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Research article

THE IMPACT OF TYPE OF GRAPE VARIETY ON THE VOLATILE AROMA COMPOUNDS AND SENSORY PROPERTIES OF GRAPE BRANDY IN SOUTH-WEST OF ROMANIA

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Abstract

This paper presents the results of a study that examined the impact of grape variety on the aromatic profile and sensory characteristics of distilled beverages produced in the Southwest of Oltenia, Romania. Six grape varieties were used: neutral (Fetească Regală and Fetească Albă), semi-aromatic (Sauvignon), and aromatic (Tămâioasă Românească, Muscat Ottonel, and Muscat of Alexandria). The pomace brandies were obtained by distilling the grape pomace from the mentioned varieties using a traditional copper still, under identical conditions.

The gas chromatography–mass spectrometry (GC/MS) method was applied to identify 30 aromatic volatile compounds belonging to the groups of alcohols, volatile acids, volatile esters, terpenes, and volatile aldehydes. Sensory evaluation of the distillates was also carried out to determine the typical characteristics of the analyzed brandies.

Alcohol content, fatty acid esters, and terpenic compounds were significantly higher in all brandies obtained from aromatic grape varieties compared to those from neutral or semi-aromatic varieties. A higher average content of 2-phenylethanol was detected in aromatic brandy samples (mg/L in TR) compared to all other brandies. In neutral/semi-aromatic grape brandies, the highest value was found in FR (6.23 mg/L).The linalool content, which contributes significantly to the aroma of roses, anise seeds, grapefruit, lime, and citrus, is significantly higher in aromatic brandies (0.68 mg/L - TR and 1.15 mg/L - MO) compared to brandies from neutral/semi-aromatic varieties (0.25 mg/L - FR and 0.34 mg/L - S).The research results showed that the grape variety had a significant impact on the aromatic volatile compounds and the sensory properties of the brandy. Sensory analysis supports the chemical determinations, showing that all brandies fall into the category of high-quality spirits.

1.Introduction

The distillates obtained from the fermentation of grape marc exhibit a much more complex chemical composition than the original fruits. Recent advances in analytical techniques and laboratory equipment have enabled a deeper understanding of the biochemical components of natural spirits. Grape marc represents the solid residue resulting from the vinification process, separated from the must, and consists—depending on the grape variety and the vinification technology used—of varying proportions of stems, skins, and seeds (Gheorghiță *et al.,* 2002).

The sensory quality of fresh grape marc distillates is determined exclusively by their volatile aromatic compound composition, which is fundamental to consumer perception.

Volatile aromatic compounds—including alcohols, volatile acids, esters, terpenes, aldehydes, and acetals—serve as key sensory markers and quality indicators in alcoholic beverages (Wei et al., 2018). The varietal aromatic potential of grapes is a major factor influencing the character, quality, and distinctiveness of distillates, especially in those produced from Muscat varieties.

Terpenes compounds represent the primary group of aromatic compounds found in grapes (V. vinifera), wines, and distillates derived from Muscat varieties. The most significant among them are terpineol, linalool, geraniol, nerol and citronellol. These compounds are chiefly responsible for the delicate floral and aromatic notes and constitute the key factors influencing the aroma profile of both Muscat and non-Muscat cultivars (Matijaševic et al., 2019).

The alcohol content has a major influence on the quality of the distillate, as it contributes to its body and aromatic complexity. High alcohol values are not beneficial to the quality of distillates (Christoph et al., 2007; Tsakiris et al., 2014). Thus, high alcohol concentrations in distilled beverages are associated with oily, green, cut grass aromas; aromas of grass and herbaceous plants (1-hexanol); and aromas of rose and honey (phenylethyl alcohol), while 1-heptanol is associated with an oily aroma (Soufleros et al., 2004; Matijaševic et al., 2019).

Esters are key compounds that predominantly enhance the pleasant fruity and floral aromas of distilled beverages (Soufleros et al., 2004; Matijaševic et al., 2019). Among them, ethyl esters of medium-chain fatty acids—such as hexanoic, octanoic, decanoic, and dodecanoic acids—are particularly important, as they positively influence the aroma profile by imparting distinct fruity and floral notes (Christoph et al., 2007; Lukic et al., 2011; Tsakiris et al., 2014; Matijaševic et al., 2019). Fatty acids themselves play a crucial role in determining the sensory quality of distilled beverages, acting as precursors for volatile compounds and contributing to aroma development through their association with various aromatic groups, including esters (Christoph et al., 2007; Tsakiris et al., 2014; Matijaševic et al., 2019).

Aldehydes, which form an extensive class of aromatic compounds, are often cited as having a negative impact on the quality of distilled beverages (Lukic et al., 2011). Research shows that total aldehyde levels exceeding 1200 mg/L (especially acetaldehyde) significantly influence the aroma of the distillate and indicate marked oxidation of ethanol during the fermentation process (Soufleros et al., 2004; Plutowska et al., 2010; Lukic et al., 2011).

Acetals, on the other hand, contribute to the aromatic profile of many fruit-based alcoholic beverages, giving them a fine and pleasant taste and bouquet (Cortés et al., 2005; Plutowska et al., 2010).

