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Impact of Different Wood Types on the Chemical Composition and Sensory Profile of Aged Tsipouro: A Comparative Study

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Abstract: The production of high-quality aged marc spirits includes a minimum period of six months of ageing in oak barrels. Lastly, producers are interested in alternative botanical origin wood. The present study is aimed at investigating the influence of Greek oak (*Quercus trojana*) and Greek chestnut (*Castanea sativa*) compared with French (*Quercus petraia*) and American oak (*Quercus alba*) on the chemical composition and sensorial characteristics of aged tsipouro, produced from marc from the Black Muscat variety. Gas chromatography–olfactometry–mass spectrometry (GC–O–MS) was used to identify volatile compounds of the aged tsipouro. Also, colour and polyphenol measurements were made, and an organoleptic evaluation was performed by 16 trained tasters. The findings revealed rather similar results between the tsipouro made from different wood species, especially between Greek and French oaks, and relative differentiation for that of chestnut. All the aged distillates exhibited a pleasant and rich aromatic potential, dominated by floral and fruity terpene varietal aromas as well as wood-related volatiles. Chestnut, with a high phenolic potential, gives pleasant organoleptic effects over time and can be an alternative wood for ageing spirits. This research highlights the importance of wood selection in the tsipouro ageing process and enables the use of Greek wood species in the ageing of spirits.

Keywords: aged tsipouro; oak wood; chestnut; American; French; Greek oak; GC–O–MS analysis; sensory analysis

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1. Introduction

Greek tradition encompasses the production and consumption of tsipouro, a traditional Greek spirit drink (named tsikoudia on the islands of Crete and Cyclades). Despite its long history, it is only over the past few decades that tsipouro has been bottled for market circulation and received geographical indication status from the European Union [1–4].

It is classified as a grape marc spirit, similar to other grape marc spirits, such as grappa in Italy, marc in France, orujo de Galicia in Spain, aguardiente bagaceiras in Portugal, and zivania in Cyprus, among others [2,4]. As a grape marc spirit, tsipouro is produced solely from fermented grape marc, with the option of adding lees not exceeding 25 kg of lees per 100 kg of grape marc used. The first distillation is carried out in the presence of the marc itself, and the minimum alcoholic strength by volume for grape marc spirit is set at 37.5% vol [1].

Tsipouro production is small, about 1.9 million litres of anhydrous alcohol (L.A.A.), which makes up to 8.8% of the Greek production of alcoholic beverages and with minimal participation in exports (0.10 mil. L.A.A., 0.7%). Nevertheless, it illustrates an upward trend and is popular in domestic consumption (about 26.4% of Greek spirit drinks), interwoven with the Greek culinary culture to complement both traditional and contemporary local cuisines [5].

Moreover, it has traditionally been produced and consumed as a clear, colourless, and unaged distillate product. However, recently, the maturation of tsipouro in barrels has gained popularity and has given rise to a distinct category of Greek spirits known as "aged tsipouro". Ageing is recommended for at least six months in oak barrels [4,6], aiming to achieve a high-quality spirit with a dark yellow-amber colour, a unique aromatic balance, and a harmonious composition that showcases the interplay between the fruity and floral character of the varietal aromas and the exceptional wood-derived aromas imparted by the barrel contact. For the barrel ageing of tsipouro, as for many well-known international aged spirits, American oak (*Quercus alba*), French oak (*Quercus petraia* and *Quercus robur*), and oak barrels from other European countries have mainly been used, depending on the desired aromatic profile of the produced distillate. Over the last few years, many producers of wines and spirits have tried other species of oak [7,8] or other types of wood [9] as an alternative for or as a revival of a traditional way of ageing. Where the law allows it, other woods, like chestnut (*Castanea sativa*) or even cherry, acacia, and ash, have been used [7,9–12].

In the international literature, a substantial amount of research related to the characteristics and volatile compounds of oak [13–16] and chestnut has been found [10,13], as well as to the ageing of spirits, such as grappa [15], orujo [17], wine spirits [18], brandies [18–22], cognac [23], and fruit spirits [24,25], which are all aged by the effect of oak or chestnut woods.

Greek forests, mainly in the northern part of the mainland, are covered with oak trees, providing an important financial contribution to local economies (Figure S1). The genus *Quercus* is represented by 11 species and a total of 14 taxa, as documented and classified in *Flora Hellenica* [26] and "Vascular Plants of Greece" [27]. Notable among these are *Q. petraia* and, to a lesser extent, *Q. robur*. While research on the suitability of Greek oak species for the cooperage of barrels and casks for wines and spirits is in its nascent stages, the endemic species *Q. trojana* subsp. *trojana*, *Q. petraea* subsp. *petraea*, and *Q. frainetto* have emerged as promising candidates for exploration and exploitation owing to positive indications regarding their properties and substantial geographical distribution [27,28]. Within the Greek territory, *Q. trojana* is categorised into two subspecies: *Q. trojana* subsp. *trojana* (or *Q. macedonica*) and *Q. trojana* subsp. *euboica* (endemic to the island of Evia, with limited distribution). The Macedonian oak (*Q. trojana* subsp. *trojana*), found primarily in Northwest Greece (mainly at the Pindos Mountain range in the region of Macedonia), is a focal species where a unique tree has been declared a "Preserved Monument of Nature" [29,30].

Chestnut, commonly referred to as "Castania" in Greek, holds significant historical and economic importance for Greece. Pollen analysis has indicated the presence of chestnut trees in Northern Greece in as early as the 10th century B.C. (Figure S1) [31]. The cultivation of chestnut, valued for both its fruit and wood, expanded into Europe and was initially introduced to the Italian Greek colonies [32] and subsequently disseminated throughout the entire Mediterranean coast [31].

Several studies have focused on the volatile profile of unaged tsipouro [33–36], and only a few have focused on the influence of wood contact with tsipouro [37,38]. To the best of our knowledge, there are no studies that have investigated the potential contribution of volatile components to the intricate aromatic profile of aged tsipouro. Hence, this study constitutes the first attempt to determine the influence of the well-known *Q. petraia* and *Q. alba* on the aromatic profile of aged tsipouro. It also marks the initial exploration of utilising the Greek endemic wood species *Q. trojana* and *C. sativa* for the ageing of tsipouro.

