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THE INFLUENCE OF ACTIVE COMPOUNDS OF WOOD CHIPS MADE FROM VARIOUS WOOD SPECIES ON THE ANTIOXIDANT, PHYSICOCHEMICAL AND SENSORY PROPERTIES OF APPLE BRANDIES

– Research paper –

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Abstract: The value of quality spirits is derived from their unique sensory profile, which is frequently achieved via the maturation of the distillate with wood. Traditionally these are barrels, but there is a growing interest in the use of wood chips, which offer new economic, environmental and sensory opportunities. The aim of this study was to ascertain the impact of wood chips (cherry, chestnut, oak, juniper) on the physicochemical attributes and sensory profile of apple distillate. Antioxidant activity (AOX), total polyphenol content (spectrophotometric method), acidity and ester content (titration method) and colour (CIELab) were determined. A sensory analysis was also conducted. The results demonstrated that the distillate was most influenced by chestnut wood, both in terms of its physicochemical characteristics (AOX 53 mg/100 mL, esters 0.83 g/L) and sensory qualities (4.5 points). The distillates aged with cherry and oak wood chips exhibited poorer outcomes (AOX 14 and 12 mg/100 mL, respectively), whereas the least impact on the physicochemical parameters was observed in the case of juniper wood chips (AOX 7.5 mg/100 mL). The maturation of apple distillates in wood chips presence led to enhanced sensory evaluation outcomes, with the most notable improvement observed in the case of chestnut wood chips.

Key words: wood chips; apple brandy; maturation; active compounds; sensory properties

INTRODUCTION

The maturation of distillates in wooden barrels represents a crucial stage in the production of alcoholic beverages, as it enhances the chemical composition and sensory characteristics of the resulting liquor. The processes that occur during maturation alter the taste, aroma and colour of the beverage, and impart a characteristic astringency (Guerrero-Chanivet et al., 2020) (Tarko et al., 2023). The primary process occurring during the ageing is the extraction of wood components. However, other chemical reactions, including oxidation, esterification, hydrolysis, ethanolysis, Maillard reactions, polymerisation and polycondensation, also occur (Guerrero-Chanivet et al., 2020).

Among the principal groups of alcoholic beverage compounds derived from wood, two are particularly worthy of note: low-molecular-weight polyphenols, such as tannins or benzoic and cinnamic acid derivatives, which originate from the thermal degradation of lignin, and furan compounds, which derive from the degradation of pentoses and hexoses (Madrera et al., 2010).

The ageing process may be conducted using barrels crafted from a variety of wood types. The selection of the appropriate wood is largely contingent upon the local production tradition of the specific alcoholic beverage in question. The most frequently utilised wood for the production of barrels is oak. This is due to the mechanical, physical and chemical properties of oak wood (Krstić et al., 2021). American oak (*Quercus alba*), pin oak (*Quercus robur*) and sessile oak (*Quercus petraea*) are utilised in cooperage (Alañón et al., 2011). In addition to oak, a variety of other wood species can be employed in the production of alcoholic beverages. These include chestnut (*Castanea sativa*), cherry (*Prunus avium*), acacia (*Robinia pseudacacia*), mulberry (*Morus alba* and *Morus nigra*), and beech (*Fagus sylvatica* L.) (Madrera et al., 2010) (Alañón et al., 2011) (Coldea et al., 2020) (Híc et al., 2020). A distinctive attribute of oak in comparison to other wood species is its lactone content, whereas chestnut is characterised by a high concentration of eugenol and isoeugenol. The

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dominant chemical compounds in oak and chestnut wood are furan compounds, phenolic aldehydes, phenolic ketones, tannins and elagotannins (Tarko et al., 2023). Cherry wood is characterised by high porosity and high oxidative capacity, which has been successfully applied for short ageing periods. Mulberry wood, in contrast, is distinguished by its fine and flexible texture, medium porosity, and low release of compounds during ageing (Coldea et al., 2020).

The maturation of beverages in the presence of wood from species other than oak, which is typically used in cooperage, is possible through the use of wood fragments, including wood chips, staves, blocks, etc. (Caldeira et al., 2009) (Canas at al., 2016) (Dumitriu et al., 2019) (Solar et al., 2021) (Krüger et al., 2022). Furthermore, the usage of wood chips enables the utilisation of stainless steel tanks for ageing. This circumvents the disadvantages inherent to barrel ageing, such as the difficulty in precise temperature control, the presence of undesirable microorganisms, evaporation losses or negative environmental impacts due to deforestation. The porosity of wood presents a significant challenge in the disinfection of barrels, a process which is not as problematic with steel tanks. Furthermore, the evaporation of liquids aged in wooden barrels occurs through the pores of the wood, which results in the loss of the final liquor. Furthermore, the necessity to produce new barrels can have a detrimental impact on the surrounding forest, particularly on slow-growing oak trees (Dumitriu et al., 2019) (Martínez Gil et al., 2018) (Baiano et al., 2016) (Smailagić et al., 2019) (Bajerski et al., 2021). Furthermore, the larger contact area between the beverage and the wood chips, in comparison to barrels, allows for a significantly reduced maturation time (Dumitriu et al., 2019). Another benefit of utilising wood fragments in lieu of barrels is the potential to employ diverse wood species during the maturation phase, thus enabling the production of a beverage exhibiting distinctive sensory attributes (Canas at al., 2016).

The aim of the present study was to investigate the influence of wood chips made from different wood species (oak, chestnut, cherry and juniper) on the sensory qualities of apple distillate and on its physicochemical and antioxidant properties.

