Food and Bioprocess Technology
https://doi.org/10.1007/s11947-024-03573-z

RESEARCH

The Interplay of Bottle Storage and Wood Ageing Technology: Volatile and Sensory Profiles of Wine Spirits Aged with Chestnut Wood

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Received: 6 June 2024 / Accepted: 19 August 2024

Abstract

Wine spirits are typically aged in wooden barrels. Recently, alternative ageing technologies, such as those using wood fragments in wine spirits stored in stainless steel tanks, have been investigated. However, a significant lack of information regarding the potential evolution of these beverages after bottling still remains. This study assessed the 12-month evolution of aroma in bottled wine spirits aged with chestnut wood using different technologies, including fragment application with several micro-oxygenation strategies and barrels (traditional). Chemical analysis using GC-FID and GC–MS methods, along with sensory analysis, was conducted on all sampled aged wine spirits. Significant changes in volatile compounds were detected over time, including volatile phenols, acids, and esters. Multivariate data analysis distinguished traditional and alternative aged samples, with slight sample discrimination based on bottle storage. Regarding the sensory results, a significant effect of the time in bottle in several sensory attributes was found, while the ageing technologies mainly affected the gustatory attributes. The tasters were also asked to rate the overall quality of the samples, which seems to be favoured by the time in the bottle. This initial assessment of the impact of 1 year of glass bottle storage on the volatile and sensory composition of aged wine spirits highlights that this stage must be considered as an additional technological factor in their production process. However, the differences induced by the wood ageing technologies applied remained evident after 1 year of glass bottle storage.

Keywords Wine spirit · Ageing technology · Bottle storage · Volatile compounds · Sensory profile · Chestnut

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Introduction

Wine spirits are typically aged in the presence of wood for a lengthy period of time. After this stage, these alcoholic beverages undergo finishing and bottling operations. Traditional ageing occurs in wooden barrels, but the potentialities of alternative ageing technologies have been explored. For this purpose, different approaches have been developed, some focusing on the wood and others on the process. One approach is based on using wood fragments or extracts in stainless steel tanks to shorten the ageing period, providing economic and environmental benefits. The use of staves or staves combined with micro-oxygenation applied to the ageing of wine spirits kept in stainless steel tanks (Canas et al., 2022) showed promising results in terms of sensory quality and volatile profile. Another alternative approach involves applying physical treatments to barrels to speed up the ageing process. Zhang et al., (2013a, 2013b) investigated the effect of an electric field (EF) applied to small oak barrels (2 L and 5 L) on the chemical composition of the wine spirit. This study revealed that the EF treatment applied increased the extraction of several phenolic compounds from the wood; after 14 months of EF treatment, the levels of these compounds in the wine spirits were higher than in those aged in wooden barrels without EF treatment.

The physicochemical and sensory changes in wine spirits that occur during the ageing process, whether through traditional or alternative technologies, have been extensively studied (Canas et al., 2022). It is well-established that, despite the influence of distillate quality and distillation conditions (Guerrero-Chanivet et al., 2023; Matias-Guiu et al., 2018), the botanical species of the wood, the heat treatment in cooperage process, and ageing time are the most determining factors regarding the composition and quality of aged wine spirits (Canas et al., 2022; Tsakiris et al., 2014). Direct extraction of wood constituents, breakdown of wood biopolymers (lignin, hemicelluloses, and cellulose), chemical reactions involving wood extractable compounds, distillate compounds, or both, evaporation of volatile compounds, concentration of volatile and non-volatile compounds, and formation of stable molecular aggregates between water and ethanol are well-documented reactions and mechanisms that occur during ageing in a complex and oxidative environment (Nishimura & Matsuyama, 1989). As a result, several volatile and non-volatile compounds enrich the wine spirits (Caldeira et al., 2021; Thibaud et al., 2020; Winstel et al., 2021). Such modifications lead to an increase in the sensory quality of these beverages over time (Caldeira et al., 2021; Granja-Soares et al., 2020).