Distillates obtained from grape marc stand out from other similar products and are highly valued by consumers due to their added value, which lies in the specific production technology closely tied to the history and traditions of a particular country or geographical region (Lukić I. et al., 2011).

In Drăgășani-Vâlcea, there are favorable environmental conditions and a long tradition of vine cultivation and, implicitly, wine and distilled beverage production (Stoica et al., 2019).

Grape marc spirit is a traditional product, obtained mainly from neutral-flavored grape varieties. The use of marc from aromatic varieties to obtain superior natural distillates contributes to enriching the sensory profile of the final product, giving it complex, fruity, and floral aromas. These distillates are distinguished by their finesse, balance, and authenticity, while also reflecting the particularities of the grape variety and the winemaking traditions of the region of origin.

The aim of this study was to analyze the aromatic profile of grape marc distillates obtained from both neutral and aromatic varieties, both autohton and international. In addition, the research aims to clarify the link between the aromatic composition and the sensory qualities of fresh grape marc distillates.

2.Materials and methods

The aim of this study was to determine the chemical composition of distilled beverages obtained from fermented grape pomace from several white grape varieties, including neutral and aromatic types, both native and international, distilled using a traditional still.

2.1. Grape samples.

For this study, six white grape varieties were used, including three neutral or semi-aromatic varieties—Fetească Regală (FR), Fetească Albă (FA), and Sauvignon (S)—and three aromatic varieties—Muscat Ottonel (MO), Muscat of Alexandria (MA), and Tămâioasă românească (TR). Each variety was used for the production of distillates in Romania, particularly in the Oltenia region (southwest of the country).

All six varieties were harvested from a vineyard located on the Olt Hill, in the Drăgășani wine region, Vâlcea County (44°39′40″N 24°15′38″E). This region enjoys a temperate climate, with weak Mediterranean influences in the south and a limited continental influence in the north. The average temperature during the hottest month, July, ranges between 21.5 °C and 22 °C. The characteristics of the climate are well-defined, featuring warm and scorching summers as well as severe winters. During the coldest months, January and February, the average temperature is between -2 °C and -2.3 °C. Temperature records indicate a maximum of 41.3 °C recorded on August 17, 1952, and a minimum of -33.5 °C on January 24, 1942. According to precipitation data, the region experiences a dry period in the summer, while the rest of the year benefits from abundant rainfall, totaling 578 mm annually. The predominant wind directions include north (14.8%), northeast (10.8%), southwest (8.6%), east (8.5%), and northwest (8.2%). Grapes were sampled at optimal technological maturity (21.7–22.2 Brix for neutral/semi-aromatic varieties and 22.5–23.7 Brix for Muscat-type aromatic varieties) between 10 and 15 September 2023.

2.2. Alcoholic fermentation of grape marcs

After harvest, the grapes were transported to the Faculty of Horticulture in Craiova, Department of Horticulture and Food Science. All grapes were vinified separately, and the resulting pomace, after crushing and pressing, was placed in vessels with a capacity of 150 L each. The vessels were filled to 70% of their volume.

Fermentation occurred spontaneously, triggered by the indigenous microflora of wine yeasts, at 20 °C ± 1 °C. The fermentation process was monitored daily, and it continued until the sugar concentration decreased to 4 °Brix.

2.3. Distillation of fermented grape marcs samples

The fermented grape pomace was distilled in a traditional copper still with an agitator, with a capacity of 80 L. Before starting the distillation, the still was sealed to prevent any vapor loss. Uniform and consistent distillations were achieved by gently heating the still at the beginning of the process, increasing the heat during the main flow, and reducing it toward the end. The water temperature in the cooling tank was maintained between 20 °C and 22 °C.

During the distillation of the fermented material, three main fractions were separated: the first fraction (heads), the middle fraction (heart of the distillate), and the tail fraction. The middle fraction was collected as the fresh distillate. Distillates were stored in dark, tightly sealed bottles at 20 °C for 12 months. All distillates were analyzed using gas chromatography and following the method used by the Laboratory of the Department of Horticulture and Food science and the laboratory of National Institute for Cryogenics and Isotopic Technologies (I.C.S.I. Rm. Valcea).

2.4. Chemical reagents

Ultrapure water, HPLC-grade dichloromethane, methyl 10-undecenoate, anhydrous sodium sulfate, and sodium chloride were purchased from Sigma-Aldrich (Steinheim, Germany). Ultrapure HPLC-grade dichloromethane from Merck (Darmstadt, Germany) was used as the extraction solvent.

2.5. Analysis of the chemical quality parameters of grape brandy

In accordance with the official Romanian legislation regarding sampling methods and the performance of chemical and physical analyses of alcoholic beverages, the following analyses were carried out:

The alcoholic strength by volume was determined after distillation using a pycnometer (Publications Office of the European Union. Commission Regulation (EC) No 2870/2000).

