beverages

Review

# Phenolic Composition and Related Properties of Aged Wine Spirits: Influence of Barrel Characteristics. A Review

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**Abstract:** The freshly distilled wine spirit has a high concentration of ethanol and many volatile compounds, but is devoid of phenolic compounds other than volatile phenols. Therefore, an ageing period in the wooden barrel is required to attain sensory fullness and high quality. During this process, several phenomena take place, namely the release of low molecular weight phenolic compounds and tannins from the wood into the wine spirit. Research conducted over the last decades shows that they play a decisive role on the physicochemical characteristics and relevant sensory properties of the beverage. Their contribution to the antioxidant activity has also been emphasized. Besides, some studies show the modulating effect of the ageing technology, involving different factors such as the barrel features (including the wood botanical species, those imparted by the cooperage technology, and the barrel size), the cellar conditions, and the operations performed, on the phenolic composition and related properties of the aged wine spirit. This review aims to summarize the main findings on this topic, taking into account two featured barrel characteristics—the botanical species of the wood and the toasting level.

**Keywords:** wine spirit; ageing; wooden barrels; oak wood; chestnut wood; toasting level; phenolic composition; chromatic characteristics; sensory properties; antioxidant activity

## 1. Introduction

The aged wine spirit is one of the most representative alcoholic beverages, taking into account production, trade [1], and consumption [2] worldwide. Its manufacture has a long history and a relevant socioeconomic role in the traditional wine countries, mainly in Europe. Among them, it is worth mentioning France and its regions of Armagnac and Cognac that date back to the 15th and 16th centuries, respectively [3–5], producing the most prestigious and top-selling aged wine spirits. In this scenario, Portugal and its Lourinhã region should also be highlighted, whose historical references on wine spirit production date back to the early 20th century; it was delimited in 1992 as an exclusive denomination for aged wine spirits, like the above-mentioned French regions [6].

According to the European legislation [7], the wine spirit can be aged for at least one year in wood containers or for at least six months in wood containers with a capacity of less than 1000 L. For wine spirits with geographical denomination, the ageing period is at least one year for Armagnac [8], and two years for Cognac [9] and Lourinhã [10].

Actually, the freshly distilled wine spirit is characterised by a high concentration of ethanol and richness of volatile compounds, but is devoid of phenolic compounds other than volatile phenols [11]. Ageing in a wooden barrel (of oak, chestnut, ... ) is traditionally included in wine spirit production technology, being recognized as a crucial step for adding value to the product. During this process, the beverage undergoes important modifications and becomes a complex mixture of hundreds of compounds in an ethanol-water matrix [12], leading to sensory fullness and improvement of its quality. There is much to know about the chemistry underpinning the ageing of wine spirits, but the scientific community unquestionably accepts that those physicochemical and sensory changes result from several phenomena [13–21] such as:

- Direct extraction of wood constituents;
- Decomposition of wood biopolymers (lignin, hemicelluloses and cellulose) followed by the release of derived compounds into the distillate;
- Chemical reactions involving only the wood extractable compounds;
- Chemical reactions involving only the distillate compounds;
- Chemical reactions between the wood extractable compounds and the distillate compounds;
- Evaporation of volatile compounds and concentration of volatile and non-volatile compounds;
- Formation of a hydrogen-bonded network between ethanol and water.

Among these phenomena, the release of wood extractable compounds into the wine spirit, namely low molecular weight phenolic compounds and tannins, plays a decisive role in its chemical composition, sensory properties [22–24] and overall quality. In addition, oxidation reactions involving these compounds and those of the distillate are of paramount importance [25–29]. They are triggered by the slow and continuous diffusion of oxygen through the space between staves and through the wood [30–32].

The research carried out over the last decades has shown that the aforementioned changes are closely related to the action of factors ruling the ageing process, namely:

(i) The wooden barrel characteristics—the wood botanical species used, and the characteristics imparted by the cooperage technology (especially the seasoning/maturation of the wood and the heat treatment of the barrel), and the barrel size [33];
(ii) The cellar conditions—temperature, relative humidity and air circulation [18,34,35];
(iii) The technological operations performed during the ageing period, such as the refilling with the same wine distillate to offset the loss by evaporation [18,20,36], the addition of water to decrease the alcoholic strength [37], and stirring to homogenize the wine spirit and to enhance the extraction of wood compounds [38].

Concerning the resulting sensory properties, positive correlations between the phenolic composition and the color were established [23,39,40], which are in accordance with the findings in studies on Porto wine [41] and wine [42–46]. Notwithstanding the intricate effect of compounds on aroma and flavour owing to the complexity of their interactions and the multiple sensations involved [47,48], some features have been often related to the phenolic composition of the aged wine spirit and of other aged beverages. There is evidence on the relationship between: the vanilla aroma and vanillin concentration [24,49,50]; the bitterness and phenolic acids, their ethyl esters, and (+)-lyoniresinol concentrations [48,51–54]. Among these properties, the vanilla aroma should be highlighted due to its outstanding importance for aged wine spirit quality [55–57]. The relationship between astringency and ellagitannin and gallotannin concentrations is still unclear for thesekind of beverages [48,51–54,58,59].

In addition, the phenolic compounds (in this case almost exclusively extracted from the wood) exhibit a wide range of biological effects, many of which have been ascribed to their antioxidant activity. Several studies mention the antioxidant activity of some phenolic acids [60–69], phenolic aldehydes [70,71], coumarins [72], tannins [65,67,73–75], lignans [76], and of some volatile phenols [77]. This topic is of great relevance in a spirit drink, since the harmful effect of high alcoholic strength on consumer's health can be offset by the intake of such bioactive compounds [74,78–84]. Such benefits are expected in beverages that have undergone ageing in wood, especially wine spirits and whisky, but not in the traditional gins and vodkas [78].

Despite the knowledge acquired through different studies on the phenolic composition and related properties of the aged wine spirit modulated by the ageing technology, so far there have been no published articles systematizing it. This review assembles the main findings on the topic, taking into account two featured barrel characteristics—the wood botanical species and the toasting level.

## 2. Phenolic Compounds Found in Aged Wine Spirits

Several low molecular weight phenolics (Table 1) have been identified and quantified in aged wine spirits using high performance liquid chromatography (HPLC) or capillary electrophoresis (EC). All of them are non-flavonoid compounds; therefore, the phenolic composition of the aged wine spirit differs from that of wine and wine aged in wood, in which flavonoid and non-flavonoid compounds coexist [85,86].

