

## **EFFECT OF NATURAL SEASONING ON THE CHEMICAL COMPOSITION OF CHESTNUT WOOD USED FOR BARREL MAKING**

### **EFEITO DA SECAGEM NATURAL NA COMPOSIÇÃO QUÍMICA DA MADEIRA DE CASTANHEIRO USADA EM TANOARIA**

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#### **SUMMARY**

The ageing of high quality brandies requires a period of storage in wooden barrels. Several studies have been showed the great influence of the seasoning process on the characteristics of oak wood and their influence on wines and brandies quality. This study provides, for the first time, specific information about the seasoning effects on Portuguese chestnut wood from two different geographical origins, namely on the moisture content and chemical composition over eighteen months of open-air seasoning. Concerning the drying process, the chestnut wood can be used for the barrel making after six months of seasoning, but the remained period will be necessary for the maturation process. The seasoning time, the geographical origin and the stack layer have a significant effect on the moisture content, colour intensity, dry extract and content of some low molecular weight compounds of the chestnut wood studied.

**Key words:** chestnut wood, geographical origin, seasoning, chemical composition

**Palavras chave:** Madeira de castanheiro, origem geográfica, secagem, composição química

#### **INTRODUCTION**

The ageing of high quality brandies requires a period of storage in wood barrels. Oak and chestnut species have been largely used for this purpose, but nowadays chestnut is rarely used.

Several studies have shown the great influence of the technological operations involved in the barrel making, namely the seasoning, on the oak wood physical and mechanical properties, as well as on its chemical composition (Wilker and Gallander, 1989; Chatonnet *et al.*, 1994a,b; Chatonnet, 1995a; Masson *et al.*, 2000). This effect is essentially caused by the modifications on wood extractable compounds, especially those of low molecular weight that are released from wood to the distilled during the ageing process and contribute to the brandies sensory properties (Viriot *et al.*, 1993; Canas *et al.*, 2000a; Canas, 2003; Caldeira, 2004).

The seasoning is absolutely necessary to lower the wood moisture content from approximately 50% to 13 – 18%, in order to ensure the impermeability and the dimensional stability of the barrel (Chatonnet, 1992; Swan *et al.*, 1992; Chatonnet *et al.*, 1994a).

The air seasoning was the most common process. The length of air seasoning necessary for optimal barrel performance is dependent on the requirements of the cooperage and the end-users but this is typically from two to three years for oak wood (Chatonnet, 1992). Therefore, as a matter of economics, in the last twenty-years more coopers are using the kiln-drying process to shorten drying-time requirements (Wilker and Gallander, 1989; Swan *et al.*, 1992; Simpson, 1993; Kallander, 2003). However, the latter requires more intensive care in order to obtain similar results to those of natural seasoning (Kallander, 2003) and even though, the shortened drying-time and the interaction between the seasoning and the wood botanical species lead some authors to question its effect on the physical and chemical characteristics of the oak wood and its consequences on the aged wines (Dubois, 1989; Swan *et al.*, 1992; Chatonnet *et al.*, 1994a,b; Chatonnet, 1995b; Ward *et al.*, 1998; Masson *et al.*, 2000).

Actually, the seasoning of the wood is not a simple process of drying (Marché *et al.*, 1975) but a refining stage, since the wood undergoes slow chemical and biochemical transformations of biopolymers and extractable compounds by fungi and bacteria that contributes to its maturation (Chatonnet *et al.*, 1994a,b; Klumpers *et al.*, 1994; Larignon *et al.*, 1994; Chatonnet, 1995a; Roulland *et al.*, 1999).

The efficiency and the result of the seasoning are dependent on the characteristics of the park, and the constitution and maintenance of the stacks (Chatonnet, 1992; Sefton *et al.*, 1993; Chatonnet *et al.*, 1994a,b; Larignon *et al.*, 1994).

The characteristics of the process, its duration and place, as well as the age of the wood and the behavior of the different botanical species, turn seasoning into an important source of the chemical variability of the wood (Chatonnet, 1992; Francis *et al.*, 1992; Swan *et al.*, 1992; Sefton *et al.*, 1993; Chatonnet *et al.*, 1994a,b).