Methanol content was analyzed following the European Community Reference Methods for the analysis of spirits, using gas chromatography (GC) with a flame ionization detector (FID). Methanol was determined by direct injection of the sample into a gas chromatography system (GC 2010 plus, Shimadzu, Japan). As an internal standard, 2-butanol was added to the spirit prior to injection. Methanol was separated using temperature programming on an SPB®-624 capillary column (30 m × 0.25 mm, df 1.40 µm) and detected with an FID detector.

The methanol concentration was quantified relative to the internal standard, using response factors obtained during calibration under the same chromatographic conditions as those used for the analysis of the spirit (Publications Office of the European Union. Commission Regulation (EC) No 2870/2000; SR 184-2:2010).

2.5.1. Extraction and concentration of minor volatile compounds

The volatile compounds were extracted following the method proposed by Raicevic et al., (2022). Fifty milliliters of distillate were mixed with 100 mL of ultrapure water, and 20 mL of the internal standard (methyl 10-undecenoate, 1 mg/L) was added. The mixture was then extracted with 40 mL of dichloromethane. Sodium chloride (10 g) was added, and the solution was stirred magnetically for 30 minutes. The layers were separated in a separatory funnel, and the organic layer was dried over anhydrous sodium sulfate for 2 hours. The extract was concentrated under a nitrogen stream to 1 mL and analyzed directly by GC/MS (Raicevic *et al.,* 2022).

2.5.2. GC/MS Analysis of minor volatile compounds

Quantitative determination of volatile compounds was performed using a gas chromatography system, a VARIAN 450 gas chromatograph GC-FID detector (flame ionization detection) with a set of 275°C temperature for both the column TG-WAXMS 60 m, ID 0.32 mm, film, 0.25 mm, injector temperature 150°C, column temperature 35°C, 3 min stand, climb to 20°C/min, up to 70 to 150°C with 27°/min, stand 2 minutes, climb 200°C, stand 2 minutes, climb to 240°C with 20°C/min and stands 6 min. The carrier gas was helium (1.2 ml/min flow rate). Injection volume is 1 µl. The identification was made by comparing the retention times of standards from the calibration curve.

2.6. Sensory analysis

The sensory evaluation of spirits samples was performed using the Buxbaum (Raicevic et al., 2022) model with a positive score of 20, which is the globally accepted method for the sensory evaluation of spirits. This model is based on five sensory analyses: color, clarity, distinctiveness, aroma, and taste. This is the most common method used in Western Balkan countries (Rankovic *et al*., 2004; Teševic *et al*., 2005; Vulic *et al*., 2012; Matijaševic et al., 2013; Stoica *et al*., 2019).

The tasting panel consisted of a group of seven tasters, including three authorized evaluators and four unauthorized evaluators. The tasters were asked to evaluate the sensory attributes on a structured scale: color and clarity (maximum 1), distinctiveness (maximum 2), smell (maximum 6), and taste (maximum 10).

The evaluation was carried out in the tasting room of the Faculty of Horticulture of the University of Craiova. All conditions were ensured for a successful evaluation, including: space, air temperature of 22 °C, white light source, tasting utensils, etc. In order to obtain a uniform taste and smell, 30 ml samples of distillate were tasted at 12/16 °C in colorless, transparent, tulip-shaped glasses. The samples were coded and tested in a balanced order to eliminate the first-order transfer effect and evaluated on the same day. Since the institution lacks an ethics committee for the sensory evaluation of products, all participants provided written consent.

The neutral/semiaromatic grape brandy was evaluated before the Muscat-type aromatic grape brandy because of the stronger aroma of the Muscat brandy.

Between samples, water was provided for rinsing the mouth and pieces of bread for mitigating the previous taste.

2.7.Statistical analysis

Experimental data were subjected to One-way ANOVA analysis of variance, followed by the Tukey`s HSD test for $p< 0.05$ in Statgraphics Centurion 18 for Windows, to assess significant differences between the different type of brandy. All data were reported as mean ± standard deviation.

3.Results and discussions

3.1. Volatile compounds of brandy samples

The aromatic profile is a determining factor of the sensory quality of grape marc distillates and is essential for consumer perception. In grape marc distillates obtained from neutral/semi-aromatic and aromatic grape varieties, using combined gas chromatography–mass spectrometry methods (GC/MS), 29 volatile aromatic compounds were identified and quantified. These belong to several classes of chemical substances such as higher alcohols, esters, acids, aldehydes, acetals, and terpenes. Although studies have been conducted on the chemical composition of alcoholic beverages made from fruits and grape marc, only a few investigations have linked this composition to their sensory properties (Cortés, S. et al., 2009; Lukić, I. et al., 2012; Stoica, F. et al., 2019, 2022; Raicevic, D. et al., 2022).

3.1.1. Alcohols

The ethanol concentration (average alcoholic strength by volume) of brandy from semi-aromatic grapes reached 50.37% vol., while the alcoholic strength of brandy from neutral and aromatic grapes did not reach 50% vol. No statistically significant difference was found between the alcohol content of the analyzed brandies.