**Table 1.** Low molecular weight phenolic compounds found in aged wine spirits.

| Class | Compound | Concentration Range * | References | Wine Spirits ** |
|---|---|---|---|---|
| Phenolic aldehydes | Sinapaldehyde | 0.05–42.31 | [13,15,29,38,74,87–105] | a,b,c,d,e,j,l |
| | Syringaldehyde | 0.20–34.20 | [13,15,17,29,38,74,87–94,98–105] | a,b,c,d,e,j,l |
| | Vanillin | 0.10–18.40 | [13,15,17,29,38,87–94,97–105] | a,b,c,d,e,j,k,l |
| | Coniferaldehyde | 0.05–12.94 | [13,15,29,38,74,87–89,91–93,98–100,102,103,105] | a,b,c,d,e,j,l |
| Phenolic acids | Gallic acid | 1.00–168.67 | [15,17,38,74,96,97,99,101,102,105] | a,c,f,k,l |
| | Ellagic acid | 3.90–104.00 | [38,74,97,99,101,102,105] | a,c,k,l |
| | Syringic acid | 0.40–17.18 | [15,17,29,38,74,88,89,99–102,105] | a,b,c,d,l |
| | Vanillic acid | 0.20–10.95 | [15,17,29,38,74,88,89,99–102,105] | a,b,c,d,l |
| | Ferulic acid | 0.05–9.94 | [15,88,89,102,105] | a,b,c |
| | Protocatechuic acid | 0.12–2.27 | [15] | a |
| | Coumaric acid | 0.02–1.20 | [15,88] | a,b |
| Coumarins | Scopoletin | 6.00–301.10 | [38,74,93–95,99,102] | a,b,c,e,f,g,h,i,l |
| | Umbelliferone | 0.11–7.00 | [38,93,102] | a,b,c |
| Lignans | Lyoniresinol | 3.40–17.50 | [92,99,106] | c,j,l |
| Phenyl ketones | Acetovanillone | 0.51–6.21 | [107] | a |

* Concentration in mg/L except for coumarins, which are in µg/L; the compounds are arranged in descending order of quantitative importance within each class; ** a—Cognac; b—Armagnac; c—Lourinhã; d—Moldovan; e—American; f—Spanish; g—Bulgarian; h—Canadian; i—Russian; j—Japanese; k—French; l—wine brandy.

Regardless the ageing conditions and the analytical methodologies employed, the results presented in Table 1 show that phenolic acids are the most abundant phenolic compounds in wine spirits, accounting for ca. 70% of low molecular weight phenolic compounds, followed by phenolic aldehydes (ca. 15%), lignans (ca. 12%), phenyl ketones (ca. 3%) and coumarins (0.1%) (Figure 1).

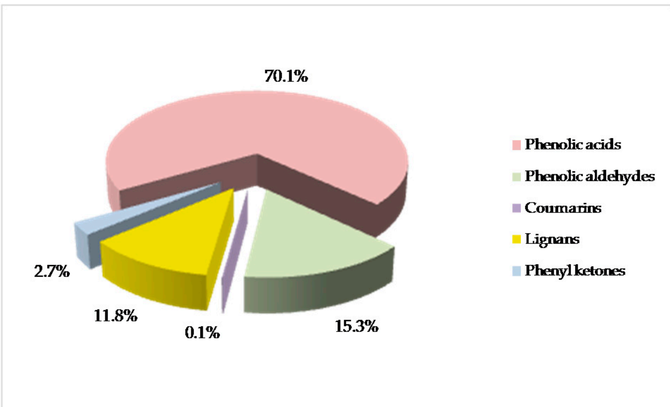



**Figure 1.** Relative importance of some phenolic classes in the aged wine spirits.

These acids, aldehydes and coumarins already exist in oak and chestnut heartwoods in the free form or linked to parietal constituents [108]. Nevertheless, their contents change considerably through the thermal degradation of the wood lignin [109–113] together with the increase of wood permeability [114,115] during the heat treatment of the barrel. Hence, higher amounts can be released into the wine spirit over the ageing process. Besides, lignin's hydrolysis occurring during ageing may also contribute to the enrichment in some phenolic aldehydes and phenolic acids [14,29,116].

Gallic acid and ellagic acid are the most representative phenolic acids, followed by syringic, vanillic, and ferulic acids. Protocatechuic acid and coumaric acid seem to be less important since they have not always been detected in the aged wine spirit.

The syringyl-type aldehydes (sinapaldehyde and syringaldehyde) are more plentiful than the guaiacyl-type aldehydes (vanillin and coniferaldehyde), probably due to the higher thermal stability of the former and their consequently greater accumulation in the toasted wood [117,118].

Among coumarins, the literature indicates scopoletin as more abundant than umbelliferone [38,93,102].

Concerning the lignans, few works report the presence of lyoniresinol in aged wine spirits. Despite the non-negligible amounts found, more advanced analytical conditions required for the separation and quantification of its two enantiomers [53,107,119] may justify their non-detection by HPLC under common chromatographic conditions. One of the enantiomers, (+)-lyoniresinol, was also quantified in oak wood [53,92,107], in which it remains stable under toasting until 200 °C [107,117].

As far as we know, acetovanillone was only quantified in small amounts in one study [106] and was also detected in toasted wood [14,120], being mainly formed by the thermal degradation of lignin [14,121].

In addition to the low molecular weight phenolic compounds, five hydrolysable tannins were identified and quantified in aged wine spirits by liquid chromatography coupled to mass spectrometry (LC-MS) and HPLC, respectively [122] (Table 2).

**Table 2.** Hydrolysable tannins found in aged wine spirits.

| Class | Compound | Concentration Range |
|---|---|---|
| Ellagitannins | Castalagin | 2.81–20.75 |
| | Vescalagin | 0.03–0.24 |
| | Roburin E | 0.08–0.19 |
| | Grandinin | 0.06–0.16 |
| Gallotannins | Monogalloyl-glucose | 0.47–5.95 |

Concentration in mg/L gallic acid. Adapted from [122].