To our knowledge, there is no published work on the study of the seasoning effects on chestnut wood, namely on the evolution of low molecular weight compounds. The results of our previous studies have demonstrated that chestnut wood is suited for cooperage (Carvalho, 1998) and has interesting properties for the ageing of brandies (Canas *et al.*, 1999; Belchior *et al.*, 2001; Caldeira *et al.*, 2002; Canas, 2003; Caldeira, 2004). The chestnut wood presents a porous structure whose poses lower resistance to liquids and gases diffusion through the wood than oaks (Feuillat and Keller, 1997; Carvalho, 1998), influencing positively its behavior during the barrel making process (Canas, 2003) and increasing the wood permeability, and therefore the access of brandy to wood extraction sites for the release of extractable compounds. Furthermore, chestnut wood exhibits a higher amount of biopolymers, namely cellulose (Fengel and Wegner, 1989), and also a higher total content of low molecular weight phenolic compounds than oaks (Canas *et al.*, 2000b) and hydrolyzable tannins (Salagoity-Auguste *et al.*, 1992; Viriot *et al.*, 1994).

These characteristics have also positive repercussions on the chemical composition and sensory properties of the corresponding aged brandies, contributing to increase their quality and to shorten the ageing period (Canas *et al.*, 1999; Belchior *et al.*, 2001; Caldeira *et al.*, 2002; Canas, 2003; Caldeira, 2004).

So, this work is focused on the evolution of the chestnut wood composition over eighteen months of open-air seasoning and the influence of geographical origin and stack layer on these parameters, in order to achieve a better control of the process and, consequently, to improve the quality of the barrels and the aged wines and brandies.

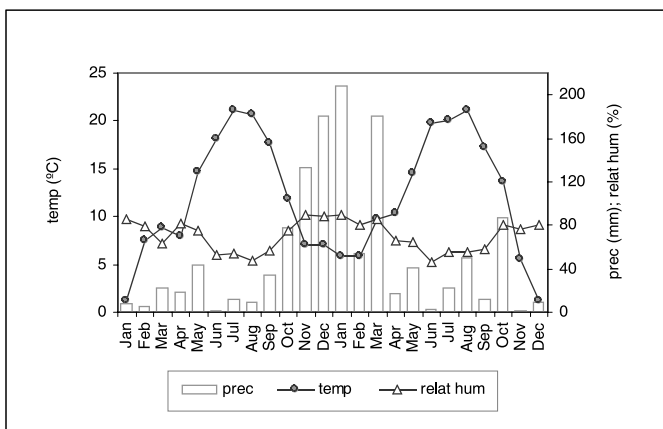
## MATERIALS AND METHODS

### *Wood origin*

The chestnut trees (*Castanea sativa* Mill.), each about 35 years old, were selected and cut in January 2000 from a 20 ha stand in a forest in the Northwest of Portugal (*Amarante* - Am) and from a 50 ha stand in a forest in the Northern of Portugal (*Carrazeda* - Carr).

### *Seasoning conditions*

The heartwood staves were obtained by sawing and were seasoned in the open air during eighteen months (from March 2000 to September 2001) at a cooperage industry (JM Gonçalves) in *Palaçoulo*, in the Northern of Portugal. Fig. 1 shows the climatic data of *Palaçoulo* region from January 2000 to December 2001.



**Fig. 1** - Evolution of precipitation (prec), air temperature (temp) and relative humidity (relat hum) from January 2000 to December 2001 in the region of Palaçoulo.

*Evolução da precipitação (prec), temperatura (temp) e humidade relativa (relat hum) entre Janeiro de 2000 e Dezembro de 2001 na região de Palaçoulo*

The staves were placed in stacks (one per geographical origin), with S-SW orientation, distanced at about 30 cm. Each stack contained approximately 25 parallel layers, with about 10-15 staves per layer, separated by sticks, and distanced of 15 cm from the floor. The stacks were irrigated in the summer and changed place within the park.

### *Sampling procedure*

The wood sampling was carried out in the control group ( $T_0$ ), after six months ( $T_6$ ), twelve months ( $T_{12}$ ) and eighteen months ( $T_{18}$ ) of seasoning. In each sampling time, two staves of each wood (two replicates) placed at the middle of the stack, were randomly selected. In addition, at  $T_6$ ,  $T_{12}$  and  $T_{18}$ , two staves of chestnut from *Amarante* placed at the top of the stack were also selected. So, the staves of each wood from the middle of the stack were used to analyse the effect of the geographical origin (total of 16 samples), and the staves of *Amarante* chestnut from the middle and the top of the stack were used to examine the effect of the stack layer (total of 12 samples). The entire thickness wood was cut, gathering the wood chips, making a homogenous group with them and then keeping the sample.