The methanol content in the analyzed samples ranged from 128.77 g/hL of 100% vol. alcohol to 148.09 g/hL in brandy from neutral/semi-aromatic grapes, and from 163.48 g/hL to 178.3 g/hL in brandy from aromatic grapes. The determined levels for all types of brandy were much lower than those stipulated in European legislation (Regulation (EU) 2019/787) and Romanian spirits law (2008, Ordinance no. 368/2008), where a methanol concentration below 1,000 g/hL of 100% vol. alcohol is indicated. Significant differences were observed only in two of the aromatic brandies (MA and MO) compared to the neutral or semi-aromatic ones (FR, FA, and S). All these values, within the legal limits, indicate that the raw material handling as well as the fermentation and distillation processes were appropriate [Silva et al., 2000; Soufleros *et al*., 2004; Stoica *et al*., 2019].

All methanol content values in the analyzed brandies fall within the same range (95.5–259 g/L) as reported for grape brandy (Rankovic *et al*., 2004; Cortés *et al.,* 2011; Lukic *et al.,* 2012; Milanov *et al*., 2014), but are lower than those reported for other fruit brandies (Silva *et al*., 2000; Soufleros *et al*., 2004, Geroyiannaki *et al*., 2007; Vulic *et al*., 2012; Arrieta-Garay *et al*., 2014).

Regarding higher alcohols, they were quantitatively the most abundant group of volatile compounds, conferring the specific aroma, taste, and basic character of alcoholic beverages (Cortés *et al.,* 2005; Christoph *et al*., 2007). Among all the higher alcohols analyzed (Table 1) in grape brandies, 2-phenylethanol and 1-hexanol had the highest contents.

Table 1. Concentration in alcohols in different types of grape brandies

Compound	FR	FA	S	TR	MA	MO
Ethanol. %vol	49.6±1.05	49.95±0.7	50.37±1.21	49.96±1.27	49.41±0.96	49.80±1.39
Methanol. g/hL of 100% vol. alcohol	132.59±7.46[c]	128.77±3.79[c]	148.09±4.44[bc]	163.48±6.91[ab]	171.37±6.92[a]	178.3±9.07[a]
Aroma compounds-high alcohol. mg/L						
1-hexanol	1.18±0.01[c]	1.12±0.04[c]	1.12±0.01[c]	3.23±0.09[a]	1.82±0.08[b]	3.07±0.09[a]
1-heptanol	0.22±0.03[bc]	0.11±0.02[c]	0.12±0.02[c]	0.81±0.07[a]	0.31±0.07[b]	0.15±0.02[c]
n-octanol	0.18±0.01[d]	0.23±0.01[c]	0.28±0.02[b]	0.36±0.02[a]	0.20±0.01[cd]	0.13±0.01[e]
n-decanol	0.12±0.01[b]	0.11±0.01[b]	0.10±0.01[b]	0.14±0.02[a]	0.08±0.004[c]	0.06±0.01[c]
Benzyl alcohol	n.d.	0.159±0.011	n.d.	n.d.	n.d.	n.d.
2 phenyl-ethyl alcohol	6.23±0.05[d]	5.85±0.07[e]	5.78±0.07[e]	7.19±0.04[a]	6.91±0.05[b]	6.38±0.03[c]
Total high alcohol	8.58±0.57[c]	7.57±0.06[d]	7.39±0.02[d]	11.81±0.29[a]	9.38±0.26[b]	9.79±0.13[b]

Note: Data are presented as mean±SD; n.d.= not detected; Different lowercase letters on the same row indicate significantly different means (HSD Tukey`s test at p<0.05) for the type of brandy from different grapevine varieties; Values without lowercase letters indicate that there were no significant differences between the analyzed variants.

A higher average content of 2-phenylethanol was detected in aromatic brandy samples (mg/L in TR) compared to all other brandies. In neutral/semi-aromatic grape brandies, the highest value was found in FR (6.23 mg/L). The results are consistent with other studies (Soufleros *et al*., 2004; Cortés *et al.,* 2011; Lukic *et al.,* 2011; Matijaševic *et al*., 2019).

For hexanol, significant differences were observed between brandies from aromatic grape varieties (TR, MA, MO) and those from neutral/semi-aromatic varieties; MA had a significantly lower content compared to TR and MO. A relatively higher content of 1-hexanol compared to the other brandies was found in the TR distillate (3.23 mg/L). The results agree with the findings of Cacho et al., 2013 and Raicevic D. et al., 2022. Compared to other fruit brandies, 1-hexanol was detected in lower concentrations in the analyzed samples (Lukic et al., 2011; Cortés et al., 2011; Matijaševic et al., 2019), which can be explained by the use of well-ripened grapes and milder pressing during grape processing, probably influencing the good quality of the brandy.

A higher average content of 1-heptanol was found in aromatic brandies compared to those from neutral/semi-aromatic grapes. Higher contents of 1-heptanol and 2-phenylethanol were recorded in the TR and MA aromatic brandies, with significant differences between the two varieties.

3.1.2. Esters

The total ester content in the analyzed distillates differs significantly depending on the grape variety from which the marc was obtained (Table 2); the highest total ester content values were recorded in aromatic grape varieties.