The four monomeric ellagitannins and the monomeric gallotannin are derived from the wood (oak or chestnut), in which they are present in higher amounts, together with four dimeric ellagitannins (roburins A, B, C and D) [67,123–126], and with other dimeric and trimeric gallotannins [127,128]. Therefore, their low content or absence in the aged wine spirits may result from the thermal degradation during the heat treatment of the wood [117,126,129–131]. Moreover, low extraction [132], as well as oxidation and hydrolysis of ellagitannins, may occur during ageing [99,101].

Among the identified ellagitannins, castalagin and vescalagin are the most representative ones, followed by xylose and lyxose derivatives (roburin E and grandinin, respectively), as in the wood. Taking into account the total average content of soluble ellagitannins in Cognacs (4–840 mg/L; [15,101]) and in Armagnacs (155–702 mg/L; [22]), the quantified ellagitannins (Table 2) only represent ca. 3%, 0.03%, 0.03% and 0.03%, respectively; that is, other unidentified ellagitannins are likely to be extracted from the wood into the wine spirit and to have a greater contribution to the total amount. Actually, so far, few soluble tannins have been found in the aged wine spirit, contrasting with the increasing number of tannins and derived compounds detected in the aged wine [43,133].

Data from Tables 1 and 2 clearly illustrate the huge variability of the phenolic compounds concentrations in aged wine spirits. Indeed, it encompasses the effect of the analytical methodology used, but especially the impact of the barrel characteristics. Details on the last aspect and its repercussion on the chromatic characteristics, color, other related sensory properties, and antioxidant activity of the aged wine spirit are presented in the following sections.

## 3. Influence of the Wood Botanical Species

The wood most commonly used for the ageing of wine spirits is from the oak species *Quercus robur* L., principally from the French region of Limousin [37,134]. However, other wood botanical species have been increasingly studied to evaluate their potential for the cooperage, focusing their chemical composition: *Quercus sessiliflora* Salisb., particularly from the French region of Allier, and *Quercus alba* L., mainly from North America [67,108,135,136]; *Quercus pyrenaica* Willd., grown in Mediterranean countries [67,108,124,126,137]. Chestnut wood (*Castanea sativa* Mill.) has also been exploited for this purpose, and is of particular significance in the countries bordering the Mediterranean Sea due to historical, economical, and social aspects of its cultivation [138]. Its suitability for the cooperage aiming the ageing of wine spirit has also been investigated [6,102,108,120,139,140].

### *3.1. Phenolic Composition*

Considerable attention has been devoted by several research teams to the chemical composition of wood used in oenology. However, only a limited number of studies about its impact on the chemical composition of the aged wine spirit have been published. Moreover, their experimental designs are not always fully described, hindering the comparison of results obtained in different approaches. To the best of our knowledge, the exception lies in four older works that examined the effect of one kind of wood. Baldwin et al. [13] found low levels of vanillin (ranging from 0.6 to 1.5 mg/L), syringaldehyde (varying from 1.2 to 7.6 mg/L), coniferaldehyde (ranging from 0.3 to 1.8 mg/L), and sinapaldehyde (ranging from 0.2 to 3.4 mg/L) in American wine spirits aged in new barrels of American oak. Similarly, Nabeta et al. [92] reported low average contents of vanillin (0.6 mg/L), syringaldehyde (0.35 mg/L), coniferaldehyde (1.4 mg/L), and sinapaldehyde (0.8 mg/L) in Japanese wine spirits aged over a six-year period in new barrels of French oak wood. Tricard et al. [94] also observed low amounts of vanillin (2.28 mg/L) and syringaldehyde (6.60 mg/L), but a considerable amount of scopoletin (109 mg/L), in Cognacs aged over a seven-year period in new barrels of French oak. In contrast, Puech and Moutounet [99] found higher contents of vanillin (8.2 mg/L), syringaldehyde (19.4 mg/L), coniferaldehyde (17.8 mg/L), and sinapaldehyde (19.8 mg/L) in wine brandies aged over a seven-year period in new barrels of Limousin oak. They also found high contents of ellagic acid (62.9 mg/L), gallic acid (31.0 mg/L), vanillic acid (7.9 mg/L), and syringic acid (8.2 mg/L).

The most recent works [6,56,105] were based on a factorial design using the same wine distillate from the Lourinhã region (produced by Adega Cooperativa da Lourinhã) aged over a four-year period in barrels made from the following kinds of wood: Limousin oak (*Q. robur* L.) and Allier oak (*Q. sessiliflora* Salisb.) from French forests; American oak (mixture of *Q. alba* L./*Q. Stellata* Wangenh. and *Q. lyrata* Walt./*Q. bicolor* Willd.) from Pennsylvania/USA; Portuguese oak (*Q.pyrenaica* Willd.) and chestnut (*C. sativa* Mill.) from the North of Portugal. The 250 L barrels were supplied by J. M. Gonçalves cooperage (Palaçoulo, Portugal) and were placed in the cellar of Adega Cooperativa da Lourinhã in similar environmental conditions.

Significant differences in the contents of the majority of low molecular weight phenolic compounds of the aged wine spirit according to the wood used were observed (Table 3). Chestnut wood induced the highest content of phenolic acids in the wine spirit, especially of gallic acid and ellagic acid, as noticed for the ageing of red wine [141,142]. The highest levels of vanillin and syringaldehyde were also found in the wine spirit aged in chestnut barrels. Portuguese oak wood promoted intermediate enrichment, with the highest level of sinapaldehyde, while the other kinds of oak had a weaker performance. However, the richness of coumarins associated with the American oak should be stressed, because these compounds can act as chemical markers of this kind of wood [56,95].