### *Determination of wood moisture (moist)*

Two pieces of wood (6 cm long, 2 cm width and 2 cm thickness) were cut from each stave and placed in an oven at  $103 \text{ }^{\circ}\text{C} \pm 2 \text{ }^{\circ}\text{C}$  until constant weight

to determine the wood moisture content (CT83, 1973).

#### *Wood extraction*

The wood chips (50 g) of each stave were grounded in a hammer-mill (Wiley, USA) and the maceration was carried out under rotary agitation for 180 min at 20 °C, with 1000 mL of an ethanol-water solution at 55% v/v adjusted to pH 4,2 with hydrochloric acid (Caldeira *et al.*, 2004). The wood extracts were filtered through a glass microfibre filter (Whatman GF/C, UK) on a Büchner funnel.

#### *Dry extract weight (DE)*

Usual method of OIV (OIV, 1994).

#### *Total polyphenol index (Ipt)*

Product of the measured absorbance at 280 nm by the dilution factor (Ribéreau-Gayon, 1970).

#### *Color intensity (A440)*

Measurement of absorbance at 440 nm (Belchior and Carvalho, 1983).

#### *HPLC analysis.*

The identification and quantification of phenolic compounds were carried out by HPLC as described by Canas *et al.* (2003). Identified compounds: Gallic acid (Gall), Vanillic acid (Van), Syringic acid (Syr), Ferulic acid (Ferul), Ellagic acid (Ellag), Coniferaldehyde (Cofde), Vanillin (Vanil), Syringaldehyde (Syrde), Sinapaldehyde (Sipde), Umbelliferone (Umb), and Scopoletin (Scop).

#### *GC analysis*

The liquid-liquid extraction of the wood extracts, and the quantification and identification by GC-MS were performed as described by Caldeira *et al.* (2004). Identified compounds: 5-Hydroxymethylfurfural (HMF), Furfural (Furf), Acetic acid (Acetic), Eugenol (Eugen), and 4-Allyl-syringol (Allyls).

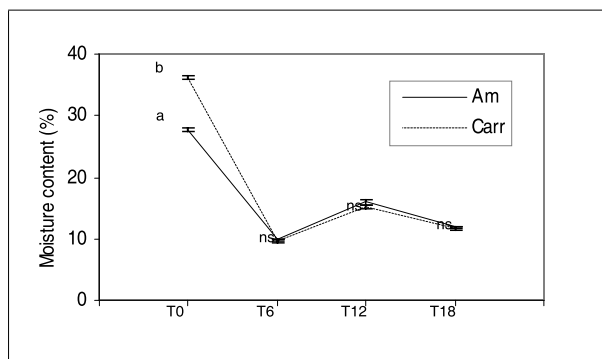
#### *Statistical analysis*

The variance analysis of the data was performed using Statistica vs '98 edition (Statsoft Inc., USA).

## RESULTS AND DISCUSSION

### *Effect on moisture content of the wood*

The average moisture content of the staves from *Amarante* and *Carrazeda* before drying was 28 and 36 %, respectively (Fig. 2).



**Fig. 2** - Evolution of moisture content in chestnut wood from *Amarante* (Am) and *Carrazeda* (Carr) during seasoning (points signed with different letters are significantly different at the 0.01\*\* level).

*Evolução da humidade da madeira de castanheiro de Amarante (Am) e de Carrazeda (Carr) durante a secagem (pontos assinalados com letras diferentes indicam diferença muito significativa\*\*)*

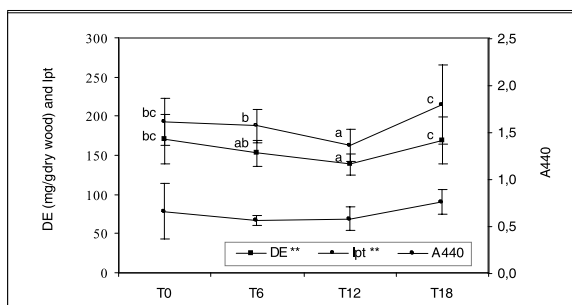
After *T6* the average moisture of the wood was already 9.5 to 10 %, and after that the equilibrium moisture content was reached. Concerning the drying process, the chestnut wood can be used for the barrel making after six months of seasoning, but the remained period will be necessary for the maturation process, as demonstrated by the changes in chemical composition of the wood (see next sections).

The length of air seasoning necessary for chestnut wood is considerable lower than that for oak wood (Chatonnet, 1992).