The results regarding the total volatile esters identified in this study show that their total content in brandies from aromatic grape varieties ranges from 19.9 mg/L in MA brandy (obtained from Muscat of Alexandria) to 23.87 mg/L in brandy obtained from Tămâioasă Românească (TR). Moreover, the total ester content is statistically significantly higher in brandies from aromatic grape varieties than in those from neutral/semi-aromatic varieties.

Table 2. Concentration in esters in different types of grape brandies

Compound, mg/L	FR	FA	S	TR	MA	MO
Ethyl butirate	0.193±0.002^{b}	0.146±0.001^{e}	0.142±0.002^{e}	0.233±0.003^{a}	0.17±0.002^{d}	0.187±0.002^{c}
Ethyl lactate	0.42±0.03^{d}	0.34±0.03^{e}	0.32±0.02^{e}	0.64±0.01^{b}	0.56±0.01^{c}	0.78±0.01^{a}
Isoamyl acetate	0.16±0.005^{d}	0.19±0.004^{c}	0.13±0.003^{e}	0.35±0.003^{a}	0.29±0.01^{b}	0.34±0.01^{a}
Ethyl succinate	2.55±0.12^{b}	2.52±0.09^{b}	2.33±0.06^{b}	2.83±0.10^{a}	2.05±0.06^{c}	2.09±0.06^{c}
Ethyl hexanoate	1.99±0.14^{cd}	1.94±0.07^{d}	1.62±0.09^{e}	3.69±0.12^{a}	2.97±0.10^{b}	2.25±0.06^{c}
Ethyl octanoate	2.03±0.09^{d}	1.90±0.07^{d}	2.01±0.08^{d}	5.72±0.06^{a}	5.34±0.07^{b}	4.83±0.07^{c}
Ethyl nonanoate	0.05±0.01^{b}	0.05±0.01^{b}	0.06±0.002^{b}	0.11±0.02^{a}	0.09±0.02^{ab}	0.12±0.03^{a}
Ethyl decanoate	6.45±0.08^{b}	4.90±0.09^{d}	4.98±0.10^{d}	7.18±0.07^{a}	5.73±0.09^{c}	7.21±0.08^{a}
Ethyl dodecanoate	2.79±0.06^{b}	2.83±0.05^{b}	2.73±0.06^{b}	3.11±0.02^{a}	2.76±0.05^{b}	3.00±0.10^{a}
Total esters	16.63±0.26^{d}	14.82±0.14^{e}	14.33±0.13^{f}	23.87±0.09^{a}	19.9±0.12^{c}	20.81±0.08^{b}

Note: Data are presented as mean±SD; Different lowercase letters on the same row indicate significantly different means (HSD Tukey`s test at p<0.05) for the type of brandy from different grapevine varieties

The most abundant esters in all samples are the ethyl esters of medium-chain fatty acids. Among these, the highest values are for ethyl decanoate (4.90 mg/L - FA and 7.21 mg/L - MO), followed by ethyl octanoate (1.90 mg/L - FA and 5.72 mg/L - TR), ethyl dodecanoate (2.73 mg/L - S and 3.11 mg/L - TR), and ethyl hexanoate (1.62 mg/L - S and 3.69 mg/L - TR). Ethyl decanoate, ethyl hexanoate, and ethyl octanoate provide a pleasant fresh tropical fruit aroma, while ethyl dodecanoate gives a pear aroma and a characteristic fruity aroma (Matias-Guiua *et al*., 2018; Matijaševic *et al*., 2019).

The distillate obtained from MO shows comparative/similar values to those recorded for distillates from the TR variety regarding the content of isoamyl acetate, ethyl nonanoate, ethyl decanoate, and ethyl dodecanoate, with no statistically significant differences between the two brandies in this regard (Table 2).

Except for the ethyl lactate content, the distillate obtained from TR stands out from the other distillates in terms of the analyzed ester contents.

The distillate obtained from Fetească Regală (FR) marc is also noteworthy for its higher ethyl butyrate content compared to the aromatic varieties MA and MO, with statistically significant differences.

Distillates obtained from FR, FA, and S do not show significant differences regarding the contents of ethyl succinate, ethyl nonanoate, and ethyl dodecanoate (Table 2).

Regarding the ethyl dodecanoate content, no statistically significant differences were observed between distillates obtained from the marc of neutral and semi-aromatic varieties (FR, FA, S) and the distillate obtained from MA. The ethyl dodecanoate content is significantly higher in distillates from TR and MO compared to distillates obtained from marc of neutral and aromatic varieties.

3.1.3. Terpenes

The values of the analyzed terpenes in the examined brandies are presented in Table 3. The results of the investigation showed that the total terpene content is significantly higher in brandies from aromatic grape varieties (1.66 mg/L - MA and 2.49 mg/L - TR) compared to brandies from neutral/semi-aromatic varieties (0.42 mg/L - FR and 0.91 mg/L - S). These results are supported by other studies (Stoica & Heroiu, 2003; Stoica *et al*., 2003; Doneva-Šapceska *et al*., 2006).