**Table 3.** Mean concentrations of low molecular weight compounds in wine spirits aged four years in different kinds of wood.

| Compound | American oak | Allier Oak | Limousin Oak | Portuguese Oak | Chestnut |
|---|---|---|---|---|---|
| Ellagic acid | 32.45 [a] | 37.19 [a] | 49.38 [b] | 81.16 [c] | 91.27 [d] |
| Gallic acid | 10.49 [a] | 13.46 [a] | 11.52 [a] | 37.80 [b] | 218.19 [c] |
| Vanillic acid | 2.97 [b] | 2.04 [a] | 2.62 [a,b] | 2.96 [b] | 6.15 [c] |
| Syringic acid | 3.56 [a] | 3.58 [a,b] | 4.09 [a,b,c] | 5.03 [c] | 19.77 [d] |
| Ferulic acid | 2.97 [a] | 2.85 [a] | 3.03 [a] | 6.06 [b] | 6.39 [c] |
| Vanillin | 6.21 [a,b] | 5.50 [a] | 6.33 [b] | 6.41 [b] | 8.28 [c] |
| Syringaldehyde | 15.06 [b] | 11.73 [a] | 15.02 [b] | 14.94 [b] | 15.89 [b] |
| Coniferaldehyde | 9.04 | 8.27 | 9.12 | 8.75 | 7.78 |
| Sinapaldehyde | 16.71 [b] | 14.63 [a,b] | 16.76 [b] | 19.65 [c] | 11.94 [a] |
| Umbelliferone | 1.48 [c] | 0.78 [a] | 0.95 [b] | 0.98 [b] | 0.92 [b] |
| Scopoletin | 164.77 [d] | 19.74 [b] | 37.12 [c] | 10.33 [a] | 8.63 [a] |
| ΣLMW | 122.68 [a] | 127.68 [a] | 144.03 [a] | 224.0 [b] | 395.93 [c] |

Concentration in mg/L absolute ethanol except for coumarins, which are in µg/L absolute ethanol; mean values ($n$ = 24) followed by different letters ([a], [b], [c], [d]) in a row are significantly different ($p < 0.05$); ΣLMW—Sum of low molecular weight phenolic compounds concentrations. Adapted from [56].

Comparing the results of the oldest and most recent works, important differences in the concentrations of phenolic aldehydes, phenolic acids and scopoletin in wine brandies aged in French oak wood and American oak wood are observed. They may express the effect of the geographical origin of the wood, as well as the interaction between the kind of wine distillate and the wood, in the extraction kinetics of such compounds. Despite the observed variability, the American oak wood had a lesser contribution to the phenolic composition of the aged wine spirit. Taking into account the results of Puech and Moutounet [99], the performance of Limousin oak wood resembled that of Portuguese oak wood.

The phenolic differentiation of wine spirits was ascribed to the pool of phenolic compounds in the different kinds of wood under study [108,126,140,143] and to lignin hydrolysis during ageing [14,29,102,116,144]. Furthermore, gallic acid and ellagic acid can be directly extracted from the wood, or derived from the hydrolysis of gallotannins [112] and ellagitannins [101], respectively, especially in the first years of ageing [22,101].

Comparing the hydrolysable tannins of the wine spirit aged in Limousin oak wood and in chestnut wood, Canas et al. [122] did not observe significant differences in their contents (Table 4). Monogalloyl-glucose was only present in the wine spirit aged in chestnut barrels, as in the corresponding wood [123], although detection of this monomeric gallotannin in *Quercus robur* wood already made by other authors [127]. The robustness of the methods used in the isolation and quantification of the tannin fraction pointed to the conclusion that the above-mentioned effect could be caused by the wood intraspecific variability [145].

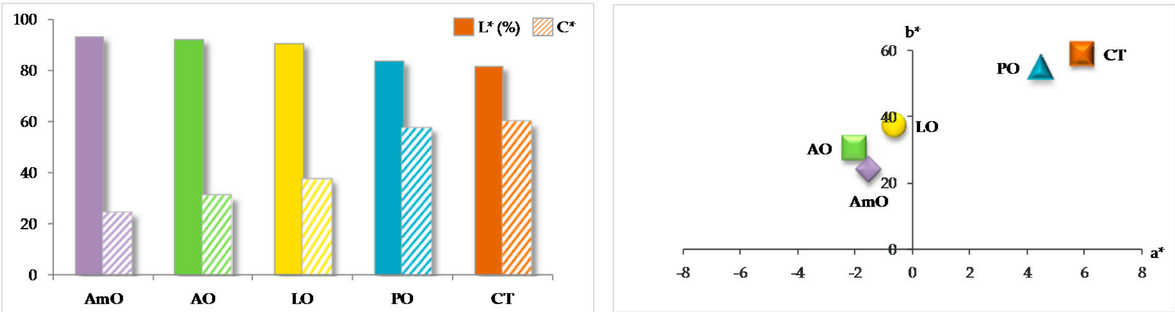
**Table 4.** Mean concentrations of hydrolysable tannins in wine spirits aged four years in different kinds of wood.

| Compound | Limousin Oak | Chestnut |
|---|---|---|
| Castalagin | 12.07 | 6.33 |
| Vescalagin | 0.11 | 0.17 |
| Roburin E | 0.14 | 0.12 |
| Grandinin | 0.12 | 0.14 |
| Monogalloyl-glucose | nd | 5.16 |

Concentration in mg/L gallic acid; mean values ($n$ = 9); nd—not detected. Adapted from [122].

### 3.2. Chromatic Characteristics and Sensory Properties

The color is a core element in the perception of wine spirit quality, as for other beverages [146]. It determines the first impression and rules the consumer's choice [147]. Among the few authors who studied it in aged wine spirits [39,40,148], Canas et al. [23] remarked that the chromatic characteristics (CIELab parameters) are closely related to the kind of wood used (Figure 2). In this investigation [23], the wine spirit aged in chestnut wood stood out by its more evolved color than the wine spirits aged in oak wood. It displayed higher color intensity (lower L*), higher red (a*) and yellow (b*) hues, and higher saturation (C*) that made it look older than the latter. Indeed, there is scientific evidence about the color evolution of wine spirits over the ageing time, which is marked by a decreasing of lightness and an increasing of saturation, red hue and yellow hue [148]. Among the wine spirits aged in oak wood, the one aged in Portuguese oak exhibited greater evolution of the chromatic characteristics than those aged in Limousin oak, American oak and Allier oak. Once the wine distillate itself is colorless due to the absence of phenolic compounds other than volatile phenols [11], the observed changes are assigned to the wood stage. Recently, Rodríguez-Solana et al. [149] reported concordant results for a grape marc spirit aged in *Q. robur*, *Q. alba* and *Q. petraea* wooden barrels.




(a) (b)

**Figure 2.** Mean values ($n$ = 24) of chromatic characteristics of the wine brandies aged in different kinds of wood: (**a**) lightness (L*) and saturation (C*); (**b**) chromaticity coordinates (a*, b*); AmO—American oak; AO—Allier oak; LO—Limousin oak; PO—Portuguese oak; CT—Chestnut. For each chromatic characteristic, the differences between the aged wine spirits are very significant ($p < 0.01$). Adapted from [23].