MATERIALS AND METHODS

Material

The raw material used in the production of apple wines was composed of the fruit of the Red Boskoop and Ruby varieties (pomological orchard of the University of Agriculture in Krakow, Poland), while the wood chips utilised in the study were derived from various species of wood. Those made from chestnut, American oak and cherry were provided by 'Arôbois' (France), while juniper chips were supplied by 'Malinowy Nos' (Poland).

Diammonium salt of the 2,2′-azino-bis (3-ethylbenzothiazoline-6-sulfonic) acid (ABTS diammonium salt), (±)-6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid (Trolox), a phosphate buffer saline (PBS: 0.01 M phosphate buffer, 0.0027 M potassium chloride, 0.137 M sodium chloride; pH 7.4 at a temperature of 25°C) were obtained from Sigma-Aldrich. The chemicals: sodium hydroxide (NaOH), hydrochloric acid (HCl), sodium carbonate (Na_2CO_3), sodium pyrosulfite ($Na_2S_2O_5$), Folin-Ciocalteu reagent were obtained from POCh (Gliwice, Poland).

The wine preparation and maturation of distilllates

Wine produced from the Red Boskoop variety and wine produced from the Ruby variety were blended in a 1:1 ratio. Then the wine was subjected to straight distillation, yielding distillate with an alcohol content of 45% vol. The distillate was then subjected to fractional distillation, from which a middle fraction of 57% vol. was collected and diluted to 45% vol. Subsequently, a quantity of 0.5 g of wood chips of a specific variety was introduced to 250 mL of distillate, which was then subjected to a 20-week ageing process at a temperature of 20°C.

Fermentation and wine aging

The antioxidant activity (AOX) was determined in accordance with the methodology described by Re et al. (1999), using an active cation-radical of 2,2'-Azino-bis (3-ethylbenzothiazoline-6-sulfonic) acid (ABTS, Sigma Aldrich). AOX was calculated on the basis of a calibration curve, prepared each time for synthetic vitamin E ((±)-6-hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid, Trolox, Sigma-Aldrich) and expressed as mg of Trolox/100 mL

Total polyphenol content (TPC)

The total polyphenol content (TPC) was determined by the method previously described in detail by Tarko et al. (2018), namely the Folin-Ciocalteu method. The results were expressed as mg of (+)-catechin/100 mL.

Determination of acidity (PN-A-79529-10: 2005)

A volume of 50 ml of distilled water was added to 50 ml of distillate, and the mixture was heated on a water bath under a reflux condenser for 30 minutes. Subsequently, the sample was cooled and titrated with 0.1 M NaOH against phenolphthalein. The acidity was calculated based on the amount of reagent used and expressed in grams of acetic acid per litre of 100% distillate.

Determination of ester content (PN-A-79529-11: 2005)

Following the determination of acidity, 10 mL of 0.1M NaOH was added to the neutralised sample and heated under a reflux condenser for one hour. The sample was then cooled, 10 mL of 0.1M HCl was added and titrated with 0.1M NaOH against phenolphthalein. The amount of reagent utilized was employed to ascertain the ester content, which was then expressed as g of ethyl acetate per L of 100% distillate.

Determination of distillate colours by L*a*b* (CIELab) method

The method of colour determination (CIELab) of the distillates was conducted in accordance with the protocol described by the International Commission of Claristics (OIV, 2006). Distillate samples were centrifuged (15 min, 5000 rpm, MPW 350R centrifuge) and poured into spectrophotometric cuvettes. The absorption spectrum within the range 380-770 nm was measured using a Konica Minolta CM-5 spectrophotometer.

From the absorption spectra obtained, the CIELab colour parameters were calculated: L* (perceptual lightness; in the range from 0 – black to 100 – white), and chromatic coordinates: a* and b*. The a* axis is relative to the green–red opponent colours, with negative values toward green (–100) and positive values toward red (+100), while the b* axis represents the blue–yellow opponents, with negative numbers toward blue (–100) and positive toward yellow (+100). Furthermore, additional colour parameters were identified, namely hab (hue) and C*ab (chroma, also referred to as colour saturation or intensity). These were calculated using the following formulas (OIV, 2006):

$$h_{ab} = \tan^{-1}\left(\frac{b^*}{a^*}\right)$$

$$C^*_{ab} = [(a^*)^2 + (b^*)^2]^{\frac{1}{2}}$$

in which: h_{ab} – hue; C^*_{ab} – chroma; a^*, b^* – color coordinates.

Organoleptic assessment of apple distillate

The organoleptic evaluation method is an inhouse development based on PN-ISO 4121 (1998). All organoleptic analyses were conducted by a trained sensory panel comprising 15 members. Prior to undertaking the evaluation, the panellists were required to complete a series of tests to ascertain their suitability for the role, including assessments of gustatory and olfactory daltonism, as well as individual gustatory and olfactory sensitivity thresholds. The sensory evaluation was carried out in a dedicated sensory laboratory, with each panellist seated in a cubicle separated from their colleagues by walls. The temperature within the laboratory was maintained at 25℃. The beverage samples were served in tasting goblets made of glass, at room temperature, and each sample was marked with a three-digit code.

The basic sensory evaluation of apple distillates was conducted according to a 5-point scale, with 1 indicating a negative evaluation and 5 indicating a positive one. Four quality attributes were evaluated with weighting factors (given in brackets): colour (0.1), clarity (0.1), aroma (0.2) and taste (0.6). The purpose of weighting factors is to increase or decrease the importance of a particular attribute being evaluated. In this case, the importance of taste was the highest, as it is the primary sensory attribute of the product under evaluation.