Linalool is the main component in all examined samples, in accordance with other research findings (Cacho *et al.,* 2013). It is followed by alpha-terpineol, geraniol, and citronellol. The linalool content, which contributes significantly to the aroma of roses, anise seeds, grapefruit, lime, and citrus, is significantly higher in aromatic brandies (0.68 mg/L - TR and 1.15 mg/L - MO) compared to brandies from neutral/semi-aromatic varieties (0.25 mg/L - FR and 0.34 mg/L - S). This means that the average linalool concentration in aromatic brandies is well above the perception threshold (0.05 mg/L) (Diéguez *et al*., 2003; Raicevic *et al*., 2022). The obtained values are in agreement with other studies (Matijašević *et al.,* 2019; Stoica *et al*., 2022).

A particular feature is observed with alpha-terpineol, which, although present in all distillates, reaches its highest values in Tămâioasă Românească (TR) brandy. Moreover, in aromatic varieties, for the two Muscats – Muscat of Alexandria and Muscat Ottonel - the dominant terpene is linalool, whereas in Tămâioasă Românească and Sauvignon (in much lower amounts compared to TR), alpha-terpineol is dominant. Alpha-terpineol provides floral aromas, as well as notes of iris and pine forest.

Table 3. Concentrations of the main terpenes in different types of grape brandies

Compund, mg/L	FR	FA	S	TR	MA	MO
α-terpineol	0.12±0.01[d]	0.24±0.01[d]	0.40±0.02[c]	1.19±0.10[a]	0.52±0.03[c]	0.67±0.07[b]
Linalool	0.25±0.01[e]	0.28±0.004[e]	0.34±0.01[d]	0.68±0.01[c]	0.89±0.03[b]	1.15±0.01[a]
Geraniol	0.021±0.003[e]	0.022±0.004[e]	0.070±0.002[d]	0.37±0.003[a]	0.15±0.01[c]	0.21±0.001[b]
Citronellol	0.030±0.002[d]	0.051±0.001[d]	0.091±0.002[c]	0.25±0.01[a]	0.11±0.01[c]	0.15±0.01[b]
Total terpenes	0.42±0.02[e]	0.59±0.01[e]	0.91±0.02[d]	2.49±0.12[a]	1.67±0.06[c]	2.19±0.09[b]

Note: Data are presented as mean±SD; Different lowercase letters on the same row indicate significantly different means (HSD Tukey`s test at $p<0.05$) for the type of brandy from different grapevine varieties

The citronellol content, which together with linalool is one of the most important terpenic compounds, is below 0.10 mg/L in brandies from neutral/semi-aromatic varieties. In brandies from aromatic varieties, the citronellol content ranges between 0.11 mg/L - MA and 0.24 mg/L - TR. These values are in line with those reported by other researchers (Diéguez *et al*., 2003; Cacho *et al*., 2013) (0.07- 43.53 mg/L) and are significantly above the perception threshold (0.018 mg/L).

The geraniol content, with values below 0.37 mg/L in aromatic brandies, will also contribute to floral aromas, especially rose, as its concentration is above the perception threshold (0.13 mg/L).

The presented results confirm the conclusions of (Tsakiris *et al*., 2014; Matijašević *et al*., 2019; Stoica *et al*., 2019, 2022; Raicevic *et al*., 2022), according to which the aroma of brandy from aromatic grapes is directly related to various monoterpenes, such as alpha-terpineol, linalool, nerol, geraniol, and citronellol.

3.1.4. Volatile acids and aldehydes

The values of total fatty acids identified (Table 4) fluctuate within similar ranges in the distillates obtained from aromatic grape varieties (3.54 mg/L - MA and 5.12 mg/L - TR) and those from neutral/semi-aromatic varieties (3.74 mg/L - FR and 5.43 mg/L - S). Regarding medium-chain fatty acids, decanoic, octanoic, dodecanoic, and hexanoic acids were identified in all brandy samples.

Medium-chain fatty acids usually do not have a significant effect on the aroma of distillates due to their relatively high odor perception thresholds, determined at 8 mg/L for decanoic acid and 15 mg/L for octanoic acid (Lukić *et al*., 2011).

In our reserche decanoic acid had the highest values among all samples of distillates (neutral/semi-aromatic and aromatic), compared to the other fatty acids. It is followed by octanoic acid, dodecanoic acid, and hexanoic acid, values that are in agreement with findings from other studies (Christoph *et al.*, 2007; Lukić *et al*., 2011, 2012; Matijašević. *et al*., 2019).

The distillate obtained from *Tămâioasa Românească* recorded the highest contents of octanoic acid and dodecanoic acid, with significant differences compared to the distillates produced both from aromatic varieties and from neutral/semi-aromatic varieties. The distillate obtained from *Sauvignon* stands out through significantly higher contents of decanoic acid and hexadecanoic acid compared to the other distillates..

Regarding long-chain fatty acids, which together with glycerol contribute to the aroma and body of the distillate through oiliness and viscosity, hexadecanoic acid (palmitic) shows the highest average value in all our samples (above 0.40 mg/L), which is consistent with other similar research (Lukić *et al*., 2011, 2012; Matijašević *et al.,* 2019; Raicevic *et al*., 2022).