The different pool of phenolic compounds and extraction kinetics (Table 3), the oxidative phenomena occurring during ageing [26,27,101], and the condensation reactions between phenolic compounds promoted by each kind of wood possibly accounted for the differences in the chromatic characteristics. Condensation reactions between tannins mediated by acetaldehyde (resulting from ethanol oxidation and which may represent more than 90% of the aldehydes' content of the aged wine spirit [14,16]), by phenolic aldehydes and furanic derivatives, such as furfural and 5-hydroxymethylfurfural, are likely to happen, as in wine during ageing [146,150–152].

Interestingly, as described in the same work [23], the color perceived by the tasters (Figure 3) was consistent with the chromatic characteristics of the aged wine spirits (Figure 2).

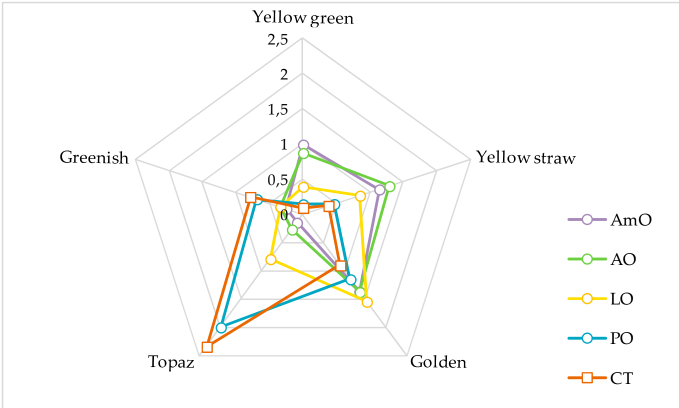



**Figure 3.** Color profile based on the average panel score's attributes of the wine spirits aged four years in different kinds of wood. For each color attribute, the differences between the aged wine spirits are very significant ($p < 0.01$). Adapted from [23].

The wine spirits aged in chestnut barrels were characterized by a more evolved color owing to the highest intensities of topaz (orange, amber color), greenish color, and the lowest intensities of golden and yellow straw (Figure 3). The wine spirit aged in Portuguese oak wood had high intensity of topaz and golden while those aged in the other kinds of oak wood showed higher intensities of golden, yellow straw and yellow green. Topaz is the main color of older wine spirits, resulting from the combination of higher positive values of chromaticity coordinates a* (red hue) and b* (yellow hue), whereas the golden prevails in wine spirits with less ageing time. Therefore, as for the analytical color, the sensory color of the wine spirits expressed the differences in their phenolic composition.

Examining other sensory properties of the wine spirits, Caldeira et al. [153] found significantly higher intensities of vanilla and astringency underlying the cluster formed by the wine spirits aged in Portuguese oak and chestnut. For these attributes, the wine spirits aged in Allier oak and American oak presented the opposite features, while those aged in Limousin oak showed an intermediate profile. Similar results were obtained for grape marc spirits aged in *Q. robur*, *Q. alba* and *Q. petraea* barrels [149]. The observed behavior for the vanilla aroma is ascribed to the vanillin content (Table 2), while the astringency seems not be related to the content of hydrolysable tannins (Table 3).

### 3.3. Antioxidant Activity

The antioxidant activity of some spirit drinks, namely the aged wine spirit, has aroused the interest of the researchers. Indeed, the "French Paradox" showed that for apparently the same level of risk factors, cardiovascular mortality rate is lower in France than in the European Northern countries [154]. Moreover, among the French regions, this phenomenon was particularly evident in the southwest France, a region where people do not drink more wine than elsewhere, but often drink Armagnac [155]. Therefore, some research was conducted to evaluate the antioxidant activity of Armagnacs and Cognacs [74,78,80] as well as of Spanish brandies [82,156], revealing its positive correlation with the ageing time. Later on, the influence of the wood botanical species used was examined [122]. Similar studies were also done with brandies [84,157] but the corresponding experimental designs raise many doubts, and do not allow comparison of outcomes with those obtained for the aged wine spirits.

Studying the in vitro antioxidant activity of the same wine spirit aged in chestnut barrels and in Limousin oak barrels, Canas et al. [122] concluded that the wood botanical species induced significantly different antioxidant activity (measured by 1,1-diphenyl-2-picrylhydrazyl radical scavenging activity—DPPH) regardless the toasting level. The antioxidant activity promoted by the chestnut wood (DPPH inhibition = 93.5%) was two-fold higher than that promoted by Limousin oak wood (DPPH inhibition = 45.7%)—Figure 4.

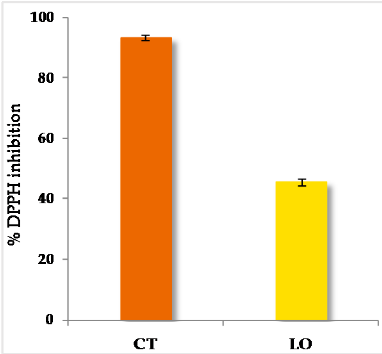



**Figure 4.** Mean values of antioxidant activity of the wine spirits aged in different kinds of wood (LO—Limousin oak; CT—chestnut). Adapted from [122].

This effect is explained by the highest phenolic content of the wine spirit aged in chestnut barrels; synergistic phenomena [81,158] and antagonistic phenomena [159] between individual phenols are also likely to occur. The antioxidant activity was mainly correlated with gallic acid ($r$ = 0.9555) and ellagic acid ($r$ = 0.6138), whose contents were significantly higher in the wine spirit aged in chestnut wood (Table 3). Actually, these phenolic acids are well-known bioactive compounds [61,62,66,69,160]. Syringaldehyde may also have contributed to this feature [70,71], especially in the wine spirit aged in chestnut barrels. It is interesting to note that a similar relationship of the antioxidant capacity was reported by Rodríguez Madrera et al. [143] for *C. sativa* and *Q. robur* wood extracts based on the levels of phenolic acids and phenolic aldehydes. Regarding the role of hydrolysable tannins in the observed behavior, no significant correlations were pointed out by Canas et al. [122] for individual compounds and for the total content. However, strong correlations between the antioxidant capacity and monomeric ellagitannins (castalagin, vescalagin, roburin E and grandinin), and dimeric ellagitannins (roburins A–D) were emphasized for wood extracts including *C. sativa* and *Q. robur* [67]. On the other hand, Da Porto et al. [74] stated that ellagitannins are the major contributors to the overall antioxidant activity of Cognac. So, this kind of discrepancy could be justified by the variability of wood composition [145] plus the variability induced by the wood heat treatment together with differential extraction from the wood and reactions involving ellagitannins during ageing, as aforementioned.