Furthermore, the perception of aromas (fruit, herbal, toast, wood, smoke, chocolate, spices, caramel, coconut, vanilla, foreign) and tastes (sweet, sour, bitter, astringent, fruit, herbal, toast, wood, smoke, chocolate, spices, caramel, coconut, vanilla, foreign) were also assessed. A five-point scale was employed to assess the perceptibility of specific tastes and odours, with a value of 1 indicating an undetectable taste or odour and a value of 5 indicating an intensely perceptible taste or odour.

Statistical analysis

The physico-chemical analyses were conducted a minimum of three repetitions, whereas the basic sensory assessment of distillates was conducted by 15 panelists. The obtained results were presented as the mean with standard deviation (±SD). For these results the statistical analysis was conducted using the InStat v. 3.01 software (GraphPad Software Inc., USA). A single-factor analysis of variance (ANOVA) with post hoc Tukey's test was employed to ascertain the significance of differences between means. The Kolmogorov-Smirnov test was employed to ascertain the normality of the distribution.

The scores of the sensory perception for the individual taste and odour characteristics were

presented as the arithmetic mean of the 15 ratings given by the panel members.
Principal component analysis (PCA) was performed (Statistica, version 13, TIBCO Software Inc.) for the physicochemical parameters and the results of the organoleptic evaluation of apple distillates matured with different wood chips.

RESULTS

Total polyphenol content and antioxidant activity
An evaluation of the polyphenolic compound content and antioxidant activity of apple distillates aged with different wood species was conducted, and the results are presented in Figure 1. The control sample, which did not contain woodchips, was devoid of polyphenolic compounds, which is a notable contrast to the samples that did contain wood.
The distillates with chestnut wood chips added exhibited the highest total polyphenol content (17.78 mg catechin/100 mL). The samples that had been treated with cherry and oak wood chips showed comparable total polyphenol contents, with values of 5.01 and 4.82 mg catechin/100 mL, respectively. The observed differences were not statistically significant (Figure 1). Polyphenol content was found to be the lowest (2.63 mg catechin/100 mL) in the distillate containing juniper wood chips, which exhibited a value approximately twice as low as that observed in distillates containing cherry or oak wood chips. The antioxidant activity of the distillates matured with the various types of wood chips employed in this study was found to be significantly correlated with total polyphenol content ($R^2 = 0.999$). The samples containing chestnut wood chips exhibited the highest antioxidant activity (52 mg Trolox/100 mL) (Figure 1). This result is associated with the enhanced extractability of polyphenolic compounds with high antioxidant potential. The antioxidant potential of the distillates matured with oak and cherry wood was found to be significantly lower (12 and 14 mg Trolox/100 mL, respectively). The lowest antioxidant potential was observed in distillates matured in the presence of juniper wood chips (Figure 1).

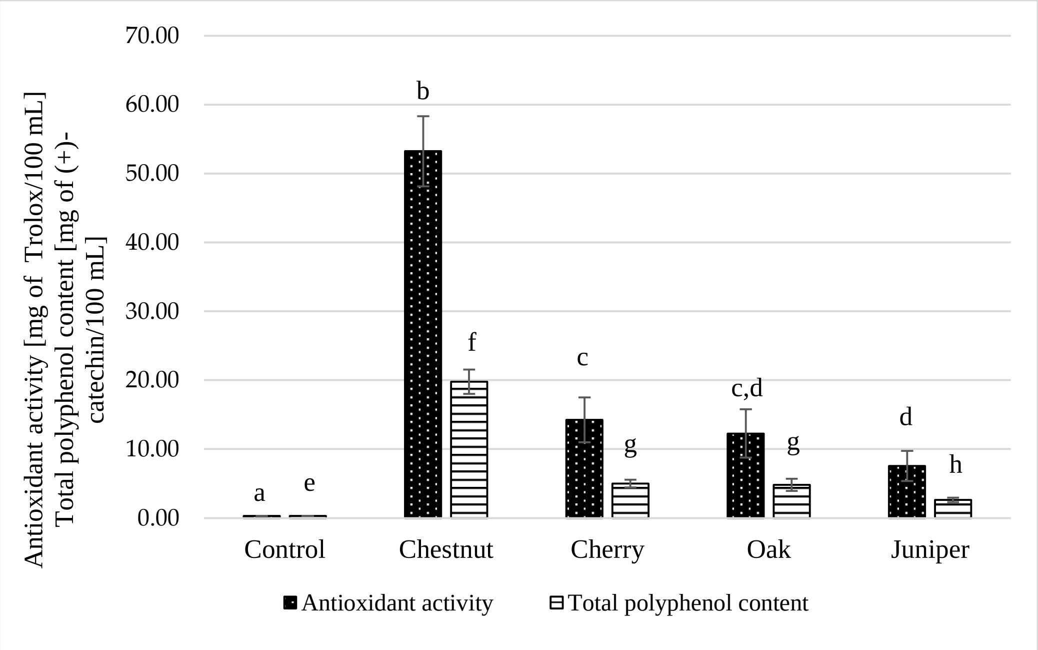

Figure 1. Total polyphenol content and antioxidant activity of apple distillates aged with wood chips from different tree species. The same letters within the analysed parameter indicate that there is no statistically significant difference for $p < 0.05$.

Acidity and ester content
The results of the acidity and ester content determination are presented in Figure 2. The control sample exhibited the lowest acidity (0.19 g/L) and ester content (0.35 g/L) among the samples tested. The ageing of apple distillates with wood resulted in a significant increase in acidity, which was dependent on the type of wood chips used. The chestnut distillates exhibited almost twice the acidity of the control sample (0.34 g/L), while the

cherry distillates demonstrated an acidity of 0.24 g/L. The lowest acidity values were observed in distillates with oak and juniper, with no statistically significant differences between them.