The concentrations of aldehydes in the distillates obtained from neutral/semi-aromatic grape varieties and from aromatic varieties are presented in Table 4. The total content of identified carbonyl compounds is not significantly different between the brandies obtained from aromatic varieties and those obtained from neutral/semi-aromatic varieties.

The main carbonyl compound in distillates, acetaldehyde, is a direct product of alcoholic fermentation and usually represents about 90% of the total aldehyde content (Silva *et al*., 2000; Spaho, 2017).

No significant differences were found between the acetaldehyde contents of the distillates from neutral/semi-aromatic varieties (195.1 mg/L of 100% vol. alcohol - FA and 220.1 mg/L of 100% vol. alcohol - S) and those from aromatic varieties (198.3 mg/L of 100% vol. alcohol - MA and 229.5 mg/L of 100% vol. alcohol - MO). These results are in agreement with those described in the literature (Hernández-Gómez *et al*., 2005; Lukić. *et al*., 2011, 2012; Raicevic *et al*., 2022).

The values determined in this study are significantly lower compared to the official limits for fruit distillates adopted by the European Council (Regulation (EU) 2019/787).

A significantly higher concentration of benzaldehyde was found in the brandy obtained from the aromatically neutral variety (0.27 mg/L-FA), while the other distillates did not differ in terms of their benzaldehyde content. The benzaldehyde content, which contributes to bitter almond, marzipan, and cherry aromas, identified in our study in the grape pomace distillates, is consistent with data reported in the literature (Christoph *et al*., 2007; Cortés *et al.,* 2009; Bortoletto, A.M. *et al*., 2016). Benzaldehyde is not desirable in large quantities, but when present in small amounts, it contributes to the complexity of the aroma.

Table 4. Concentrations of volatile acids and aldehydes in different types of grape brandies

Compund, mg/L	FR	FA	S	TR	MA	MO
			Volatile acids			
Hexanoic acid	0.51±0.02^{c}	0.53±0.02^{c}	0.59±0.05^{bc}	0.77±0.06^{a}	0.58±0.05^{c}	0.69±0.04^{ab}
Octanoic acid	0.76±0.02^{e}	0.82±0.01^{cd}	0.88±0.01^{c}	1.02±0.04^{a}	0.68±0.01^{f}	0.94±0.02^{b}
Decanoic acid	1.61±0.02^{e}	1.94±0.02^{cd}	2.80±0.02^{a}	1.89±0.01^{d}	1.30±0.03^{f}	2.22±0.07^{b}
Dodecanoic acid	0.45±0.03^{e}	0.51±0.02^{d}	0.66±0.01^{c}	0.97±0.02^{a}	0.62±0.02^{c}	0.74±0.01^{b}
Hexadecanoic acid	0.41±0.01^{c}	0.41±0.01^{c}	0.51±0.02^{a}	0.45±0.01^{b}	0.43±0.01^{bc}	0.46±0.02^{b}
Total acids	3.74±0.07^{d}	4.22±0.03^{c}	5.43±0.05^{a}	5.12±0.03^{b}	3.60±0.08^{d}	5.05±0.06^{b}
			Aldehydes			
Acetaldehyde	212.00±12.49	197.73±10,74	220.10±9.90	218.30±7.05	198.33±11.95	226.20±13.35
Benzaldehyde	0.19±0.01^{b}	0.27±0.02^{a}	0.21±0.04^{b}	0.20±0.02^{b}	0.18±0.01^{b}	0.18±0.01^{b}
Furfural	0.15±0.01^{c}	0.30±0.03^{b}	0.52±0.04^{a}	0.31±0.02^{b}	0.47±0.05^{a}	0.18±0.02^{c}
Total aldehydes	212.30±12.41	198.30±10.76	220.79±9.89	218.80±7.09	198.95±12.01	226.56±13.35

Note: Data are presented as mean±SD; Different lowercase letters on the same row indicate significantly different means (HSD Tukey`s test at $p<0.05$) for the type of brandy from different grapevine varieties

The furfural content in the analyzed distillates ranges between 0.15 and 0.52 mg/L, being significantly lower than the perception threshold of this compound, which is 150 mg/L. Significantly lower amounts of furfural were recorded in the distillates from FR and MO.

Furfural is a normal constituent of fruits and can be used as an indicator of the natural origin of the distillate; however, increased concentrations may contribute to the appearance of "roasted" notes in the spirit (Tsakiris *et al*., 2016; Spaho, 2017). These results are in agreement with those reported in the literature (Ranković *et al*., 2004; Tešević *et al*., 2005; Hernández-Gómez *et al*., 2005; Lukić *et al*., 2011; Cacho. *et al*., 2013; Cortés *et al*., 2011; Arrieta-Garay *et al*., 2014; Raicevic *et al*., 2022).

3.2. Sensory analysis

The sensory characteristics of the examined brandies are presented in Table 5.

All distillates obtained from neutral/semi-aromatic or aromatic grape pomace are colorless and clear, exhibiting excellent clarity and being remarkable for the characteristic aromas and flavors of these types of beverages. In the sensory evaluation, all samples received the maximum score (1.0) for color and clarity, reflecting the superior visual quality of the distillates.