## 4. Influence of the Heat Treatment of the Barrel

Research on oak wood and chestnut wood have shown that the heat treatment of the barrel is of remarkable importance to the pool of extractable compounds that can be released into the beverage when it contacts the wood [102,109–113,117,118].

The heat treatment is part of the barrel making process, being performed by the French technique using fire, or by the American technique using heated steam for bending the staves followed by fire [161]. In European cooperage, the barrel is heated over a fire of wood shavings with various techniques of spraying or swabbing with water to enable the bending of the staves to the concave shape of a barrel without breaking—the bending phase [110,161]. Then, the barrel is placed again over the fire to heat the inner surface and to cause significant toasting in order to modify the structure [114], the physical properties [115], and the chemical composition of the wood [109,110,121], which confer a distinct character to the wine or distillate aged in it—the toasting phase. Despite the diversity of toasting protocols, the toasting level is usually classified as light, medium or heavy. In practice, the result mostly depends on the binomial temperature/time applied to each wood botanical species [109,110,143,162,163].

### 4.1. Phenolic Composition

Scientific data about the influence of the wood toasting level on the phenolic composition and related properties of the aged wine spirit are rather scarce. Notwithstanding, older works made on Spanish brandies by Artajona et al. [96], on Cognacs by Cantagrel et al. [164] and Viriot et al. [101], and on French wine spirits by Rabier and Moutounet [97] and Puech et al. [165] are noteworthy. Artajona et al. [96] found increasing contents of phenolic aldehydes in brandies with an increasing of barrel toasting intensity: ca 18 mg/L, 30 mg/L, and 58 mg/L under the influence of light, medium and heavy toasting, respectively. Rabier and Moutounet [97] observed increasing contents of ellagic acid (ca 15 mg/L and 60 mg/L), gallic acid (ca 4 mg/L and 9 mg/L), and vanillin (ca 0.5 mg/L and 1 mg/L) in a wine spirit aged over a two-year period in new oak barrels with light and heavy toasting levels. Puech et al. [165] also studied the influence of the toasting level (light, medium and heavy) in a wine spirit aged over a two-year period in new barrels of Limousin oak. They found an increasing content of vanillin with the rise of toasting intensity, which remained below 5 mg/L; a similar behavior was observed for syringaldehyde with ca.1 mg/L, 7 mg/L and 11 mg/L under the effect of light, medium and heavy toasting, respectively; for coniferaldehyde and sinapaldehyde, a sharp increase between light and medium toasting (from ca 3 to ca 13 mg/L, and from ca 2 to ca 22 mg/L, respectively) and a slight decrease under heavy toasting (ca 11 mg/L and 21 mg/L, respectively) were described.

In recent years, a comprehensive investigation was performed [6,56,120]. In that study, the same wine distillate from *Lourinhã* region (produced by Adega Cooperativa da Lourinhã) was aged over a four-year period in 250 L barrels. The barrels were made by J. M. Gonçalves cooperage (Palaçoulo, Portugal) using the following kinds of wood: Limousin oak (*Q. robur* L.) and Allier oak (*Q. sessiliflora* Salisb.) from French forests; American oak (mixture of *Q. alba* L./*Q. Stellata* Wangenh. and *Q. lyrata* Walt./*Q. bicolor* Willd.) from Pennsylvania/USA; Portuguese oak (*Q. pyrenaica* Willd.) and chestnut (*C. sativa* Mill.) from the North of Portugal. These barrels were divided into three groups. Then, each group were submitted to one of the three levels of toasting—light (LT), medium (MT) and heavy (HT)—according to the cooperage protocol: 10 min for light toasting, 20 min for medium toasting and 25 min for heavy toasting [120]. They were filled with the same wine distillate and kept in the cellar of Adega Cooperativa da Lourinhã in similar environmental conditions.

Analysing the low molecular weight phenolic compounds of the wine spirits aged in them, Canas [56] showed that the toasting level had a significant effect on the concentration of all phenolic compounds, except for scopoletin (Table 5), confirming the results of previous studies [96,97,164,165].

**Table 5.** Mean concentrations of low molecular weight compounds in wine spirits aged four years in barrels with different toasting levels.

| Compound | Light Toasting | Medium Toasting | Heavy Tosating |
|---|---|---|---|
| Ellagic acid | 38.85 [a] | 57.69 [b] | 87.36 [c] |
| Gallic acid | 42.27 [a] | 55.44 [b] | 54.22 [b] |
| Vanillic acid | 2.05 [a] | 3.20 [b] | 4.48 [c] |
| Syringic acid | 5.08 [a] | 6.00 [b] | 8.64 [c] |
| Ferulic acid | 4.30 [a] | 4.49 [a] | 5.00 [b] |
| Vanillin | 2.94 [a] | 6.43 [b] | 10.07 [c] |
| Syringaldehyde | 4.31 [a] | 13.00 [b] | 26.28 [c] |
| Coniferaldehyde | 3.25 [a] | 8.64 [b] | 13.55 [c] |
| Sinapaldehyde | 3.99 [a] | 14.49 [b] | 30.60 [c] |
| Umbelliferone | 0.47 [a] | 0.92 [b] | 1.64 [c] |
| Scopoletin | 37.36 | 39.42 | 36.58 |
| ΣLMW | 117.82 [a] | 197.10 [b] | 295.08 [c] |

Concentration in mg/L absolute ethanol except for coumarins, which are in µg/L absolute ethanol; mean values ($n$ = 56) followed by different letters ([a], [b], [c]) in a row are significantly different ($p < 0.05$); ΣLMW—Sum of low molecular weight phenolic compounds concentrations. Adapted from [56].