Despite the extensive knowledge of the aforementioned changes, information regarding potential post-bottling modifications in beverages stored in glass bottles is scarce, both for wine spirits and other wood aged or unaged distilled beverages. Among the few published studies, the following are noteworthy: Aguiar et al. (2021) observed significant changes in the physicochemical composition of bottled Madeira Rum after 14 months of underwater storage; Matias-Guiu et al. (2020) monitored unaged fruit spirits bottled under various storage conditions (pH, temperature, and sunlight exposure) for over 7 months, highlighting temperature as the primary factor influencing volatile compound levels; Flouros et al. (2003) examined the major volatile compounds in bottled Tsipouro samples (grape marc spirit produced in Greece) stored at room temperature for 12 months in different containers (PET, PVC, and glass bottles) and found a significant increase only in furfural. In the latter work, some volatile compounds showed slight increase, while others remained stable (acetaldehyde and 1-propanol) or slightly decreased (2-methyl-1-propanol), with no significant influence observed based on container type. Furthermore, Cigic and Zupancic-Kralj (1999) observed the influence of glass colour on flavour quality in Bartlett pear spirit, finding lower quality in spirits stored in colourless bottles compared to green bottles.

In the case of aged wine spirits, Belchior and San-Romão (1982) noticed that aged wine spirits preserved in green glass bottles and stored in darkness had more appealing sensory features than those stored in transparent glass bottles exposed to light. Another study (Belchior et al., 1990) was focused on the bottle storage conditions, including different types of cork stoppers and bottle positions (laying down or standing). It was found that storing the bottle upright allowed for greater oxygen intake, suggesting that this position may not be ideal for long-term storage. However, Cognac literature advises keeping bottles upright at room temperature, in moderate environments shielded from light to prevent cork-wine spirit contact (Parvulesco, 2002). More recently, Oliveira-Alves et al. (2022) monitored the non-volatile composition (total phenolic index and low molecular weight compounds) and the antioxidant activity evolution of aged wine spirits from different alternative ageing processes and found that differences between ageing modalities were mostly retained during bottle storage, with only minor modifications detected. Furthermore, Valaer and Frazier (1936) conducted a study on whiskey samples throughout a 4-year bottle storage period, using global chemical methods to quantify aldehydes, esters, and acids, which revealed a decrease in acids and a tendency for ester increase.

Given the published results on bottle storage, it is hypothesized that there may be changes in the volatile composition and sensory properties of the aged wine spirits during this stage. Thus, this study aims to evaluate whether these changes of spirits aged with chestnut wood, resulting from different ageing technologies (alternative using fragments under different micro-oxygenation conditions and traditional using 250 L wooden barrels), persist after 1 year of storage in glass bottles. The assessment was carried out using complementary analytical approaches, including the quantification of volatile compounds by gas chromatography and the evaluation of the sensory properties of the wine spirits.

Materials and Methods

Experimental Design

Wood Ageing Modalities

The wood ageing experiment consisted of five ageing modalities: the traditional technology using 250 L chestnut wooden barrels (B) and alternative ageing technology using wood staves combined with micro-oxygenation at three different levels (O15, O30, and O60). Additionally, a fifth modality combining wood staves with nitrogen (N) was performed in order to decrease the dissolved oxygen as much as possible. These ageing conditions (Fig. 1) were maintained for one year. Further details about each modality are provided below:

I) B—250 L chestnut wooden barrel
II) O15—50 L glass demijohns with chestnut staves and micro-oxygenation (flow rate of 2 mL/L/month for the first 15 days, followed by 0.6 mL/L/month until 365 days)
III) O30—50 L glass demijohns with chestnut staves and micro-oxygenation (flow rate of 2 mL/L/month for the first 30 days, followed by 0.6 mL/L/month until 365 days)
IV) O60—50 L glass demijohns with chestnut staves and micro-oxygenation (flow rate of 2 mL/L/month for the first 60 days, followed by 0.6 mL/L/month until 365 days)
V) N—50 L demijohns with chestnut staves and nitrogen application (flow rate of 20 mL/L/month for 365 days)

These flow rates were selected based on the results of a previous study (Canas et al., 2019), considering the subsequent principles: guaranteeing adequate oxygen supply but avoiding excessive oxidation, shortening the ageing period, and decreasing the costs associated with ageing to attain a sustainable ageing technology.

Two replicates (1 and 2) of each modality were used, resulting in ten containers filled with the same wine distillate from Adega Cooperativa da Lourinhã (alcoholic strength 78.3% vol.; pH 5.33; total acidity, as acetic acid 0.12 g/L of absolute ethanol). All experimental units were placed in the cellar of Adega Cooperativa da Lourinhã (Lourinhã, Portugal).