From the perspective of *Distintion,* significant differences were recorded only for the distillate obtained from TR (average score 1.90).

Brandy from aromatic grape varieties stood out with a relative higher average score for aroma (5.78), due to the distinctive character of Muscat-type grape varieties, which impart a pronounced and intensely fruity aroma to the distillate, appreciated by consumers for its olfactory finesse and expressiveness, however, the differences were not statistically significant between the brandies from neutral/semi-aromatic varieties and those from aromatic varieties (Table 5).

Table 5. Sensory analyses of grape brandies (average values _ STD)

Type of grape brandy	Colour (max 1)	Clarity (max 1)	Distintion (max 2)	Odour (max 6)	Taste (max 10)	Total (max 20)
FR	1.0	1.0	1.81±0.04^{b}	5.59±0.17	9.23±0.13^{b}	18.62±0.22^{c}
FA	1.0	1.0	1.83±0.03^{ab}	5.63±0.12	9.37±0.18^{b}	18.83±0.11^{bc}
S	1.0	1.0	1.88±0.04^{ab}	5.70±0.10	9.48±0.07^{ab}	19.07±0.17^{ab}
Averages	1.0	1.0	1.84±0.03	5.64±0.10	9.36±0.09^{B}	18.84±0.02^{B}
TR	1.0	1.0	1.90±0.03^{a}	5.81±0.10	9.77±0.08^{a}	19.49±0.19^{a}
MA	1.0	1.0	1.84±0.02^{ab}	5.75±0.08	9.58±0.11^{ab}	19.18±0.06^{ab}
MO	1.0	1.0	1.86±0.03^{ab}	5.79±0.10	9.68±0.07^{ab}	19.33±0.16^{a}
Averages	1.0	1.0	1.87±0.02	5.78±0.08	9.68±0.07^{A}	19.32±0.13^{A}

Note: Data are presented as mean±SD; Different superscript small letters on the same column indicate significantly different means (HSD Tukey`s test at $p<0.05$) for the type of brandy from different grapevine varieties; different superscript capital letters indicate significantly different means (HSD Tukey`s test at $p<0.05$) for the type of brandy (neutral/semi aromatic and aromatic).

Regarding taste, both types of brandies-those from neutral/semi-aromatic grapes and aromatic grapes-achieved a high average score above 9. The distillates from aromatic grapes were better appreciated, achieving an average score of 9.68, significantly higher compared to the distillates from neutral/semi-aromatic varieties. The distillate from TR stands out in terms of taste, with an average score of 9.77 points.

The average total score obtained by the brandies from aromatic varieties (19.32) was significantly higher compared to the brandies from neutral/semi-aromatic varieties (18.84), most likely due to the greater taste complexity of the distillates from aromatic varieties. It is also noteworthy that there were no significant differences between the average total scores of the distillates from aromatic varieties and the distillate from Sauvignon, indicating similar olfactory–taste qualities of these distillates, as perceived and appreciated by consumers.

These results demonstrate that Romanian brandies produced from grape pomace have an exceptional sensory profile, characterized by an intense and pleasant aroma, high drinkability, and a perfectly balanced harmony between flavor and taste. All these qualities make both types of analyzed brandies-particularly those from Tămâioasă Românească and Muscat Ottonel-true unique brands, representative of the tradition and specific characteristics of their origin area in southwestern of Romania, in Drăgășani vineyard region.

4.Conclusions

All the analyzed grape marc distillates were produced using the same technology, the difference between them being determined by the grape variety used - three neutral/semi-aromatic grape varieties (Fetească Regală - FR, Fetească Albă - FA, and Sauvignon - S) and three aromatic varieties (Tămâioasă Românească - TR, Muscat of Alexandria - MA, and Muscat Ottonel - MO).

The content of volatile aromatic compounds (higher alcohols, volatile esters, and terpenes) was higher in the distillates obtained from aromatic varieties than in those from neutral/semi-aromatic grape varieties.

A particular characteristic is shown by α-terpineol which, although present in each variety and implicitly in each distillate, reaches the highest levels in the Tămâioasă Românească (TR) distillate. Moreover, in aromatic varieties it can be observed that in the two Muscats - Muscat of Alexandria and Muscat Ottonel - the dominant terpene is linalool, whereas in Tămâioasă Românească the dominant compound is α-terpineol.

The results of the study showed that the grape variety used for producing the distillates influences certain sensory characteristics (smell and taste).

Distillates from aromatic varieties stood out due to a higher average score for aroma, owing to the distinctive character of Muscat-type grape varieties, which impart a pronounced and intensely fruity aroma to the distillate - highly appreciated by consumers for its finesse and expressive olfactory profile.

Sensory analyses showed that all distillates belong to the category of high-quality spirits and exhibited uniform quality. Both types of distillates were distinguished by excellent drinkability, full and balanced flavor, and overall harmony in the sensory profile.

These results indicate a high quality (volatile aromatic compounds and sensory properties) of the analyzed Romanian distillates, and the aromatic ones (TR and MO) could be certified as a new traditional product—a distinctive brand in Romania, comparable to Grappa from Italy.

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