The impact of wood chips on the colour paramaters of distillates

Table 1 illustrates the colour parameters of the analysed apple distillates with the addition of different wood species. The brightness of the samples, as indicated by the L* parameter (ranging from 0 to 100), exhibited high values across all samples. The control sample, prepared without the addition of woodchips, exhibited the highest brightness, while the apple distillates with the addition of chestnut chips displayed the lowest brightness. Samples containing cherry, oak, and juniper chips exhibited similar L* values, with no statistically significant differences.

A strong positive correlation (R^2 = 0.92) was identified between the acid and ester contents of the apple distillates. The control sample exhibited the lowest ester concentration (0.35 g/L), while the remaining distillates demonstrated ester concentrations ranging from 0.55 to 0.85 g/L. The highest concentration of esters was observed in distillates that had been matured with chestnut wood (0.85 g/L). The addition of cherry wood resulted in a nearly twofold increase in ester concentration compared to the control sample. The lowest ester concentrations were observed in distillates matured with oak and juniper (Figure 2).

The h_{ab} parameter (0°-360°) was employed to determine the hue of the samples, while the C^*_{ab} parameter (0-100) was utilised to characterise the colour saturation. All samples exhibited a green-yellow hue. The control sample had the highest H_{ab} value (99.88) and a low C^*_{ab}, indicating a grey hue. The distillates with chestnut, cherry, and oak demonstrated comparable h_{ab} values (97.40-98.06), with a greenish tinge and no statistically significant differences. The distillate containing juniper (h_{ab} = 94.07) exhibited a yellowish tinge. The degree of colour saturation demonstrated a notable variation between samples, with the chestnut sample exhibiting the highest level and the juniper sample displaying the lowest.

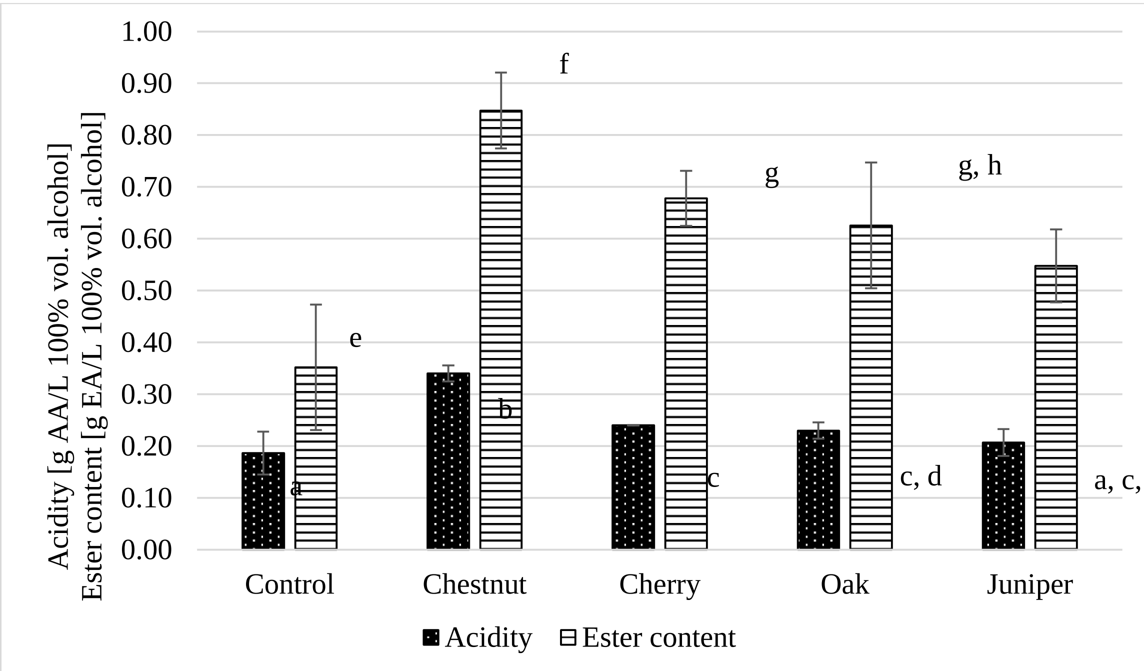

Figure 2. Acidity and ester content in apple distillate aged with wood chips from different tree species.
The same letters within the analysed parameter indicate that there is no statistically significant difference for $p < 0.05$

Table 1. The impact of apple distillate maturation in the presence of diverse wood species on colour parameters.

Sample	L*	a*	b*	h_{ab}	C^*_{ab}
control	95.81±0.12[a]	-0.08 ± 0.01[a]	0.48 ±0.01[a]	99.88±1.26[a]	0.48±0.01[a]
chestnut	93.22±0.28[b]	-1.54±0.11[b]	11.87±1.34[b]	97.40±0.41[b]	11.97±1.35[b]
cherry	94.18±0.38[c]	-1.26±0.12[c]	8.86±1.16[c]	98.06±0.31[b,c]	8.95±1.17c
oak	94.24±0.52[c,d]	-1.01±0.14[d]	7.34±1.78[c]	97.93±0.82[b,c]	7.41±1.78[c]
juniper	94.71±0.14[c,d]	-0.26±0.03[e]	3.60±0.19[d]	94.07±0.70[d]	3.61±0.19[d]

The same letters indicate a lack of statistical significance, for $p < 0.05$.