2. Materials and Methods

2.1. Tsipouro Production

A total of 1500 kg of grapes of the Mavro Moscato (Black Muscat) grapevine variety, cultivated organically at the foothills of Agrafa mountains (communities of Paliouri and Daphnospilia, Karditsa, Greece; 39°14′53.455″ N, 21°57′40.995″ E), were manually harvested at the optimum grape maturity level (21.5 °Brix) on 11 September 2014. Grapes were destemmed, crushed, and slightly pressed. The free-run juice and the pressed juice were separated from seeds and skins and remained grape juice (marc). Approximately 270 kg of marc was transferred to a stainless-steel temperature-controlled tank. The marc was fermented spontaneously and yielded 7.5 L per 100 kg of anhydrous ethyl alcohol.

2.2. Distillation

The marc was fermented spontaneously in a stainless-steel tank with a controlled temperature (24 °C) and without SO_2 additions, similar to red vinification. After the completion of alcoholic fermentation, the fermented marc was used to produce tsipouro by a double distillation process. A traditional simple distillation without rectification copper-made alembic of 130 L was used, composed of a gas heating system, a boiler, a still head, and a cooling system (Figure S2). In the first step, the grape marc was transferred to the still, and the produced distillate was separated at several fractions of "heads", "hearts", and "tails". The aim was to have a better separation of the fractions, especially for the "heads" and "tails," and to identify and remove, in real-time, defective odours, optimising the quality of the final product through a repetitive sensory (olfactory) evaluation. The selected "heart" fractions were joined all together and redistilled. Following the same procedure with the strict separation of "heads" and "tails", finally, a distillate of 70% v/v was collected in a stainless-steel tank. After a period of 3 months of maturation, it was diluted with pure water produced by an ion-exchange resin column for water purification (Ionel S.A., Athens, Greece) down to 40% v/v, which is the usual ethanol content of commercial brands.

2.3. Woods

The wooden cubes used in this study were from four different botanical species: French oak (FO) (*Quercus petraia*), American oak (AO) (*Quercus alba*), Greek oak (GO) (*Quercus trojana* or *Querqus macedonica*), and Greek chestnut (GC) (*Castanea sativa*). The first two (French and American oak) were kindly donated by the Tonnellerie Nadalie (Bordeaux, France) in the form of untreated staves, and the other two, in the form of tree logs, from the Department of Forestry, Wood Sciences, and Design (DFWSD, Karditsa, University of Thessaly, Greece). The tree of Greek chestnut came from the Athos peninsula in Northeast Greece, from the monastic community of Mount Athos. The tree of Greek oak, which for the first time is being studied for its effect on ageing, came from the Kouri peri-urban forest at Kozani, West Macedonia (Greece). It was cut, with permission of the Forestry Office of Kozani, especially for this study, and a log of 2 m (from height 30–230 cm) and diameter of approx. 32 cm was used. The cutting of wood into small pieces of dimensions $2.0 \times 2.0 \times 1.0$ cm took place at the laboratories of DFWSD (Figure S3). The same protocol was followed for all the different types of wood.

2.4. Toasting

Various toasting levels were tested at the Technology and Quality Control of Wine and Spirits Laboratory (Department of Food Science and Nutrition, University of Thessaly, Karditsa, Greece). Keeping the woods of different toasting techniques in tsipouro for 3 months and after a sensory evaluation of the samples, a toasting technique was finally selected in 2 stages: the first was at 160 °C (for 6 h), and the second stage of already toasted wood was at 200 °C (for 8 h) in a drying and heating chamber (Binder GmbH, Tuttlingen, Germany). The toasted wood pieces of the four different types of wood were weighed before and after the toasting, and only the pieces with similar weights ($\pm$5% w/w) were used for the ageing process. The toasted wood pieces (Figure S3) were placed into 4 L glass containers that were first filled up with 4 L of tsipouro. The surface/volume ratio of a barrel was considered to be 2.01 m^2/225 L, according to a previously published research work [39]. The number of cubes added to the 4 L glass containers was based on that given information and the fact that the cubes had dimensions of $2.0 \times 2.0 \times 1.0$ cm and a total surface of 16 cm^2.

All conditions were treated in triplicates. The samples were maintained in a dark room with stable conditions (80% humidity and 18 °C) for 5 years. Also, tsipouro, without the addition of wood cubes, was kept in the same glass containers in triplicate.

2.5. Aged Tsipouro

2.5.1. Colour Measurements and Total Polyphenolic Index

Measurements of the chromatic characteristics were realised in the tsipouro aged samples. The CIELAB coordinates L* (lightness), a* (redness/greenness), b* (yellowness/blueness), C* (chroma), and h* (hue) were calculated according to Ayala et al. [40] with the application of the MSCV software [41]. The total colour difference (ΔEab*) between samples was calculated using the following formula:

$$\Delta Eab^* = ((\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2)^{1/2}$$

The total phenolic index (TPI) was determined by measuring the absorbance at 280 nm [42]. Analyses were realised with a double-beam UV–vis spectrophotometer (Shimadzu UV-1900, Shimadzu Scientific Instruments, Kyoto, Japan).

2.5.2. Extraction of Volatile Compounds

The extraction of volatile components was performed using liquid–liquid extraction with dichloromethane as the solvent, adapting a protocol for the wine sample analysis [43]. A total of 100 mL of the sample was diluted with 200 mL of ultrapure water in a 500 mL Erlenmeyer flask. A total of 20 mL of dichloromethane was added, and the mixture was stirred vigorously for 15 min. The organic phase, with the use of a separation funnel, was collected in a glass centrifuge tube. The process was repeated twice with the addition of 10 mL of dichloromethane each time. The separated organic extracts were combined and centrifuged at 4000 rpm for 10 min at 4 °C. After phase separation, the lower organic phase layer was collected, and anhydrous sodium sulphate was added to achieve the sample's dehydration, followed by decantation into another vial with salt removal. The extract was concentrated to 500 µL under a nitrogen flow and transferred to a 1.5 mL vial. Sample storage was maintained at −20 °C prior to concentration when the extract was in the vial containing salt. The extraction was performed in duplicate for each sample.