Furthermore, Viriot et al. [101] and Canas [56] emphasized a positive relationship between ellagic acid concentration in the wine spirits and the toasting intensity of the barrel. A different pattern is identified for gallic acid; its concentration in the aged wine spirits increases under the influence of medium toasting and slightly decreases under heavy toasting. Recent results obtained for grape marc spirit corroborate it [149]. This pattern expresses the behavior of gallic acid in the toasted wood, in which it undergoes degradation from the medium toasting as a consequence of higher thermal sensitivity [97,166]. In contrast, higher level of ellagic acid is ascribed to its high fusion point and greater accumulation in the toasted wood, as observed by Rabier and Moutounet [97]. As in the untoasted wood [108] and toasted wood [109,137], ellagic acid and gallic acid still remain the major phenolic acids of the aged wine spirits, being mainly derived from the wood ellagitannins and gallotannins [118,129,130,167].

It was also demonstrated that the rise of toasting level of the barrel promoted an increase of vanillic acid, syringic acid, ferulic acid and phenolic aldehydes contents in the aged wine spirits, as in the aforementioned studies, except for coniferaldehyde and sinapaldehyde [165]. It is well-known these compounds resulted from the wood lignin's decomposition [117] (Figure 5). Under mild temperatures, decarboxylation and cleavage of the aryl-alkyl ether bonds of the terminal units of this biopolymer take place, originating the cinnamic aldehydes (coniferaldehyde and sinapaldehyde). At higher temperatures, an oxidative cleavage of double C-C bond of the aliphatic chain of these aldehydes may occur, yielding the corresponding benzoic aldehydes (vanillin and syringaldehyde). The resulting concentrations express the balance between synthesis and degradation reactions. Therefore, the slight decrease of coniferaldehyde and sinapaldehyde contents reported by Puech et al. [165] for heavy toasting should have resulted from specificity of the toasting protocol, which induced higher degradation of these aldehydes.As the temperature rises, the phenolic aldehydes thus formed give rise, by decarboxylation, to the corresponding phenolic acids. Hence, they accumulate in the toasted wood [109,130,137,163].

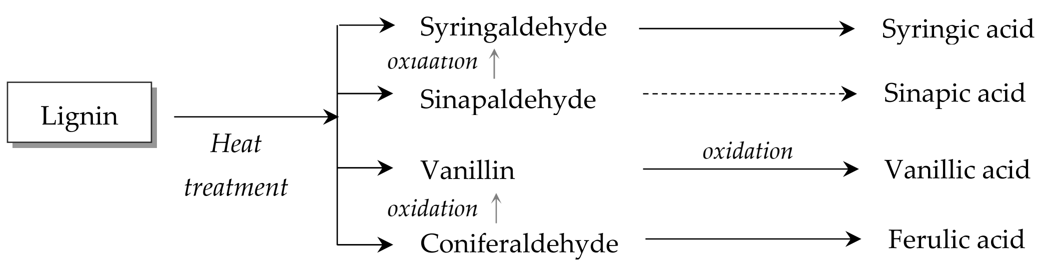



**Figure 5.** Mechanism of lignin's decomposition and formation of derived compounds; proposed by [117].

Furthermore, higher permeability of the wood and better access of the wine spirit to wood extraction sites caused by fragmentation of cell structures and reorganization of lignocellulose network [114,115] may also facilitate their release into the wine spirit. Likewise, lignin'shydrolysis during the ageing period may contribute to their increase in the beverage [14,29,102,116,144]. The presence of oxygen and the mild acidity of the medium, mainly modulated by the increase of acetic acid content over time, favor this pathway [37].

Regardless the toasting level of the barrel and the ageing time, it has been found [29,56,102] that syringyl-type aldehydes (sinapaldehyde and syringaldehyde) prevailed over those of guaiacyl-type (vanillin and coniferaldehyde) in the aged wine spirits. On the other hand, an increase in the syringyl/guaiacyl ratio with the toasting intensity has been referred [14,56,97]. In the above-mentioned work [56], mean values of 1.34, 1.82 and 2.41 were obtained for the same wine spirit aged during four years in barrels with light, medium and heavy toasting levels, respectively. This suggests that higher thermal stability of the syringyl compounds and subsequent higher availability in the toasted wood [117,118] was the causal effect.

There is also evidence of the increase of umbelliferone content in the wine spirit with the toasting level of the barrel [56], but the chemical mechanisms underpinning its formation/degradation during the heat treatment of the wood are still unknown.

Concerning the hydrolysable tannins, no significant differences in wine spirits aged in barrels with different toasting level were reported [122].

### *4.2. Chromatic Characteristics and Sensory Properties*

As for the wood botanical species, data from the literature [23] show the modulating effect exerted by the toasting level of the barrel on the chromatic characteristics of this beverage (Figure 6). The higher the toasting level of the barrel the more the evolution of the aged wine spirit color (higher intensity, saturation and red and yellow hues) with significant increments between levels. Acquisition of these chromatic characteristics makes the wine spirit aged in heavy toasting barrels look older than those aged in medium and light toasting barrels (as noticed for the wine spirit aged in chestnut wood when compared with those aged in different kinds of oak wood). These outcomes are correlated with the phenolic compounds extracted from the wood (Table 5) and are in agreement with those obtained for wine aged in barrels with different toasting levels [167]. In addition, the oxidative phenomena underlying the ageing process may also be responsible for the color acquired by the aged wine spirit; the higher the toasting level the higher the wood permeability to oxygen [114,115], and therefore greater extension of oxidation reactions are expected.

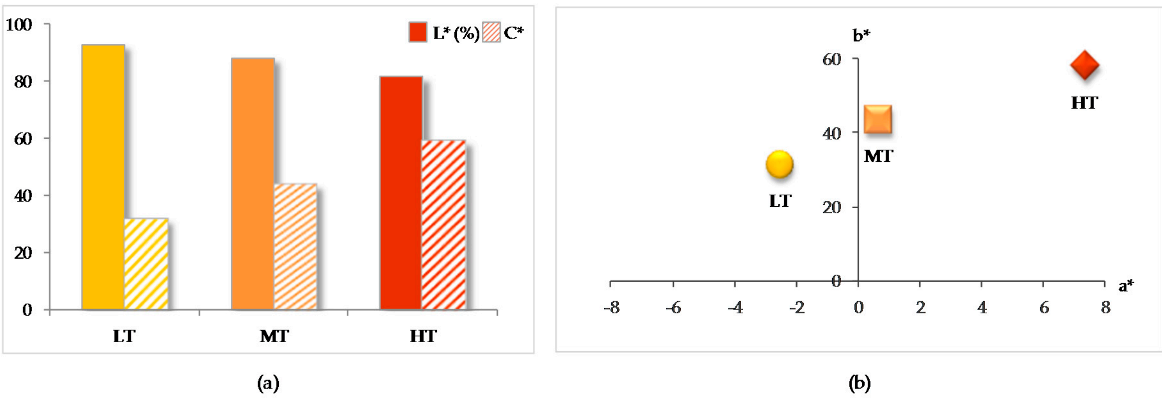



**Figure 6.** Mean values ($n$ = 56) of chromatic characteristics of the wine spirits aged in barrels with different toasting levels: (**a**) lightness (L*) and saturation (C*); (**b**) chromaticity coordinates (a*, b*); LT—light toasting; MT—medium toasting; HT–heavy toasting. For each chromatic characteristic, the differences between the aged wine spirits are very significant ($p < 0.01$). Adapted from [23].