Organoleptic assessment of distillates

An organoleptic assessment of the apple distillates obtained and aged with different types of wood chips was conducted. The following indicators were considered in the evaluation: colour, clarity, aroma, flavour. The results are presented in Table 2. The distillate constituting the control sample was deemed the least favourable by the sensory panel, with a rating of 3.9. The addition of different wood chips to the samples resulted in higher ratings, yet no statistically significant differences were observed between the various types of wood employed for ageing (Table 2). Distillates matured with chestnut and oak wood chips exhibited the highest ratings (4.5 and 4.4 points, respectively), indicating a positive effect of even short-term maturation on the sensory evaluation results.

A comprehensive assessment of the aroma and taste profile of the apple distillates was also conducted, evaluating the intensity of selected characteristics by sensory panel. The apple distillate without wood (control sample) was distinguished by a pronounced fruit aroma, stronger than that of the distillates aged with wood chips (Figure 3). All distillates possessed a discernible fruit aroma, with nearly undetectable spicy or floral aromas. Furthermore, no toasty aromas were exhibited. It is noteworthy that all distillates, including the control, manifested a perceptible woody aroma. Distillates aged with chestnut wood chips exhibited moderately perceptible woody aromas, while those aged with cherry wood chips displayed herbal and vanilla-like notes. Distillates aged with oak exuded woody, smoky, and vanilla aromas. Very unusual notes were observed in samples aged with juniper woodchips - mainly citrus and herbal.

The primary factor influencing the overall sensory evaluation was taste. Apple distillate matured without wood was characterised by low perceptibility of the taste notes evaluated in the sensory test (Figure 4).

The addition of wood chips during the ageing stage increased the perspicuity of the woody flavour. Chestnut wood had a particular influence on the astringent and spicy taste. Distillates aged with oak and cherry wood chips exhibited comparable levels of wood flavour, with a more pronounced sour aftertaste in the case of oak wood and a spicy aftertaste in the case of cherry wood. Distillates made with the use of juniper wood chips wood chips were characterised by sweet, citrus and bitter flavours.

Table 2. The impact of apple distillate maturation in the presence of diverse wood species on the sensory evaluation outcomes.

Sample	Organoleptic assessment
control	3.86±0.53[a]
chestnut	4.51±0.51[b]
cherry	4.09±0.58 [b]
oak	4.41±0.50 [b]
juniper	4.01±0.64 [b]

The same letters indicate a lack of statistical significance, for $p < 0.05$, 5-point scale, n=15.

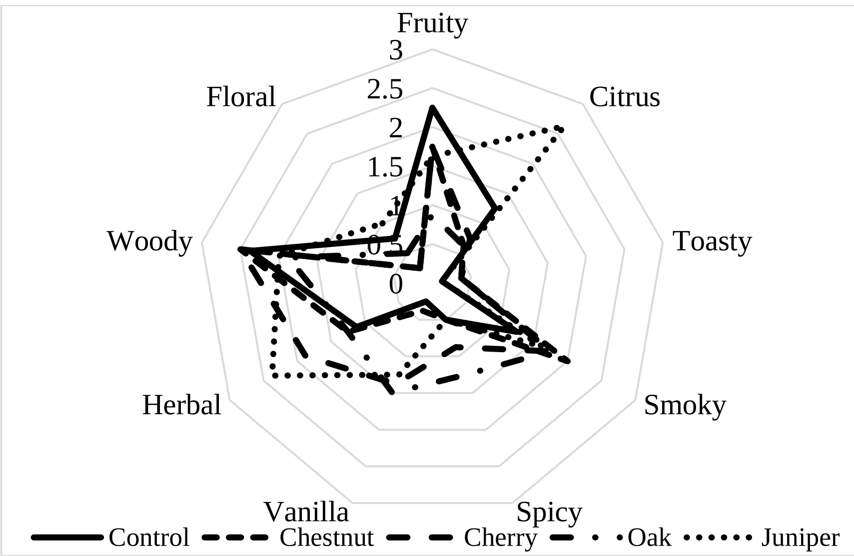

Figure 3. The impact of apple distillate maturation in the presence of diverse wood species on additional aroma characteristics, 5-point scale, n=15.

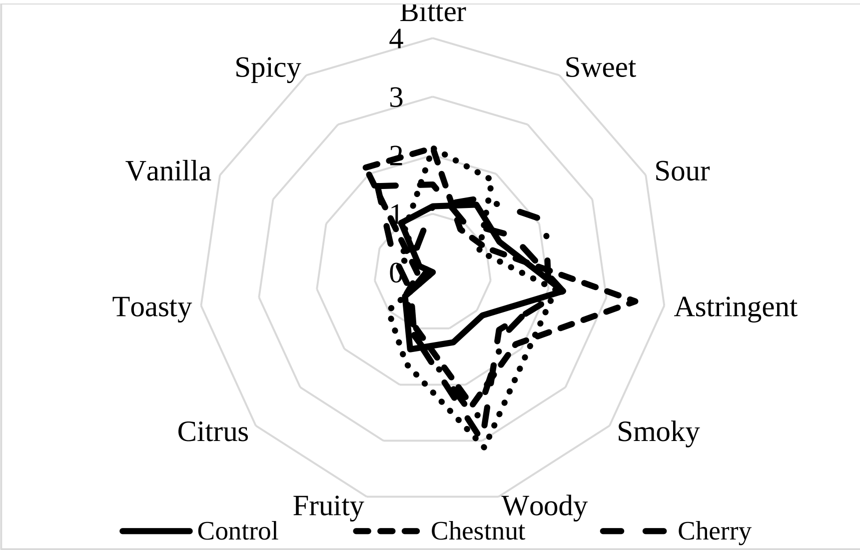

Figure 4. The impact of apple distillate maturation in the presence of diverse wood species on additional taste evaluation 5-point scale, n=15.