2.5.3. Gas Chromatography–Olfactometry–Mass Spectrometry (GC–O–MS) Analysis

Analysis with gas chromatography–olfactometry was performed according to Goulioti et al. [44]. Briefly, a Perkin Elmer olfactory detector port (SNFRTM Olfactory Port, N6590100) was fitted to the GC-MS system (Perkin Elmer Clarus 590 gas chromatograph coupled to a Perkin Elmer Clarus SQ8S mass spectrometer—Waltham, MA, USA), connected by a flow splitter to the column exit (SwaferTM technology, Perkin Elmer) to achieve a 1:5 split ratio. GC effluent was combined with humidified air (99.999% purity, Revival, Greece) at the bottom of the detector port, and 1 µL of the sample was injected in splitless mode (injector temperature, 250 °C), and the split vent was opened after 5 min. A DB-Wax column (50 m × 0.25 mm i.d., 0.25 µm film thickness Agilent J&W GC Column, Folsom, CA, USA) was used with helium as the carrier gas in a constant flow rate of 1.9 mL/min. The temperature program was as described by Goulioti et al. [44]: 40 °C for 2 min, then increasing by 4 °C/min to 240 °C, with a 20 min isothermal phase at 240 °C. The MS transfer line was set at 250 °C, and the electron ionisation mass spectra at 70 eV were recorded in the range of m/z 40–400.

For the GC–O–MS analysis, three panellists (males, 35–55 years old) trained in descriptive sensory analysis and who were able to identify the odours of different aged tsipouro samples were used to identify the odours by the olfactory detector port. The panellists noted each odour-active compound detected in their retention time, as well as their odour description, and finally, their intensity on a scale of 1 to 3. The absolute average intensity value from the three panellists was used for the comparison of the different samples. The collected MS data were analysed with the TurboMass Ver6.1.2 GC/MS Software (Perkin

Elmer, Waltham, MA, USA) and compared with the NIST 2008 Mass Spectra library (US National Institute of Standards and Technology, Gaithersburg, MD, USA). The Kovats retention index (RI) of the target compound was calculated using C7–C30 alkanes (Sigma-Aldrich, St Louis, MO, USA) and compared with the NIST database [44]. Subsequently, a comparison to the literature and the Flavornet website for all aroma descriptors was performed [45].

2.5.4. Sensory Analysis

The produced tsipouro grape marc spirits were used for the sensorial evaluation by 16 trained panellists (8 females and 8 males; 23–55 years of age, all experts on spirits and mainly marc distillates) who provided their informed consent to take part in the study. The panellists were trained in two training sessions, and following this process, they evaluated the samples. During the training sessions, the panellists were asked to smell the reference odorants to become familiarised with the odours that can be found in aged marc spirits produced by Muscat grapes. The reference odorants were prepared with the use of natural products prepared in a 40% (v/v) ethanol solution made of neutral alcohol of agricultural origin (Table 1). The reference odorants were prepared and macerated overnight before the training session. The evaluation of the marc spirits was realised in a temperature-controlled testing room (18–20 °C) in individual booths with natural lighting, and thereupon the samples were left to reach room temperature. Each sample for a given marc spirit was prepared by blending in equal proportions of the respective replicates. Hence, 30 mL of each sample was poured into black INOA/ISO tasting glasses covered with plastic Petri dishes and coded with a three-digit monadic sequence according to a Latin square design. The panellists evaluated each sample ortho-nasally and rated them for each sensory attribute intensity according to a five-point scale (ranging from 1, not perceived, to 5, very strong). During the tasting, the panellists had a 1 min break. All sensory attributes were subjected to statistical analysis with the application of one-way ANOVA ($p < 0.05$) followed by Tukey's HSD post hoc test ($a = 0.05$).

Table 1. Odour descriptors based on reference standards prepared in 100 mL of 40% (v/v) ethanol solution.

Odour Category (Descriptor)	Substance/Flavouring	Quantity
Floral (rose)	Rose water (flavoured water by steeping rose petals)	1 mL
Citrus (bergamot)	Bergamot peel	2.5 g
Dry fruits (apricot, plum, raisins, fig)	Dried apricots Dried plums Raisins Fig spoon sweet	2.5 g 15 g 12 g 20 g
Vanilla	Vanilla extract, 1 g/L	1 mg
Honey	Honey	15 g
Caramel	Caramel extract	15 µL
Wood	Uncharred oak chips	5 g
Spicy (cinnamon, allspice, pepper, clove)	Ground cinnamon Ground allspice Ground pepper Ground clove	0.21 g 0.25 g 0.3 g 0.5 g
Smoke (tobacco)	Hand rolling tobacco	0.8 g
Roast (coffee, hazelnut)	Coffee Hazelnut	2 g 2 g
Chocolate	Black chocolate	5.4 g
Coconut	Ground coconut	2 g