Furthermore, there is a very significant effect of the geographical origin on the initial moisture content of the wood. After *T6* there is a similar evolution pattern, and differences become not significant.

### *Effect on dry extract, polyphenol index and colour intensity of the wood*

The results of the variance analysis demonstrate that the seasoning time has a very significant effect on the dry extract and the polyphenol index of the wood extracts (Fig. 3). Their evolution profiles with time seem to be



**Fig. 3** - Evolution of dry extract, total polyphenols index and color intensity in chestnut wood extracts from *Amarante* (Am) and *Carrazeda* (Carr) during seasoning (points signed with different letters in a line are significantly different at the 0.01\*\* level).

*Evolução do extracto seco, índice de polifenóis totais e intensidade dos extractos de madeira de castanheiro de Amarante (Am) e de Carrazeda (Carr) durante a secagem (pontos assinalados com letras diferentes na mesma linha indicam diferença muito significativa\*\*)*

independent of the season conditions, and indicate the existence of two main phases. During the first twelve months there is a decrease, probably due to the rain leaching (Chatonnet, 1992; Chatonnet *et al.*, 1994a; Masson *et al.*, 2000), degradation by hydrolysis or oxidation (Chatonnet *et al.* 1994a; Klumpers *et al.* 1994; Viriot *et al.*, 1994) and/or polymerization/insolubilization of phenolic compounds (Peng *et al.*, 1991; Viriot *et al.*, 1994), as already observed in oak wood. After that there is an inversion of this tendency that suggests the predominance of the synthesis, as a consequence of biochemical and/or chemical reactions. According to Vivas and Glories (1996) this change could reflect the retention of extractable compounds within the wood owing to a lower moisture content and modification of the fibre conformation. Other authors (Chatonnet *et al.*, 1994a; Ward *et al.*, 1998) have stated that a certain period of time is needed to ensure the level of colonization and the change in dominance species with time that is compatible with the required chemical and anatomical modifications of the wood.

The evolution of the chemical composition is faster than that observed in Allier oak by Chatonnet *et al.* (1994a), suggesting that these phenomena are dependent on the botanical species and the seasoning conditions.

The evolution profile with time of the colour intensity seems to reflect some of the chemical modifications of the wood during the seasoning process, namely the oxidation of phenolic compounds that origins more coloured compounds (Haluk *et al.*, 1988; Chatonnet *et al.*, 1994a; Klumpers *et al.* 1994).

From the quantitative point of view, the dry extract, the polyphenol index and the colour intensity of the wood extracts are strongly influenced by the

geographical origin, which has a very significant interaction with the seasoning time on the dry extract and polyphenol index (Table I).

TABLE I  
Average and standard deviation of dry extract (mg/g dry wood), polyphenol index and colour intensity in chestnut wood extracts from different geographical origins  
*Média e desvio padrão do extracto seco (mg/g peso seco), índice de polifenóis totais e intensidade da cor dos extractos de madeira de castanheiro de diferentes origens geográficas*

		DE <sup>a</sup>	Ipt <sup>b</sup>	A440 <sup>c</sup>
<i>Effect</i>		**	*	**
<b>Am</b>	x	151.1 <b>a</b>	175.8 <b>a</b>	0.715 <b>b</b>
	SD	24.0	23.9	0.117
<b>Carr</b>	x	164.2 <b>b</b>	202.9 <b>b</b>	0.576 <b>a</b>
	SD	20.9	38.2	0.172
<i>origin x time</i>		**	**	ns

<sup>a</sup>Dry extract weight; <sup>b</sup>Total polyphenol index; <sup>c</sup>Color intensity;

x – mean of eight values; means followed by the same letter in a column are not significantly different at the 0.05\* or 0.01\*\* level.

On the other hand, the results of the seasoning process are also dependent on the stack layer, but there is no interaction between this factor and the seasoning time (Table II).

TABLE II  
Average and standard deviation of dry extract (mg/g dry wood), polyphenol index and colour intensity in chestnut wood extracts from *Amarante* as a function of the stack layer  
*Média e desvio padrão do extracto seco (mg/g peso seco), índice de polifenóis totais e intensidade da cor dos extractos de madeira de castanheiro de Amarante em função da posição na pilha*

		DE <sup>a</sup>	Ipt <sup>b</sup>	A440 <sup>c</sup>
<i>Effect</i>		**	**	ns
<b>Middle</b>	x	137.1 <b>b</b>	163.2 <b>b</b>	0.663
	SD	10.1	14.3	0.091
<b>Top</b>	x	103.0 <b>a</b>	125.1 <b>a</b>	0.623
	SD	8.4	13.7	0.100
<i>Stack layer x time</i>		ns	ns	ns

<sup>a</sup>Dry extract weight; <sup>b</sup>Total polyphenol index; <sup>c</sup>Color intensity.

x – mean of six values; means followed by the same letter in a column are not significantly different at 0.01\*\* level.