Principal component analysis (PCA)
The component variable PC1 is positively correlated with floral and citrus aroma and fruity and sweet taste, and to a lesser extent with citrus aroma and taste. The PCA1 values are inversely correlated with the basic organoleptic evaluation, with all physico-chemical analysis results and with the toasty and smoky aroma and toasty taste (Figure 5). The component variable PC2 is correlated with sour and vanilla taste and spicy aroma. PC2 values are inversely correlated with bitter taste and fruity aroma.

A clear correlation exists between antioxidant activity and polyphenol content, as well as between these variables and sample acidity. Additionally, there is a positive correlation between organoleptic evaluation results and the perception of toasty taste and aroma, smoky aroma, and ester content. The results of the organoleptic evaluation do not correlate with the perception of sour, vanilla, or bitter taste, nor with the perception of spicy or fruity aroma.

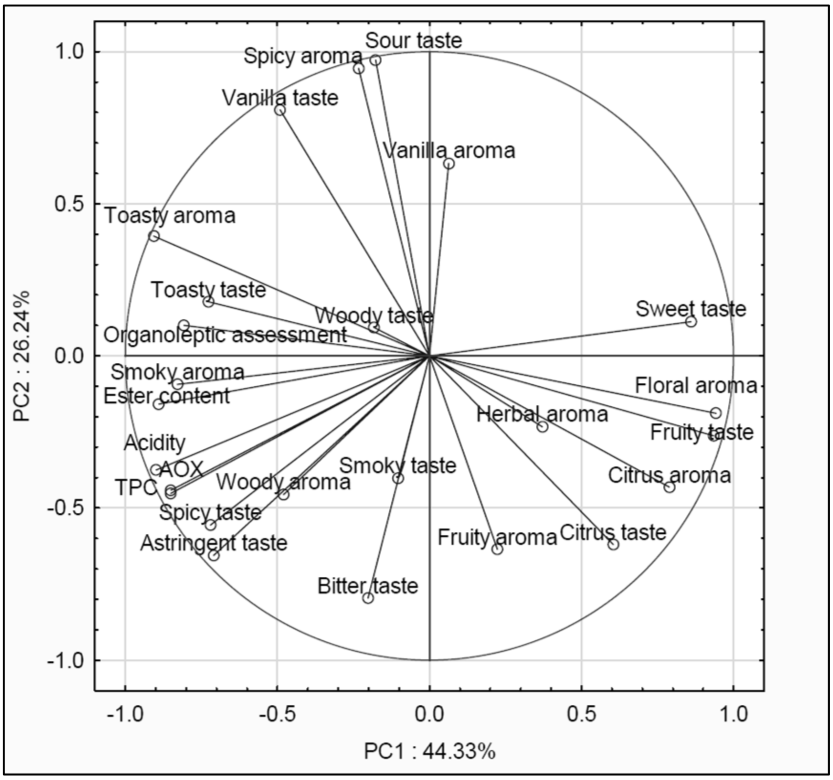

Figure 5. Principal component analysis demonstrating the impact of the addition of chestnut, cherry, juniper and oak chips on the chemical composition and organoleptic assessment of apple distillates.

The values of the analysed parameters remained largely unchanged in the samples that had been matured with juniper wood chips in comparison to the control sample. Similar results were obtained for the samples matured with cherry and oak wood chips. It is evident that the analysed attributes exhibit elevated values in the samples that underwent maturation with chestnut wood chips (Figure 6). The component variable PC2 was found to be correlated with the perception of sour taste, spicy aroma and, to a lesser extent, vanilla taste and aroma. The PC2 values were inversely proportional to the intensity of the bitter taste (Figure 6). The control and juniper samples exhibited a higher floral and citrus aroma, as well as a relatively low acidity, AOX, TPC and toasty, smoky aroma. The samples that had been matured with oak and cherry chips displayed a higher sour taste, spicy aroma and the least bitterness. The samples that had been matured with chestnut chips exhibited an astringent and spicy taste.

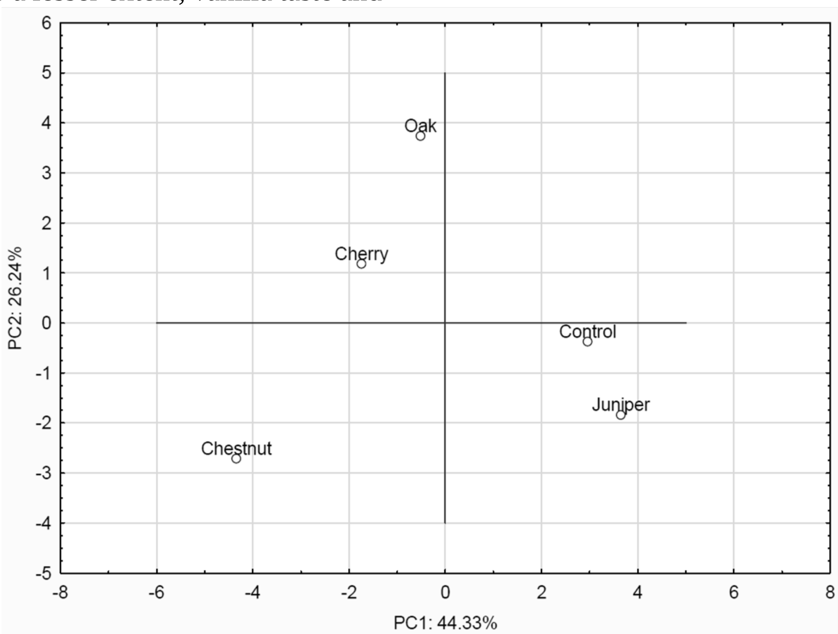

Figure 6. Principal component analysis – plot of component weights.