3. Results and Discussion

3.1. Identification of Volatile Compounds by GC–O–MS of Aged Tsipouro

Gas chromatography–olfactometry–mass spectrometry (GC–O–MS) was applied to identify the volatile compounds of aged tsipouro with and without wood contact and their influence on the organoleptic characteristics of the analysed samples. While not all volatile compounds contribute to the distinct aroma of a particular spirit drink, it is essential to identify those that genuinely have an olfactory impact on this specific spirit. In agreement with this statement, the current survey revealed that 29 odour-active compounds were determined by GC–O analysis. The vast majority of the identified compounds were confirmed through a combination of retention index, mass spectrum, and reference compounds. The odorants contributing to the overall aroma of the samples, as revealed by GC–O, belong to the chemical compound groups of esters, higher alcohols, terpenoids, lactones, and furan derivatives. Among these compounds, alcohols, esters, and terpenes were found in quite high intensity and were further responsible for the fruity and floral aromas in all samples, namely 2-methyl-1-propanol, 2-methyl-1-butanol, ethyl hexanoate, ethyl octanoate, 2-phenylethyl acetate, linalool, and geraniol, as listed in Table 2. Those compounds are acknowledged as the key aromatic compounds present in grapes (*V. vinifera*), overripe pomace, and wines produced from Muscat cultivars [46–48]. The family of Muscat grape varieties has significant amounts of terpenoids, which exceed the odour threshold. These amounts will, therefore, also pass into the distillate, which is why linalool and geraniol were determined by using GC–O in high intensity, followed by citronellol, nerol and *a*-terpineol (Table 2). Linalool scored the highest intensity perception by the GC–O, and subsequently, this was found for the control sample, in comparison to the aged samples, followed by the geraniol that had similar odour intensity, with and without wood contact. Citronellol, nerol, and *a*-terpineol displayed a low intensity for the control sample in addition to the aged with wood samples. Other marc spirits, such as grappa [49–53], orujo [49,54], bagaceiras [55], zivania [56], as well as wine distillates, such as wine spirit [57], brandy [58–61], cognac [60,62], and pisco [63,64], have similar chemical compositions with tsipouro.

Regarding the higher alcohols, such as 2-methyl-1-propanol (solvent and fruity odorants) and 2-methyl-1-butanol (cheesy, green, and solvent), a high intensity for all the samples was found. It is worth noting that 1-hexanol is not an alcoholic fermentation product, and its presence is related to the grape variety and maturation stage. When it is detected in low concentrations, it positively influences the aroma of the distillate. Otherwise, it would provide an intense cut-grass aroma. Additionally, it serves as an indicator of the pressing degree when sensing the green-like notes [65–67]. In this case, 1-hexanol was detected in low-aroma intensity levels, suggesting that the grapes were not subjected to high pressure during the pressing process. Furthermore, in spirits, it is commonly recognised that 2-phenylethanol contributes to the aroma of rose [68]. However, the findings of the current study indicate that another compound, specifically 2-phenylethyl acetate, exhibited a more pronounced presence and played a greater role in generating the rose aroma. It makes sense, as the boiling point of the alcohol is higher than that of the acetate.

Moreover, ethyl esters were also detected in the studied samples, as already found in previous studies on brandy, cognac, apple, and apricot distillates [67–69]. The presence of ethyl hexanoate, ethyl octanoate, and ethyl decanoate, which contributes to the fruity character of the distillates, was detected in all the samples. Based on the findings of this study, ethyl hexanoate had a higher odour intensity, and when the alkyl group was increased, the active aroma compound intensity was decreased, which is in line with the research of de-la-Fuente-Blanco et al. [70].

From the above, it is clear that tsipouro, without contact with the wood after ageing for five years, still exhibited predominant fruity, floral, and citrus notes, which were influenced by the grape variety used for its production. On the other hand, the aged tsipouro produced after a period of wood contact displayed more prominent aromas of caramel, smoked, and vanilla. This was expected, as the four different types of wooden cubes, namely Greek oak (GO), American oak (AO), French oak (FO), and Greek chestnut (GC), play a significant role in the flavour and aroma profiles of the aged tsipouro. In addition, as stated by Tarko et al. [71], throughout the maturation process of alcoholic beverages in the presence of carefully chosen wood species, different chemical reactions take place between the beverage constituents and the extracted compounds from the wood. These interactions significantly influence the aromatic complexity of alcoholic drinks.

Table 2. Aroma description, compound identification, and aroma intensity, determined by GC–O–MS for the different tsipouro aged samples (French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC)).

Num [a]	**RI GC-MS** [b] **Column**		**Aroma Description** [c]	**Compound** [d]	**Intensity** [e]				
	DB-WAX	**DB-5**			**CONTROL**	**FO**	**AO**	**GO**	**GC**
1	1104	655	Solvent, fruity	2-methyl- 1-propanol (isobutanol)	3	3	3	3	3
2	1210	756	Cheesy, green, solvent	2-methyl-1-butanol	3	3	3	3	3
3	1210	756		3-methyl-1-butanol (isoamyl alcohol)	n.d. [1]	n.d.	n.d.	n.d.	n.d.
4	1240	1007	Fruity	ethyl hexanoate	3	3	3	3	3
5	1251	1012	Caramelised, heavy, sweet	unknown	1	1	1	1	1
6	1356	865	Cut-grass	1-hexanol	1	1	1	1	1
7	1391	858	Cut-grass	(*Z*)-3-hexen-1-ol					
8	1433	1193	Fruity	ethyl octanoate	2.3	2	2	2	2
9	1457		Floral, rose	unknown	1	1	1	1	1
10	1533	1104	Citrus, bergamot	linalool	3	2	2	2	1.7
11			Sweet, tropical	unknown	1.3	1	1	1	1
12	1640	1401	Brandy, fruity	ethyl decanoate	1	1	1	1	1
13			Citrus, sweet orange	unknown	1.3	1	1	1	1
14	1700	1192	Floral, lilac	*a*-terpineol	1	1	1	1	1
15	1768	1235	Lemon, citronellal, lemongrass	citronellol	1.3	1	1	1	1
16	1792	1230	Honeysuckle, lavender	nerol	1	1	1	1	1
17	1837	1250	Rosy	2-phenylethyl acetate	2.3	2	2	2	1.7
18	1852	1088	Smokey	guaiacol	n.d.	2.3	2	2.3	1
19	1857	1103	Coconut-like	(3S,4R)-*trans*-whiskylactone	n.d.	2	2	2	n.d.
20	1862	1266	Citrus	geraniol	2.3	2	2	2	2
21	1914	1115	Rose/honey	2-phenylethanol	1.3	1	1	1	1
22	1971	1310	Coconut-like	(3*S*,4*S*)-*cis*-whiskylactone	n.d.	1	3	1	n.d.
23	2033	1282	Roasty, spicy	4-ethyl-2-methoxyphenol (4-ethyl guaiacol)	n.d.	1.3	2	1.3	n.d.
24	2038	1077	Caramel-like	4-hydroxy-2,5-dimethyl-3(2*H*)-furanone (furaneol)	n.d.	1.7	2	1.7	1
25	2212	1314	Clove-like	2-methoxy-4-vinylphenol (4-vinylguaiacol)	n.d.	1.7	2	1.7	1
26	2235	1117	Seasoning-like	3-hydroxy-4,5-dimethyl-2(5*H*)-furanone (sotolon)	n.d.	2	2	2	2.3
27	2566	1405	Vanilla	vanillin	n.d.	3	1	2.7	1
28	2600	1525	Vanilla	methyl vanillate	n.d.	2	1.7	2	1
29	2665		Vanilla, chocolate	ethyl vanillate	n.d.	2.3	2	2.3	1

