syringic acid	*ns*	5.89 ± 1.68	5.81 ± 1.29	*
Vanillin	**	6.88 ± 2.26 *b*	5.41 ± 0.61 *a*	**
Syringaldehyde	*ns*	15.63 ± 4.89	15.59 ± 2.31	*
Coniferaldehyde	*ns*	7.96 ± 2.59	7.48 ± 3.66	*ns*
Sinapaldehyde	*ns*	21.51 ± 7.13	24.69 ± 6.18	**
Ellagic acid	*	85.76 ± 16.20 *b*	65.11 ± 10.93 *a*	*ns*
Umbelliferone	*ns*	0.004 ± 0.001	0.005 ± 0.001	*ns*
Scopoletin	***	0.047 ± 0.016 *a*	0.153 ± 0.037 *b*	*ns*

x ± s – mean ± standard deviation of six values; Means followed by the same letter in a row are not significantly different at 0.05*, 0.01** or 0.001*** level of significance; ns - without significant difference; CT – chestnut; LO – Limousin oak. Adapted from Canas *et al.* (2011)

In spite of the effort made to reproduce the ageing conditions, these variations probably result from the combined action of the following factors: the botanical species (Canas *et al.*, 2000b), the toasting process (Canas *et al.*, 1999); the raw distillate used, and the ageing time (Canas, 2003). In addition, there is the effect of the barrel dimension (Belchior *et al.*, 2005; Canas *et al.*, 2008b).

Nevertheless, the HPLC data confirm that the wine spirits aged in chestnut barrels are significantly richer in low molecular weight phenolic compounds, namely gallic acid, vanillin and ellagic acid. Scopoletin is still higher in the wine spirits aged in Limousin oak (Table 4). The results strengthen the role of gallic acid and scopoletin as chemical markers of chestnut wood and Limousin oak wood, respectively.

The wine spirits aged in chestnut barrels still present the highest phenolic content (Table 5), which is in agreement with the higher content of low molecular weight phenolic compounds (Table 4).

Consequently, the chestnut wine spirits have a more evolved colour, as already verified (see 3.1.1.), regardless the second factor under study.

Table 5. Chromatic characteristics of the wine spirits aged three years in chestnut barrels and in Limousin oak barrels.

	Effect	CT	LO	Interaction wood x barrel dimension
Ipt	**	58.21 ± 23.49 *b*	29.77 ± 3.64 *a*	*ns*
L*	**	85.69 ± 2.42 *a*	90.86 ± 1.52 *b*	*ns*
C*	*ns*	51.81 ± 7.49	42.39 ± 10.43	*ns*
a*	*ns*	2.66 ± 2.24	4.32 ± 12.45	*ns*
b*	*	51.72 ± 7.38 *b*	41.12 ± 7.64 *a*	*ns*

x ± s – mean ± standard deviation of six values; Means followed by the same letter in a row are not significantly different at 0.05* or 0.01** level of significance; ns - without significant difference; CT – chestnut; LO – Limousin oak.
Adapted from Canas *et al.* (2011)

The sensory analysis and the statistical treatment of the sensory data were performed according to the methodology described in the first study (see 3.1.1.). The discriminating attributes for the wood botanical species are golden, topaz, greenish, vanilla, toasted, dried fruits, green, unctuous, flavour evolution, flavour complexity, retronasal aroma and flavour persistence. All of them are positively correlated with the overall quality and the age of the wine spirits, except golden and green (Caldeira *et al.*, 2006a).
The sensory outcomes confirm those of the first study (see 3.1.1.), showing that the wine spirits aged in chestnut wood present more evolved colour, more complex aroma and flavour profiles and higher overall quality that make them look older than those aged in Limousin wood (Figure 3).
It is evident the predominance of positive attributes in the wine spirits aged in chestnut wood, which are closely related to the richness in wood extractable compounds, regardless the barrel dimension. Among the attributes negatively correlated with the wine spirit's quality, only the green score, noted for the wine spirits aged in Limousin oak wood, had significant importance.
The sensory differences found in the aroma and flavour for each kind of wine spirit between the two studies presented are probably due to the factors cited to justify the differences observed in the chemical composition of these wine spirits.