The wood placed in the middle of the stack presents the highest levels of dry extract and polyphenols, and consequently higher colour intensity than the wood placed at the top of the stack. These results show the strong influence of the stack layer on the chemical and biochemical phenomena that occur

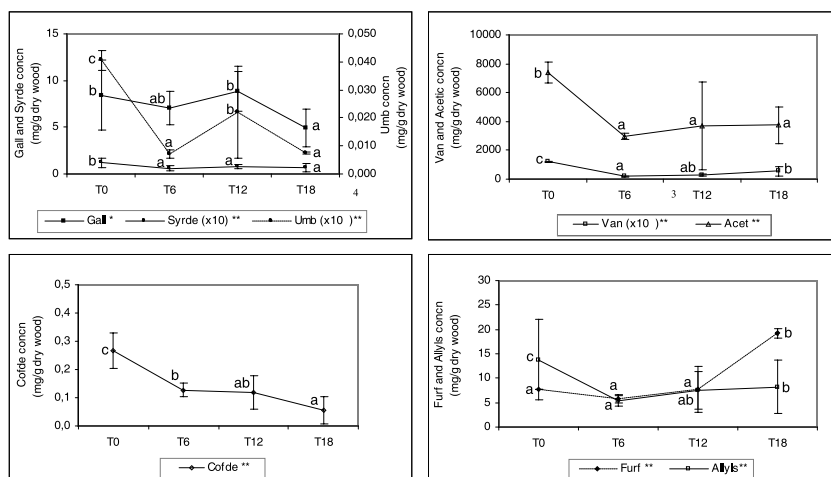


during the seasoning process. This aspect gives the scientific support for the requirement of regular change of the stacks referred by Larignon *et al.* (1994).

### *Effect on low molecular weight compounds of the wood*

The compounds were selected to reflect the changes taking place in the wood extractable constituents during the seasoning process, which have a great influence on the chemical characteristics and sensorial properties of the aged brandies (Canas *et al.*, 1999; Caldeira *et al.*, 2002; Caldeira 2004).

According to the variance analysis, the seasoning time affects very significantly the contents of vanillic acid, syringaldehyde, coniferaldehyde, umbelliferone, furfural, 4-allyl-syringol and acetic acid, and affects significantly the content of gallic acid (Fig. 4).



**Fig. 4** - Changes in the average concentration of low molecular weight compounds in chestnut wood extracts from *Amarante* (Am) and *Carrazeda* (Carr) during seasoning (points signed with different letters in a line are significantly different at the 0.05\* or 0.01\*\* level).

*Evolução dos teores médios de compostos de massa molecular baixa dos extractos de madeira de castanheiro de Amarante (Am) e de Carrazeda (Carr) durante a secagem (pontos assinalados com letras diferentes na mesma linha indicam diferença significativa\* ou muito significativa\*\*)*

Three different groups of compounds are identified according to their evolution pattern with time (Fig. 4): i) the first group consists of gallic acid, syringaldehyde and umbelliferone that presents a periodic variation; ii) the second group consists of vanillic acid, acetic acid, furfural and 4-allyl-syringol which contents decrease in the first six months, followed by a continuous increase with time; iii) the third group consists of coniferaldehyde which concentration has a continuous decrease during the seasoning period.

The results indicate a more pronounced influence of the season on the first group of compounds which contents seem to be related to biochemical reactions (Chatonnet *et al.*, 1994a) that are conditioned by the temperature and wood moisture content.

The behaviour of the second group seems to be independent of the season, and could reveal the predominance of the rain leaching (Chatonnet *et al.*, 1994a; Masson *et al.*, 2000) and the degradation phenomena, namely hydrolysis and or oxidation (Chatonnet *et al.*, 1994a) in the first six months, and the preponderance of the synthesis in the remained period. The results obtained by Masson *et al.* (2000) in oak wood also suggested that furfural and acetic acid, resulting from the hydrolysis of hemicelluloses, are independent of the drying conditions.