[a] Elution order from the DB-WAX column. [b] Retention index on DB-WAX and DB-5 columns. [c] Odour quality, as perceived at the sniffing port during GC–O. [d] Identified odorants based on retention indices on both columns and on mass spectra obtained by MS. [e] Intensity, as perceived from the panellists, from 1 = low to 3 = high intensity. n.d. [1]: not determined.

In line, the findings demonstrated that each type of wooden cube contributed unique flavours and aromas to the tsipouro, as presented in Table 2. In particular, the addition of different wood species led to the formation of specific substances in the tsipouro, such as guaiacol, (3*S*,4*R*)-*trans*-whiskylactone, (3*S*,4*S*)-*cis*-whiskylactone, 4-ethyl-2-methoxyphenol, 4-hydroxy-2,5-dimethyl-3(2*H*)-furanone, 2-methoxy-4-vinylphenol, 3-hydroxy-4,5-dimethyl-2(5*H*)-furanone, and several derivatives of vanillin. These substances were directly derived from the wood and were not present in the non-aged wood contact in the sample of tsipouro. From those compounds, guaiacol, associated with the smoky character of the samples, was detected at an average level of intensity for the three different oaked samples, while the chestnut was at lower levels, and is in accordance with the results of Culleré et al. [72]. The 4-ethyl guaiacol (4-ethyl-2-methoxyphenol), characteristic of the roasted and spicy odour, was found to be at a higher intensity in the AO samples, followed by the GO and the FO, while the presence of the specific odour-active compound could not be detected in the GC samples. From the two whiskylactones contributing to the coconut odour, the *cis* form was at a higher intensity level for the AO samples, followed by lower levels for the other two oaked samples, whereas for the chestnut sample, there was no possibility of detection. For the *trans* form, the levels of intensity were found to be at the middle level for all the oak samples and undetected for the GC samples. Culleré et al. [72] were able to identify by GC–O–MS the *cis*-whiskylactone in the extracts from chestnut wood samples, something that was not possible in the present study; in all probability, due to a different type of chestnut wood. The 4-hydroxy-2,5-dimethyl-3(2*H*)-furanone (furaneol), with the caramel-like odour, had higher levels in the AO samples, followed by the FO, GO and GC samples had the lower levels. Similarly, for the 2-methoxy-4-vinylphenol (4-vinylguaiacol), the powerful clove-like spicy odour had the lowest detected levels in the GC, followed by higher levels from the GO and FO and the highest identified in the AO samples. Those results followed the opposite trend that was observed in a previous study [72]. Moreover, the 3-hydroxy-4,5-dimethyl-2(5*H*)-furanone (sotolon), a potent aroma compound with a seasoning-like odour, had the same middle-intensity levels for all the samples. Finally, from the vanillin and its derivatives, vanillin was higher in the FO and the GO tsipouro aged samples. Both methyl and ethyl vanillate were identified at similar middle levels for all three oak samples, followed by lower detection levels in the chestnut-aged samples. Vanillin had the same trend as Culleré et al. [72]; however, there was a contrasting trend for the methyl vanillate. The difference presented between the studies can be related to the prolonged ageing period in the present study and the different botanical origins.

3.2. Colour Measurements and Total Polyphenolic Index of Aged Tsipouro

Several types of woods covering a range of different botanical origins (American, French, and Greek oak) and species (Greek oak and chestnut) were assessed for their ability to influence the organoleptic characteristics of the spirits aged in contact with wood fragments for five years. It is well known that alcoholic beverages aged with different types of wood had a considerable impact on their phenolic composition and colour characteristics [73–76]. As shown in Figure 1, the total polyphenolic index (TPI) of the spirits aged in contact with GC wood was observed to be almost double that of the TPI at the tsipouro marc spirit that was aged with the FO and GO oaks. The spirit aged with AO had the lowest TPI, with a statistically significant difference from the other spirits. Híc et al. [74] found a higher level of total phenolic index in the wine spirits aged with wood fragments of chestnut, which were also related to a higher antioxidant capacity. Similarly, Belchior et al. [77] mention that a higher phenolic content was observed in the spirits aged in barrels made from chestnut wood, followed by the European oak and, finally, the American oak wood barrels. Differences were detected by Canas et al. [18] in the concentrations of phenolic aldehydes, phenolic acids, and scopoletin, and mainly with the higher concentrated phenolic compounds, such as gallic acid and ellagic acid, which are directly extracted from the wood or hydrolysed from gallotannins and ellagitannins; this was due to the prolonged period of ageing [78].

Figures 2 and 3 illustrate the colour parameters of the spirits aged with different wood fragments. Figure 2 shows the chromatic coordinates a* and b*. The colour-opponent dimension a*, expressing the red-greenness of the studied spirits, shows a clear trend. The Greek chestnut wood influenced the a* coordinate more; the GC-aged spirit had the higher value (26.88 $\pm$ 1.3), followed by the Greek oak-aged spirit (9.61 $\pm$ 0.09), and the French oaked spirit (8.06 $\pm$ 0.07) being slightly lower but with a significant difference from the GO. The American oaked spirit had a lower value, with a high difference from the other three spirits (3.14 $\pm$ 0.05). A similar tendency appeared for the b* yellow-blueness coordinate. GC exhibited a higher yellow colour tint (87.0 $\pm$ 2.7) with a significant difference from the other three spirits. The GO and the FO had similar yellow colours (74.0 $\pm$ 3.1 and 71.8 $\pm$ 2.4, respectively), while the AO spirit had a lower yellow tint (59.0 $\pm$ 1.3) with statistically significant differences from the other spirits. Based on the a* and b* values, all aged spirits had yellow colour hues with a considerable proportion of red colour. Similar results were observed from other research groups when comparing the ageing of spirits that had been in contact with chestnut and oak woods [74,78,79]. Regarding the values related to the a* and b* coordinates, there are differences that are most probably related to the toasting process and mainly the toasted level in addition to the oxidation process [73,80]. In general, the tsipouro aged in contact with chestnut wood had more intense topaz hues than golden and yellow straw; the Greek and French oak had more golden hues, respectfully, followed by the American oak, which produced more yellow straw tints.