DISCUSSION

In our study, apple distillates aged with chestnut wood chips were characterised by the highest polyphenol content, while samples matured with cherry and oak wood exhibited lower concentrations. Similar outcomes have been reported by other researchers. In a study by Coldea et al. (2020), investigating the phenolic composition of Romanian apple brandy matured with different types of wood, it was demonstrated that the control sample, which did not contain wood, exhibited a limited quantity of phenolic compounds, with chlorogenic acid (presumably derived from the fruit) representing the predominant component of the phenolic profile of apple wine. Converging results were also obtained by Hic et al. (2020) and Madrera et al. (2010), who also identified the highest polyphenol content in distillates matured with chestnut wood, compared to other wood species. In contrast, Guerrero-Chanivet et al. (2020) demonstrated that wine distillate matured with unburned cherry wood chips exhibited a significantly higher concentration of polyphenolic compounds than that matured with chestnut wood chips. This is because the content of polyphenolic compounds is influenced by two factors: the tree species and the degree of wood roasting. During high temperatures, lignins and polysaccharides undergo decomposition, which leads to an increase in polyphenolic compounds (Le Floch et al., 2015). Although juniper chips induced the least significant increase in polyphenol concentrations in comparison to control, these results are of interest as the maturation of apple distillates with juniper wood chips has not previously been reported in the scientific literature. Only ethanolic and aqueous extracts of juniper fruit have been the subject of study (Elmastaş et al., 2006), and the determined content of total polyphenols (16.2 μg gallic acid/mL) was several times higher than those obtained by maturing apple distillate with juniper wood chips used in the present study. It is important to note, however, that juniper fruit contains a significantly higher concentration of terpenoid compounds than wood. Nevertheless, the profile of these compounds is found to be similar in fruit and wood (Barjaktarowic et al., 2005).

Polyphenolic compounds are natural antioxidants that are found in plants. During the maturation of alcoholic beverages, polyphenols are extracted from the wood into the distillate, thereby increasing its antioxidant activity (Alañón et al., 2011). The principal compounds accountable for the antioxidant capacity of chestnut distillates are thought to be tannins and their derivatives, including gallic acid (mean value 151 mg/L) (Madrera et al., 2010). The reduced antioxidant capacity of distillates aged with cherry and oak wood may be attributed to their compact structure, which impedes the penetration of the distillate into the wood chips. The profile of polyphenolic compounds in the different wood species also has a significant impact on the antioxidant activity of the extracts. In contrast with the findings presented in this paper, a study by Híc et al. (2020) demonstrated that the total polyphenol content was not correlated with antioxidant activity in the case of cherry wood. The authors suggest that this may be due to the presence of other types of polyphenols in cherry wood, which exhibit lower antioxidant capacities compared to the other wood species tested.

The organic acids present in fruit distillates are primarily the result of yeast metabolism, whereas the esters are compounds formed by the esterification of acids and alcohols during fermentation. These compounds, despite their low concentrations, significantly affect the aroma and character of alcoholic beverages (Biernacka, 2012). Their concentration depends on the raw material, the yeast strain and the microbiological purity of the fermentation. The maturation of quality vodkas results in the formation of particularly high amounts of acids and esters, particularly when the vodka is in contact with wood (Wiśniewska. 2016). In a study conducted by Januszek et al. (2020), the most prevalent group of volatile compounds in apple brandy was identified as esters, comprising over 60 distinct compounds. These included methyl esters, ethyl esters, acetates, benzoates, and others. Methyl and ethyl esters were the most abundant group, comprising over 26 compounds.

The predominant esters among the 23 identified in apple distillates in the work of Balcerek et al. (2023) were ethyl acetate, ethyl decanoate, ethyl benzoate, ethyl tetradecanoate, ethyl acetanoate, ethyl hexanoate, ethyl propanoate, methyl acetate, 2-phenylethyl acetate, isoamyl acetate. In other studies on apple distillates (Spaho et al., 2021) the authors found the following esters: isobutyl acetate, ethyl lactate, ethyl isovalerate, isoamyl acetate, diethyl succinate, ethyl decanoate, ethyl tetradecanoate, ethyl octanoate, ethyl hexanoate. In the experiments of Coldea et al (2020), 28 esters were identified in Romanian apple brandy, with ethyl acetate, ethyl 2-methylbutyrate, ethyl isovalerate, isoamyl acetate, ethyl hexanoate, ethyl octanoate, hexyl 2-methylbutanoate, ethyl decanoate, ethyl laurate in the highest concentrations. The compounds described are characterised by different aromas, as shown in Table 3.

In their 2020 study, Coldea and colleagues (2020) observed an increase in the concentration of esters, particularly compounds such as ethyl acetate, isobutyl acetate, isoamyl 2-methylbutyrate, ethyl benzoate, ethyl nonanoate, methyl decanoate, ethyl decanoate and ethyl dodecanoate, during the maturation of apple brandy with wood.

Table 3. Aromas of esters occurring in highest concentrations in apple distillates (Madrera et al., 2015).