From the sensory point of view, Canas et al. [23] indicated the predominance of yellow straw and yellow green in the wine spirits aged in light toasting barrels, and the prevalence of golden and topaz in those aged in medium and heavy toasting barrels, respectively (Figure 7). These results are consistent with those obtained by the CIELab method (Figure 6), showing a faster ageing of the wine spirit associated with the heavy and medium toasting levels.

Other sensory properties related to the phenolic composition, such as the vanilla aroma and astringency, had higher intensities associated with the heavy toasting barrels [153]. The wine spirits aged in light toasting barrels and medium toasting barrels revealed opposite and intermediate intensities of these attributes, respectively. Increasing concentrations of vanillin with the toasting intensity (Table 5) should explain the effect on the vanilla aroma. Regarding astringency, the existing information does not allow the establishment of a reliable relationship with the phenolic composition.

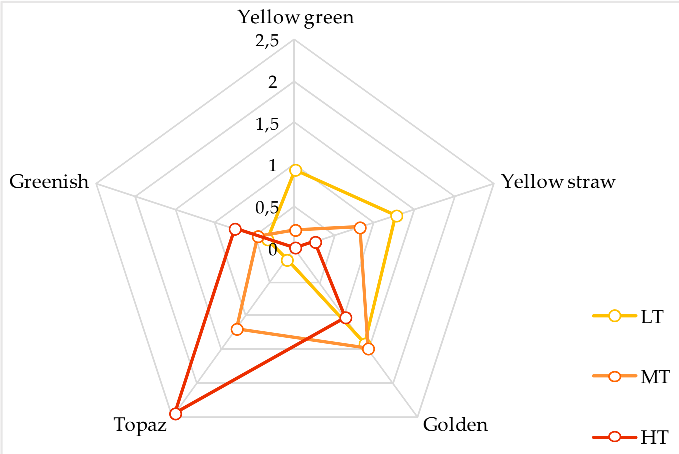



**Figure 7.** Color profile based on the average panel score's attributes of the wine spirits aged during four years in barrels with different toasting levels (LT—light toasting; MT—medium toasting; HT—heavy toasting). For each color attribute, the differences between the aged wine spirits are very significant ($p < 0.01$). Adapted from [23].

### 4.3. Antioxidant Activity

Only one approach [122] is found for the influence of the toasting level on the antioxidant activity of the aged wine spirit. Surprisingly, in this work, it was observed that a non-significant variation of the DPPH inhibition with the toasting intensity existed (60.6%, LT; 69.6%, MT; 63.5%, ST) (Figure 8). Such an effect was assigned to the high variability associated with the toasting operation [102,164], despite the significant influence found for the majority of low molecular weight phenolic compounds (Table 5). Nevertheless, Híc et al. [168] noticed a similar behavior for the oak wood antioxidant activity under the toasting effect.

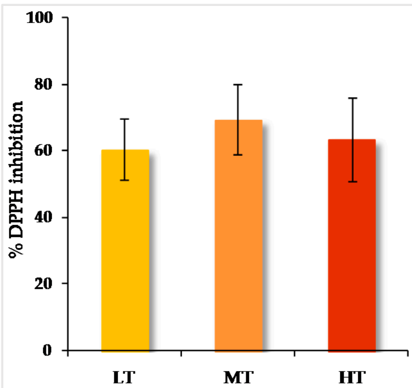



**Figure 8.** Mean values of antioxidant activity of the wine spirits aged in barrels with different toasting levels (LT—light toasting; MT—medium toasting; HT—heavy toasting). Adapted from [122].

## 5. Concluding Remarks

The reviewed literature demonstrates that the phenolic composition and some related properties of the aged wine spirit are effectively modulated by the kind of wood and the toasting level of the barrel. It is worth mentioning the highest enrichment of the wine spirit in low molecular weight phenolic compounds through the contact with chestnut wood (*Castanea sativa*). As a consequence, greater evolution of the chromatic characteristics and sensory color, as well as higher intensities of vanilla aroma, and higher antioxidant activity are achieved. *Q. pyrenaica* and *Quercus robur* exerts a similar influence. It means that these botanical species contribute to accelerate the ageing process and to give singular physicochemical characteristics and sensory profile to the aged wine spirit. The other kinds of oak (*Q. petraea* and *Q. alba*) show a weak performance, providing lower contents of phenolic compounds and promoting less intense related properties.

Regarding the toasting levels commonly used, higher concentrations of low molecular weight phenolic compounds, more evolved chromatic characteristics, sensory color, and other related sensory attributes are induced by the heavy toasting, followed by the medium toasting.

Thus, the wood botanical species together with the toasting level of the barrel are important resources for the industry for more sustainable management of the ageing process, to differentiate and to improve the quality of aged spirits. In addition, knowledge on the antioxidant activity of this beverage resulting from different ageing conditions may support a proper management of the ageing technology in order to add value to the final product. To be successful, the chemistry underlying the ageing process must be better understood. For this purpose, further research, supported by more advanced analytical methodologies, is needed on for key aspects such as: (1) identification and quantification of other phenolic compounds, coumarins and tannins of the aged wine spirit; (2) chemical reactions in which they are involved, and the relationship with chromatic characteristics and sensory properties of the aged wine spirit; (3) bioactive properties of the aged wine spirit modulated by thebarrel characteristics and other ageing factors. More studies about the heat treatment effect on the wood constituents and derived phenolic compounds, namely the coumarins, will also be of great relevance for a comprehensive insight into the ageing process.

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**Conflicts of Interest:** The author declares no conflicts of interest.

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