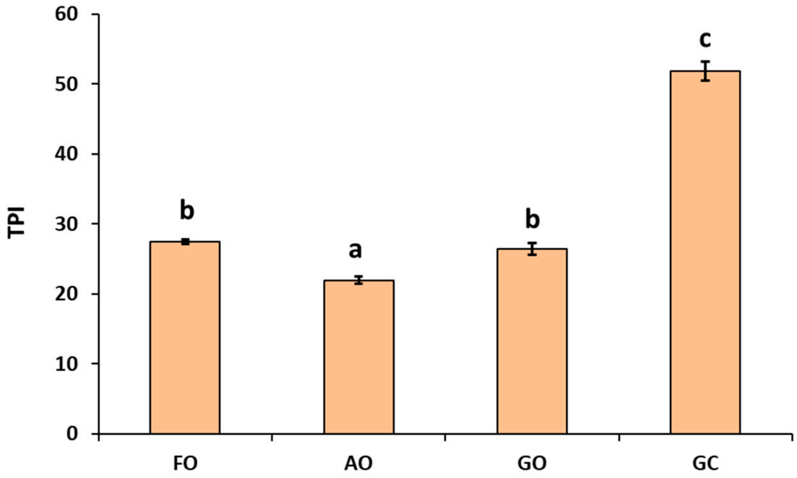

Figure 1. Total phenolic index (TPI) of tsipouro marc spirits aged for 5 years while in contact with French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC). All data are expressed as the average of 3 samples ± standard deviation. Different Latin letters indicate statistical differences ($p < 0.05$).

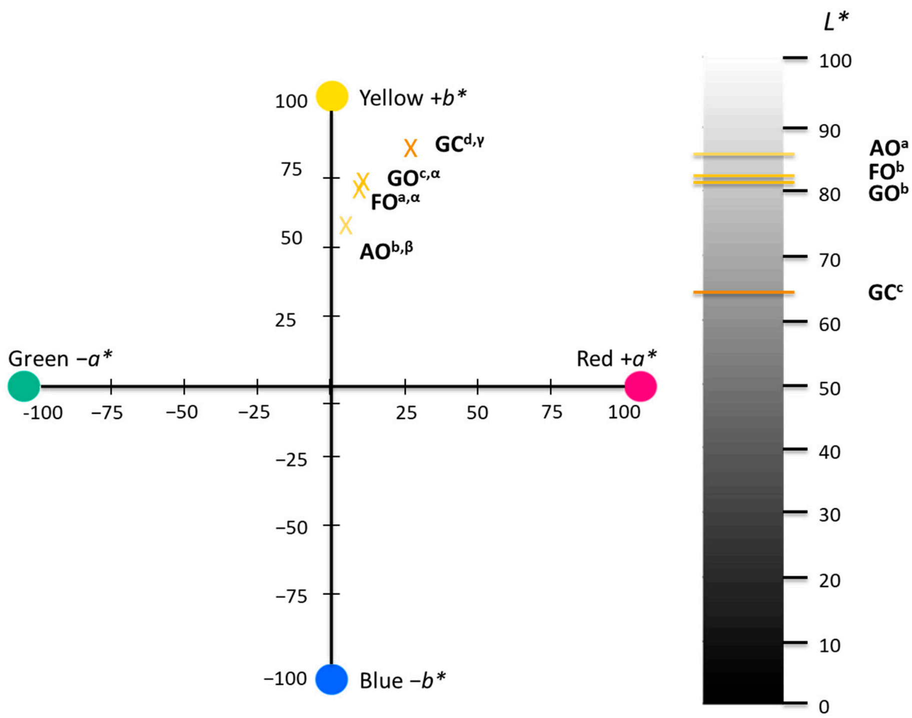

Figure 2. CIELAB chromatic coordinates: a*, b*, and lightness L* of tsipouro marc spirits aged for 5 years while in contact with French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC). All data are expressed as the average of 3 samples. Different Latin letters indicate statistical differences ($p < 0.05$) for the L* and a* parameters and Greek letters for the b* parameter.

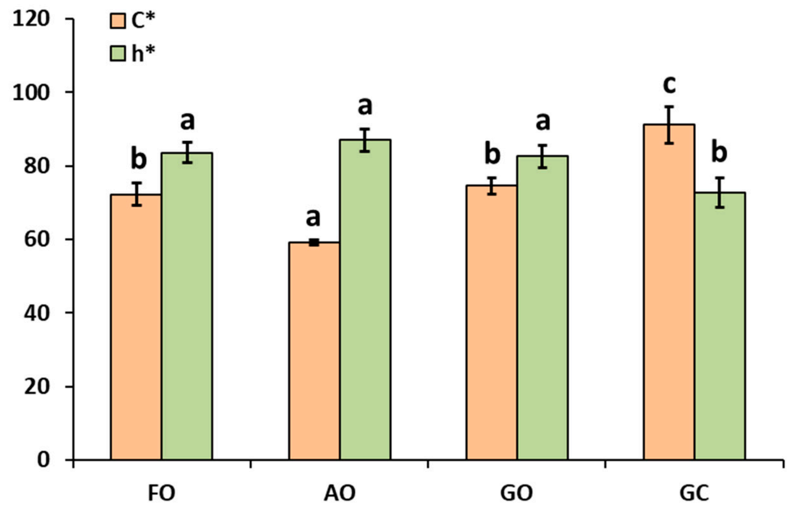

Figure 3. CIELAB chromatic coordinates: chroma C* and hue h* of tsipouro marc spirits aged for 5 years while in contact with French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC). All data are expressed as the average of 3 samples ± standard deviation. Different Latin letters indicate statistical differences ($p < 0.05$).