Compound	**Aroma**
Ethyl acetate	Fruity, solventy, perfumy
Isoamyl acetate	Banana
2-Phenylethyl acetate	Roses honey
Methyl acetate	Glues and nail polish removers
Ethyl propanoate	Pineapples, grapes, and tropical fruit
Ethyl hexanoate	Apple peel fruit
Ethyl octanoate	Fruity fat
Ethyl decanoate	Grape
Ethyl lactate	Mild, buttery, creamy, with hints of fruit and coconut
Ethyl benzoate	Floral camomile
Ethyl tetradecanoate	Ether
Isobutyl acetate	Fruity, sweet, and floral
Ethyl isovalerate	Fruity, sweet apple, pineapple
Diethyl succinate	Very mild, predominantly fruity, with hints of leaf green and cooked apples
Hexyl 2-methylbutanoate	Strawberry
Ethyl 2-methylbutyrate	Green-fruity
Ethyl laurate	Floral, fruity

The greatest increase in these compounds was observed in samples containing mulberry, chestnut, and cherry wood, while fir and walnut wood samples exhibited a decrease in ethyl isobutyrate and ethyl isovalerate. Ethyl acetate typically constitutes the majority of esters in spirits, representing approximately 80% of all esters (Bortoletto and Alcarde, 2013).

Coldea et al. (2020) determined the ethyl acetate content to be between 3.9 and 4.2 g/L, which is several times higher than the ester content of the distillates in the present work (0.35-0.85 g/L). The discrepancies between the findings of the Coldea et al. (2020) study and the present investigation can be attributed to the quantity of wood chips incorporated. In the aforementioned study, the authors employed 30 g of wood chips per L of distillate, whereas the present study utilized 2 g/L of distillate. In both studies, the addition of chestnut and oak wood chips resulted in an increase in the amount of esters.

The formation of ethyl acetate is enhanced when distillates are matured with roasted wood chips (García-Moreno et al., 2020), as acetic acid is also produced as a by-product of the thermal degradation of wood. Additionally, the authors observed that the incorporation of cherry wood chips resulted in a reduction in the ethyl acetate concentration. This finding contrasts with the results of the present study, where the ester content in the distillate with cherry wood chips was comparable to that of the distillate with oak wood chips. The reduction in ethyl acetate concentration may have been attributed to the oxidative properties of cherry wood (Coldea et al. 2020) (Chinnici et al., 2015).

Brandy is characterised by a golden-yellow or amber colour, the intensity of which in distillates aged in wood depends on the content of phenolic compounds (Canas, 2017). In the present study, the control sample was almost colourless, displaying the highest brightness, while the L* parameter for the chestnut woodchip distillate exhibited the lowest value (Table 1). This may be related to the higher content of polyphenolic pigments, which correlates with the results of the polyphenol analysis (Figure 1). Similarly in their study, Híc et al. (2020) examined the colour of distillates aged with wood chips from various wood species. They observed that distillates from cherry, oak, and chestnut exhibited a greenish hue. Additionally, they found no correlation between the degree of wood toasting and the values of the a* and b* parameters. An increase in the toasting temperature of the wood chips resulted in an elevated b* value, towards a yellow colour. This phenomenon may be attributed to the oxidation of elagotannins and the condensation reaction of tannins with aldehydes, as observed in the aging of wine (García-Moreno et al., 2020). The species of wood exerted an impact on the chromatic characteristics of the distillates, with the greatest colour differences occurring in distillates matured with chestnut wood, which may be explained by its high porosity and the increased extraction of tannins and phenolic compounds (Delgado-Gonzalez et al., 2020).

Coldea et al. (2020) reported an increase in certain fruit-flavoured esters (such as isobutyl acetate, isoamyl acetate, hexyl acetate, hexyl 2-methylbutanoate, ethyl decanoate) in Romanian apple brandy with the addition of wood chips, primarily derived from oak, but also from chestnut, mulberry, and walnut.

The woodchips used in the study were not heat-treated, so that the toasty aroma so characteristic of toasted wood was not identified, but they were characterised by a strong woody aroma. This aroma is characteristic of raw wood chips (Martínez Gil et al. 2018) (Carpena et al.,2020). A faint vanilla aroma was perceptible in the distillates. According to Caldeira et al. (2002), wood from the chestnut tree is particularly rich in vanillin, a substance that lends a distinctive vanilla aroma to spirits distilled in barrels made from this wood. Juniper wood chips contributed to the identification of intense citrus and herbal aroma notes. A similar pattern was observed in the aroma evaluation of gins in a study conducted by Aumatell (2012), in which all tested gin samples were characterised by a citrus aroma.

The addition of chestnut wood chips resulted in a greater perception of the astringent and spicy flavours, which is likely attributable to the high tannin content (Madrera et al., 2010). The maturation of distillates with non-heat-treated wood chips typically results in the weak perception of toasty, smoky, and spicy flavours (Híc et al., 2020). Guaiacol and 4-methylguaiacol, compounds responsible for these flavours are formed after heat treatment of the wood (Kennison et al., 2008). In turn, juniper wood chips are a source of terpenes, including α-terpineol and limonene (Zawirska-Wojtasiak, 2006), which may be responsible for the sweet, citrus and bitter taste in apple distillates aged with these wood chips.

CONCLUSIONS

The utilisation of assorted wood chips for the maturation of apple distillates exerts a considerable influence on their sensory and physicochemical attributes. The utilisation of chestnut wood has been demonstrated to enhance the antioxidant activity and optimise the aroma and flavour of the distillates. The effects of oak and cherry wood chips on the physicochemical and sensory characteristics of the distillates are comparable to those of chestnut wood chips. Conversely, the utilisation of juniper wood chips can impart distinctive citrus notes, yet does not markedly influence the antioxidant potential, polyphenol content, acidity, or ester content. The selection of an appropriate wood species is contingent upon the desired sensory and technological characteristics of the beverage, thereby facilitating the customisation of the product to align with diverse consumer preferences.

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