The content of coniferaldehyde demonstrate the prevalence of the degradation phenomena, especially oxidation, all over the seasoning period. Swan *et al.* (1992) reported a similar trend in American oak wood during 36 months of seasoning.

The concentrations of eugenol, HMF and scopoletin in chestnut samples vary only slightly (Table III) and most of these differences were not significant, but their evolution is similar to that of the first group.

TABLE III

Average and standard deviation of low molecular weight compounds concentration in chestnut wood extracts as a function of the seasoning time (mg/g dry wood, except for Scopoletin and volatiles which are in µg/g dry wood)

*Média e desvio padrão dos teores de compostos de massa molecular baixa dos extractos de madeira de castanheiro em função do tempo de secagem (mg/g peso seco, excepto para a escopoletina e compostos voláteis, em µg/g peso seco)*

		Syr <sup>a</sup>	Ferul <sup>b</sup>	Ellag <sup>c</sup>	Vanil <sup>d</sup>	Sipde <sup>e</sup>	Scop <sup>f</sup>	Eugen <sup>g</sup>	HMF <sup>h</sup>
<i>Effect</i>		<i>ns</i>	<i>ns</i>	<i>ns</i>	<i>ns</i>	<i>ns</i>	<i>ns</i>	<i>ns</i>	<i>ns</i>
<b>T0</b>	x	4.46	0.89	0.73	0.02	0.015	22.55	10.03	49.20
	SD	1.57	0.12	0.16	0.004	0.007	0.013	5.19	3.32
<b>T6</b>	x	2.71	0.71	0.90	0.01	0.018	20.00	6.71	22.62
	SD	1.48	0.13	0.07	0.003	0.004	0.003	2.55	8.53
<b>T12</b>	x	2.37	0.62	0.68	0.01	0.008	37.41	9.99	39.85
	SD	0.53	0.14	0.14	0.001	0.000	0.045	2.01	21.50
<b>T18</b>	x	2.13	0.41	0.98	0.01	0.013	23.41	8.87	27.08
	SD	1.82	0.06	0.12	0.008	0.003	0.017	0.87	16.04

<sup>a</sup>Syringic acid; <sup>b</sup>Ferulic acid; <sup>c</sup>Ellagic acid; <sup>d</sup>Vanillin; <sup>e</sup>Sinapaldehyde; <sup>f</sup>Scopoletin; <sup>g</sup>Eugenol; <sup>h</sup>5-Hydroxymethylfurfural.  
x = mean of four values.

The evolution pattern of eugenol is similar to that observed in Allier oak wood (Sefton *et al.*, 1993). Concerning the HMF, Masson *et al.* (2000) have also observed a decrease in the first three months of seasoning of oak wood. The strong resistance of cellulose to degradation could explain the behavior of this furanic derivative.

The amounts of syringic acid, ferulic acid and vanillin in chestnut samples present a little variation (Table III), with an evolution similar to that of the

third group. Contradictory results have been published regarding changes in the level of vanillin in oak woods during air-drying process, probably due to the strong effect of the botanical species and the seasoning conditions. Some authors (Chatonnet *et al.*, 1994b; Simon *et al.*, 1999; Cadahia *et al.*, 2001) reported a continuous increase in concentration, while others (Sefton *et al.*, 1993; Doussot *et al.*, 2002) observed insignificant changes during the seasoning period. Chatonnet *et al.* (1994a) consider that vanillin, like the other phenolic aldehydes, derives from lignin only by depolymerization and oxidation. The biochemical pathway suggest by Marché *et al.* (1975) is unlike because neither the different bacteria nor fungi isolated seem to be able to modify the lignin polymers (Chatonnet *et al.*, 1994a; Ward *et al.*, 1998).

The decrease of phenolic aldehydes contents is probably due to rain leaching, photodegradation and biochemical degradation, originating the corresponding phenolic acids. The subsequent chemical and biochemical degradation of these acids originates the corresponding volatile phenols (Chatonnet *et al.*, 1994b).

The amounts of ellagic acid and sinapaldehyde in chestnut samples also have a slight variation (Table III). These compounds show the inverse behavior of the first group.

The decrease of concentration of all the analyzed compounds in the first six months of seasoning seems to be associated with the great loss of water. In fact, when the wood is outside the hygroscopic domain (water content higher than 30%), there is a migration of the extractable compounds towards the surface of the staves where they are subsequently eliminated or modified, along with the migration and evaporation of the water (Masson *et al.*, 2000).