Related to the lightness L*, the visual perception of the luminosity in the CIELAB colour space has values from 0% (dark/opaque) to 100% (transparent/bright). From the tsipouro aged marc spirits, the chestnut samples showed a lower value, thus being the darkest/most opaque aged spirit (Figure 2). Higher values were presented in the French and Greek oak spirits, with the highest value observed in the American oak marc spirit, presenting the brightest colour. Comparable results were shown in other scientific works [74,78], with the values decreasing with ageing and increases in the toasting level and the level of oxidation [78,81,82]. On the other hand, chroma C* expresses the colourfulness of an area judged as a proportion of the brightness of a similarly illuminated area that appears white or highly transmitting [83], meaning that at a higher chroma value, the colour intensity perceived by the human eye is greater [82]. Lower values were found with the AO marc spirit, followed by the FO and the GO aged spirits (Figure 3). The tsipouro that was aged in contact with chestnut wood had a higher value with higher colour intensity, as was also indicated by Canas et al. [78]. Related to the hue h* parameter, the AO spirits had a slightly higher but no significant difference from the other two oak-aged spirits (Figure 3). The chestnut had a significantly lower value, indicating that it released more coloured compounds than the spirits aged with oak. In general, it could be concluded that the extraction of different amounts and compositions of phenolic compounds from the different wood fragments had a clear influence on the chromatic parameters of the aged spirits [74,78,79].

Table 3 shows the total colour differences (ΔEab*) among the tsipouro aged in contact with different woods. It is considered that when the ΔEab* values are ≤ 3 units, the human eye cannot perceive the visual difference in the colour between two samples [84]. In the case of the present study, the total colour differences between marc spirits were all higher than 3 units, indicating that the human eye could distinguish among the different samples. The ΔEab* was higher when the chestnut sample was compared with the three different oak samples. The smallest difference (almost at the limit of the human eye's capacity to distinguish them) was between French and Greek oak-aged marc spirits, which are in agreement with the results from the colour parameters presented above.

Colour had a great influence on the organoleptic perception of the foods and beverages and, in particular, on the perception of the marc spirits. It is well known that the visual aspect of a sample will influence the verbalisation of olfactory information and, indeed, its odour perception [85]. As the above data demonstrate (Figures 1–3, Table 3), the differences in the colour for the four aged grape marc spirits can be distinguished by the human eye. For that reason, the sensory analysis of the spirits was performed with the use of dark-coloured glasses in order to avoid bias for the odour perception of the grape marc spirits aged in contact with the four different wood types.

Table 3. Total colour differences (ΔEab*) between aged tsipouro samples by French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC).

Types of Wood			ΔEab*
FO	"vs"	AO	14.4
AO	"vs"	GO	17.4
GO	"vs"	GC	27.1
FO	"vs"	GC	30.2
FO	"vs"	GO	3.1
AO	"vs"	GC	43.0

3.3. Sensory Analysis

A trained panel of sixteen panellists orthonasally evaluated the aromas of all five samples. The sensory analyses of the tsipouro samples focused on the olfactory perception, as the visual differences among the samples were evident, as already mentioned above. The aim of the present work was to study the volatile compounds extracted from the tsipouro marc spirits, and not the non-volatile part, where the gustatory sensory analysis could affect the judgement of the tasters. The results from the tasting after the statistical treatment of the data are shown in Table S1, while Figure 4 presents the mean values in a spider plot form.

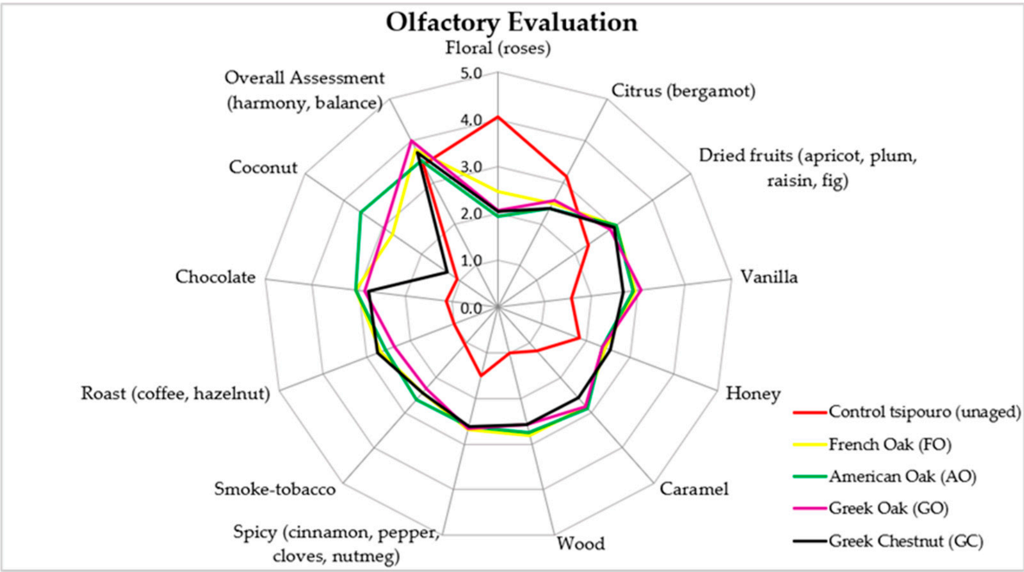

Figure 4. The mean value of the sensory attributes of the five different tsipouro marc spirits aged without wood contact (control) and in contact for 5 years with French oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC).

It is clear that for the majority of the studied sensory parameters, the control samples were rated differently from the samples aged in contact with wood to a statistically significant level (Table S1) and had a clear opposing trend, even if the attributes were not statistically different (Table S1 and Figure 4).