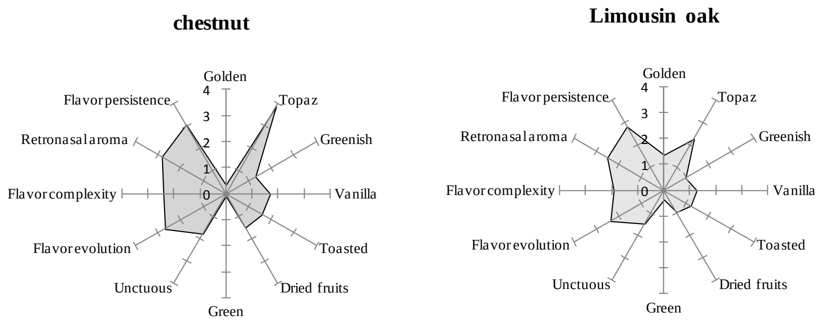

Figure 3. Sensory profiles with the average LSD attributes of three-years wine spirits aged in chestnut wood and in Limousin oak wood. Adapted from Canas *et al.* (2011)

3.2. Suitability of the chestnut wood for the cooperage

Complementarily, the study of the wood botanical species used in 3.1.1., demonstrate that the chestnut wood combines the required anatomical and mechanical properties, namely easy sawn, suitable flexibility, good thermal isolation, adequate hardness and durability, and slight porosity (Carvalho *et al.*, 1998). The absence of nodes is a crucial aspect to assure the leak-proof (Keller, 1987; Moutounet *et al.*, 1999), especially in the case of chestnut wood since the coopers often use wood with nodes (proceeding from the upper part of the trunk and/or from the branches) in order to improve the yield of younger trees and/or those from orchards (Carvalho A., personal communication). Therefore, a more appropriate forestry technology leading to the attainment of trees whose height is compatible with wood production and a proper procedure adopted by the coopers are possible ways to solve this problem.
Moreover, the chestnut wood present chemical characteristics that make it appropriate for cooperage, exhibiting a distinct chemical profile owing to the richness in phenolic compounds and in furanic derivatives (some of which are key-odorant compounds) and poorness in volatile compounds, regardless the toasting level (Canas *et al.*, 1999; Canas *et al.*, 2000b; Caldeira, 2004). These characteristics are partially related to the specific behaviour of chestnut wood during the seasoning operation (Canas *et al.*, 2006) and the heat treatment of the barrel (Canas *et al.*, 2007).

Recent studies made by Flamini *et al.* (2007), De Rosso *et al.* (2009) and Fernández de Simón *et al.* (2009) on the chemical composition of different kinds of wood confirm the interest of chestnut wood for the cooperage intending its use in the ageing of wines and wine spirits. This statement is supported by higher contents of total phenolics, tannins and several low molecular weight compounds with odorant power.

In addition to these more exhaustive studies, other scientific work performed by our team reinforces the adequacy of chestnut wood for the ageing of wine spirits: i) Study of the influence of wood botanical species on the extraction/oxidation and impregnation/evaporation kinetics of the wine spirits under model conditions (Canas *et al.*, 2002; Patrício *et al.*, 2005); ii) Study of alternative systems for the ageing of wine spirits (Canas *et al.*, 2009a,b; Caldeira *et al.*, 2010; Canas *et al.*, 2013).

4. Conclusions

The chestnut wooden barrels prove to be suitable for the ageing of wine spirits in different oenological conditions in the first years of ageing owing to suitable chemical composition given to the wine distillate, improving the chemical composition and enhancing the sensory properties of the aged wine spirit. Therefore the chestnut wood contribute to improve the complexity and the quality of the wine spirits, including the nutraceutical quality (health benefits that partially neutralize the harmful effect of alcohol for wine spirit’s consumer), inducing a faster evolution of the wine spirits, that is the wine spirits aged in chestnut wooden barrels show a chemical composition and a sensory profile closest to that of oldest wine spirits.

Moreover, the use of this kind of wood will allow the introduction of a competitive product in national and international markets - aged wine spirits of high quality and highly differentiated.

From the economic point of view, the ageing of wine spirits in chestnut wooden barrels is cheaper than in Limousin oak wooden barrels due to the lower price of the barrels, together with a faster evolution and the possible reuse more often.

The use of the chestnut wood for the ageing of wine spirits also imply the recovery of a typically species used in the Mediterranean countries for this purpose, which ecosystem have other remarkable collateral benefits related to its multifunctional role, providing other agro-forestry co-products and contributing to the social and economic development of some rural areas.

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