All of the studied compounds, except furfural and scopoletin, present a final concentration that is lower than the initial concentration. Simon *et al.* (1999) and Cadahia *et al.* (2001) pointed out the opposite effect for the majority of the low molecular weight compounds of different oak species.

The results of variance analysis (Table IV) show that the geographical origin has a very significant effect on the amounts of gallic acid, syringaldehyde, umbelliferone and furfural, and a significant effect on the amounts of coniferaldehyde, eugenol, acetic acid and HMF.

It is possible to verify that the chestnut wood from *Amarante* has lower content of all of the analyzed compounds than the chestnut wood from *Carrazeda*, except eugenol.

It is interesting to confirm that gallic acid is the most important compound in chestnut wood, being about 50% to 60% of all the phenolic acids (Canas *et al.*, 1999; Canas *et al.*, 2000b).

We also observe that independently of the geographical origin, the phenolic acids present the main contribution (97%) to the total content of low molecular weight phenolic compounds.

TABLE IV  
Average and standard deviation of low molecular weight compounds concentration in chestnut wood extracts as a function of the geographical origin and the stack layer (mg/g dry wood, except for coumarins and volatiles which are in µg/g dry wood)  
*Média e desvio padrão dos teores de compostos de massa molecular baixa dos extractos de madeira de castanheiro em função da origem geográfica e da posição na pilha (mg/g peso seco, excepto para a escopoletina e compostos voláteis, em µg/g peso seco)*

	Gall <sup>a</sup>	Van <sup>b</sup>	Syr <sup>c</sup>	Vanil <sup>d</sup>	Syrde <sup>e</sup>	Ferul <sup>f</sup>	Cofde <sup>g</sup>	Sipde <sup>h</sup>	Ellag <sup>i</sup>	Umb <sup>j</sup>	Scop <sup>k</sup>	Acetic <sup>l</sup>	Fur <sup>m</sup>	Eugen <sup>n</sup>	HMF <sup>o</sup>	Allys <sup>p</sup>
	**	ns	ns	ns	**	ns	*	ns	ns	**	ns	*	**	*	*	ns
<b>Am</b>	x 5.55 <b>a</b>	0.56	2.49	0.01	0.07 <b>a</b>	0.67	0.11 <b>a</b>	0.01	0.87	0.0015 <b>a</b>	0.25	3914.53 <b>a</b>	8.85 <b>a</b>	11.56 <b>b</b>	24.00 <b>a</b>	8.08
	SD 1.79	0.44	1.44	0.01	0.05	0.30	0.07	0.09	0.09	0.0012	0.23	2213.89	6.87	3.62	15.90	6.14
<b>Carr</b>	x 9.10 <b>b</b>	0.56	3.35	0.01	0.10 <b>b</b>	0.64	0.17 <b>b</b>	0.02	0.78	0.0024 <b>b</b>	0.59	4975.93 <b>b</b>	11.37 <b>b</b>	6.24 <b>a</b>	45.38 <b>b</b>	9.34
	SD 2.11	0.39	2.31	0.01	0.01	0.27	0.09	0.01	0.26	0.0017	0.66	2235.84	5.76	2.72	16.20	3.22
	<i>Origin effect<sup>q</sup></i>															
	ns	ns	ns	ns	**	ns	ns	ns	ns	*	ns	ns	**	ns	ns	**
<b>Middle</b>	x 5.48	0.34	2.20	0.01	0.04 <b>b</b>	0.63	0.08	0.27	0.94	0.0007 <b>a</b>	0.12	2928.16	9.21 <b>a</b>	10.33	14.49	4.22 <b>a</b>
	SD 1.96	0.31	1.49	0.01	0.01	0.31	0.05	0.63	0.09	0.0001	0.12	1721.28	7.50	3.35	5.67	0.70
<b>Top</b>	x 4.75	0.23	1.50	0.01	0.03 <b>a</b>	0.47	0.09	0.02	0.86	0.0011 <b>b</b>	0.17	2869.83	16.55 <b>b</b>	7.70	14.38	6.97 <b>b</b>
	SD 1.52	0.19	1.14	0.01	0.02	0.20	0.04	0.01	0.17	0.0002	0.14	984.50	11.12	2.13	3.52	1.50
	<i>Layer effect<sup>r</sup></i>															
<i>Origin x time</i>	ns	ns	ns	ns	**	ns	ns	ns	ns	**	ns	**	*	ns	ns	**
<i>Stack layer x time</i>	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	*	ns	ns	ns	ns