From Figure 4, it is evident that the tsipouro without wood contact exhibited aromas of the Muscat variety, primarily floral and citrus (bergamot), at a higher olfactory intensity than the other samples. The control sample scored higher for the floral notes and tended to have a higher aroma intensity for the citrus (bergamot) attribute, while the addition of wood reduced the intensity of floral aromas, possibly by masking the panellists' perception at a certain level. From the samples aged in contact with wood, the FO tended to have a higher perception of the floral attribute. On the other hand, the ageing process enhanced the perception of dried fruit senses (apricot, plum, raisin, and fig), with the control samples scoring relatively lower than the aged in contact with wood samples.

Comparing different types of wood, the sensory analysis provided a similar profile to what was observed from the GC–O–MS analysis (Table 2). Specifically, oak provided more intense aromas compared to chestnut, while the intensities of the aromas from the French and Greek oaks were at similar levels. American oak provided more pronounced notes of toasting and coconut, a characteristic aroma of this spice of oak, due to its presence of *cis*-whiskylactone, in accordance with the GC–O–MS analysis and other studies [16,84–88].

For the other attributes (vanilla, honey, caramel, wood, tobacco, spicy notes, coffee, hazelnut, chocolate, and coconut), it is clear that the extraction of the volatile compounds from the wood into the tsipouro during the ageing process produced spirits with a different organoleptic perception. For all those attributes, the aged with wood contact samples scored higher than the control sample, which is aligned with the results found during the GC–O analysis (Table 2). The samples with different wood types did not present significant differences for the majority of the attributes. However, a trend was observed. The chestnut samples (GC), although generally had lower intensities, especially in the "sweet" aromas such as vanilla, caramel, and chocolate, gave a greater aromatic intensity in characteristics such as roasted.

The sample of Greek oak, studied for the first time, showed a balance in the individual olfactory parameters with quite high intensities. It had the highest values in parameters related to typical oak barrel aromas, such as vanilla, due to the vanillin it contains, as well as caramel aromas related to the wood-toasting process. These characteristics make its aromatic profile similar to that of FO wood, with which they share a common botanical origin due to their European origin. The most significant differences between wood types were related to the intensity of the coconut aroma descriptor due to *Quercus alba*, as also reported by others [84–88].

The organoleptic evaluation of the various samples showed that, in general, all the aged spirits had a very good result regarding the overall aromatic assessment, quality, harmony, and balance. Given that the particular witness referred to a spirit of a particular, aromatic and pleasant character, it seems that the ageing of tsipouro can produce special, excellent spirits whose long-term ageing improves its characteristics in all the most well-known types of wood used. The organoleptic evaluation of the aromas of the tsipouro samples aged in the French and American oaks—the two well-known, most studied, and most used species of oak—confirms the qualitative dynamics of these two oak species in the case of the aged tsipouro as well. The use of Greek oak, for the first time in the ageing of spirits, produced spirits of great intensity in many qualitative aromatic characteristics and provided a harmonious and balanced aged marc spirit, which mainly resembles the characteristics of a spirit aged in French oak. Chestnut, also for the first time, was studied in relation to aged tsipouro and rated a high score in the overall assessment.

4. Conclusions

Ageing tsipouro with different oak species and chestnut wood led to a rich aromatic potential, imparting many pleasant volatiles derived from the wood, creating a complex effect, especially regarding the floral and fruity distillates of aromatic varieties, as are the majority of Greek tsipouro. Tsipouro from Muscat grapes produces high levels of terpenes, such as linalool, *a*-terpineol, citronellol, nerol, and geraniol, and additionally high levels of phenylethanol, 2-phenylethyl acetate. The compounds with floral and fruity aromas are distilled to tsipouro, and these compounds are even retained in aged spirits.

Greek oak (*Quercus trojana* or *Querqus macedonica*) was studied for the first time as an alternative oak species in terms of its aromatic profile regarding distillates. It appears to have a high-quality potential, imparting an intense and complex aroma to the spirit with a pleasant organoleptic character. It has a lot in common (botanically, chemically, and organoleptically) with the other oak wood species used in the ageing of wines and spirits and, especially, with the French oak (*Quercus petraia*). They share a similar colour, total polyphenolic index, chemical and organoleptic characteristics, and a number of substances derived from the toasting of the wood, such as guaiacol, *cis*- and *trans*-whiskylactones, 4-ethyl guaiacol, furaneol, 4-vinyl guaiacol, sotolon, vanillin, methyl vanillate, ethyl vanillate, are common with similar intensities in their aroma. Further investigation is needed to explore its potential for marc spirits' ageing with Greek oak, as well as other spirits and even wine.

On the other hand, chestnut (*Castanea sativa*), with its high phenolic potential and, which, over the years, has provided strong characteristics to aged spirits, produced very satisfactory organoleptic results, as well as the possibility of using it as an alternative to oak for ageing tsipouro and other spirits. However, its aromatic potential is less intense than that of the oak.

Both the Greek oak and the chestnut are endemic trees in Greece with great botanical, environmental, historical, and economic importance. Consequently, exploring the possibility of ageing spirits with these types of woods can contribute to the production of distinctive aged spirits and help strengthen the Greek economy.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/beverages10030076/s1, Figure S1: Distribution of (a) *Quercus trojana* (*Q. trojana* subsp. *trojana* (or *Q. macedonica*) with green colour, and *Q. trojana* subsp. *euboica* with violet colour, (b) *Quercus petraia*, and (c) *Castanea sativa* (probable native range and isolated population with green colour and introduced and naturalised range and isolated population since Neolithic with light brown colour); Figure S2: Picture of the traditional simple distillation without rectification copper-made alembic of 130 L composed of a gas heating system, a boiler, a still head, and a cooling system; Figure S3: Cutting process of the different type of woods (a) and (b) at cubes of dimensions of $2.0 \times 2.0 \times 1.0$ cm (c). Toasting process of wood cubes (d); Table S1: Mean values of sensory attributes of tsipouro marc spirits aged without wood contact (Control) and in contact for 5 years with France oak (FO), American oak (AO), Greek oak (GO), and Greek chestnut (GC).

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