<sup>a</sup>Gallie acid; <sup>b</sup>Vanillic acid; <sup>c</sup>Syringic acid; <sup>d</sup>Vanillin; <sup>e</sup>Syringaldehyde; <sup>f</sup>Ferulic acid; <sup>g</sup>Coniferaldehyde; <sup>h</sup>Sinapaldehyde; <sup>i</sup>Ellagic acid; <sup>j</sup>Umbelliferone; <sup>k</sup>Scopoletin; <sup>l</sup>Acetic acid; <sup>m</sup>Furfural; <sup>n</sup>Eugenol; <sup>o</sup>5-Hydroxymethylfurfural; <sup>p</sup>4-Allyl-5-hydroxy-2-methylfuran-3-carboxaldehyde; <sup>q</sup>Origin; <sup>r</sup>Layer; x = mean of six values; means followed by the same letter in a column are not significantly different at the 0.05\* or 0.01\*\* level.

The results of variance analysis (Table IV) also indicate that the stack layer has only a very significant effect on the content of syringaldehyde, furfural and 4-allyl-syringol and a significant effect on the content of umbelliferone. The staves in the middle of the stack have the highest contents of the majority of the low molecular weight compounds analyzed. These results reinforce the effect of the stack layer on the modifications of wood chemical composition during the seasoning process.

So, in general, the seasoning has a great influence on the quality of chestnut wood, as referred for the oak wood by several authors (Marché *et al.*, 1975; Chatonnet, 1992; Chatonnet 1995a), although its specific effect is dependent on the geographical origin of the wood and the stack layer.

## CONCLUSIONS

This study provides, for the first time, specific information about the seasoning effects on chestnut wood from two different geographical origins. Furthermore, it gives the scientific support for the requirement of regular change of the stacks during the seasoning period.

The seasoning time, the geographical origin and the stack layer have a significant effect on the moisture content, colour intensity, dry extract and content of some low molecular weight compounds of the chestnut wood studied. Since the geographical origin can influence the physical and chemical composition of the wood, it is important to separate trees according to their origin and so the differences can be exploited.

The results obtained may allow a better understand of the process and, consequently, its optimization in order to maximize the economic profit and the quality of the wooden barrel by the coopers, as well as the quality of the brandies aged therein.

## RESUMO

### **Efeito da secagem natural na composição química da madeira de castanheiro usada em tanoaria**

O envelhecimento em vasilhas de madeira é indispensável para a obtenção de aguardentes de qualidade. Diversos estudos têm evidenciado o importante efeito do processo de secagem, a montante do fabrico da vasilha, nas características da madeira de carvalho e na qualidade de vinhos e aguardentes nela envelhecidos. O presente trabalho constituiu o primeiro estudo científico sobre os efeitos da secagem natural, durante dezoito meses, no teor de humidade e na composição química de madeira de castanheiro português, de duas origens geográficas distintas. Relativamente à perda de humidade, os resultados indicam que logo após seis meses de secagem a madeira de castanheiro pode ser utilizada no fabrico de vasilhas, no entanto o período remanescente é absolutamente necessário para assegurar o processo de maturação da madeira e, portanto, a sua aptidão para tanoaria. O tempo de secagem, a origem geográfica e a posição das aduelas na pilha de secagem são factores que afectam significativamente o teor de humidade, a intensidade da cor, o extracto seco e os teores de alguns compostos de massa molecular baixa das madeiras de castanheiro estudadas.

## RÉSUMÉ

### Effet sur sa composition chimique du séchage naturel du bois de châtaignier utilisé en tonnellerie

Le vieillissement en fûts de bois est indispensable pour l'obtention d'eaux-de-vie de qualité. Différentes études ont mise en évidence l'importance du processus de séchage, avant fabrication du fût, qui influence les caractéristiques du bois de chêne et la qualité des vins et des eaux-de-vie. Le présent travail est la première étude scientifique sur les effets du séchage naturel, pendant dix-huit mois, sur l'humidité et la composition chimique du bois de châtaignier portugais, de deux origines géographiques différentes. En ce qui concerne la perte d'humidité les résultats indiquent qu'après six mois de séchage le bois de châtaignier peut être utilisé pour la fabrication des fûts, bien qu'une période supplémentaire est absolument nécessaire pour permettre le processus de maturation. La période de séchage, l'origine géographique et la position des merrains dans la pile sont des facteurs qui influencent significativement l'humidité, l'intensité de la couleur, l'extrait sec et les teneurs en quelques composés de faible masse moléculaire des bois de châtaignier étudiés.

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