Portuguese chestnut wood staves (*Castanea sativa* Mill.) were used in all modalities. These staves, with 50.0 cm length, 5.0 cm width, and 1.8 cm thickness, were supplied by J. M. Gonçalves cooperage (Palaçoulo, Portugal). A medium plus toasting level was applied to the staves, involving 90 min of toasting at an average temperature of 240 °C, with a toasting thickness of 1.8 cm. The staves were heated in an industrial oven, and the barrels were toasted over a fire of wood scraps to ensure a consistent level of toasting across all units.

Bottle Storage Experiment

After 1 year of wood ageing, the wine spirits from the ten experimental units were bottled on the same day (Fig. 1). The entire set of amber glass bottles (0.75 L) is from each ageing wood modality, comprising 32 bottles (replicates

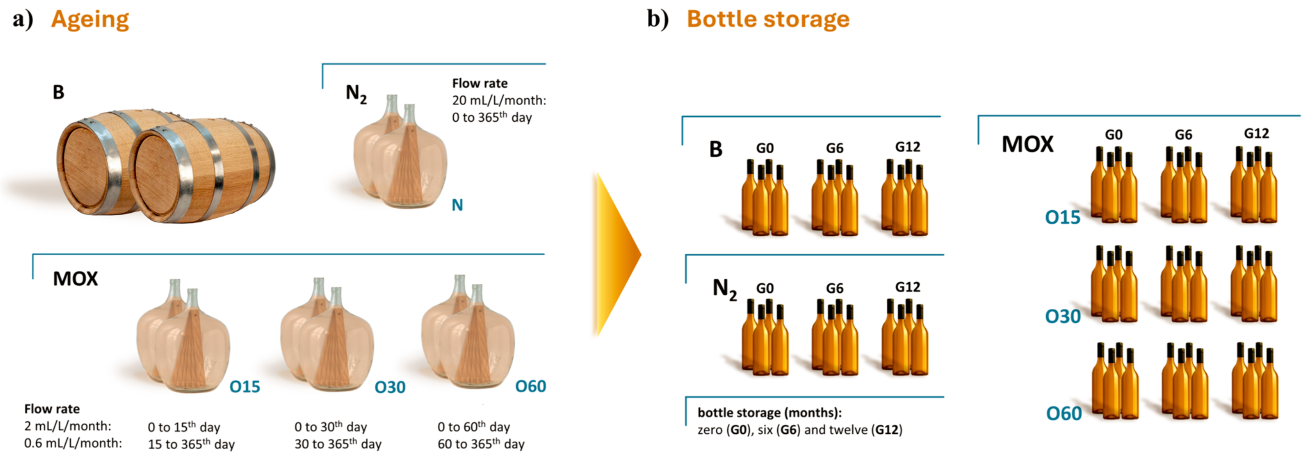


Fig. 1 Illustration of the experimental design for investigating the bottle storage of aged wine spirits from different ageing technologies

of bottle storage). All bottles were sealed with natural cork stoppers and had an average headspace of 9.8 mL. The bottles were stored upright in the cellar of INIAV – Polo de Inovação de Dois Portos (Portugal) in darkness at 19 °C and 80% humidity, for 1 year. All modalities of the bottle storage experiment were sampled at three time points: initially (G0), after 6 months (G6), and after 12 months (G12), to perform volatile chemical quantification and quantitative descriptive sensory analysis. A total of 60 samples were analysed, with 20 samples at each sampling point (5 experimental ageing modalities × 4 replicates).

Reagents

Pure oxygen (X50S Food) and nitrogen (X50S Food), used in the wood ageing experiment, were supplied by Gasin, Portugal. For volatile analyses, anhydrous sodium sulphate and ethanol were supplied by Merck (Darmstadt, Germany); dichloromethane was obtained from Honeywell Riedel-de Haën (Steinheim, Germany), and silanized glass wool was provided by Supelco (Steinheim, Germany). Ultrapure water was obtained using an arium®comfort I equipment from Sartorius Lab Instruments, Germany. GC-FID and GC–MS standards were as follows: acetic acid was purchased from Riedel-de-Haen (Seelze, Germany); ethyl acetate, ethyl butanoate, and ethyl octanoate from Merck (Darmstadt, Germany); isobutyl acetate, ethyl 2-methylbutyrate, ethyl hexanoate, *trans*-2-hexen-1-ol, linalool, butanoic acid, 3-methyl butanoic acid, hexanoic acid, 2-phenylethanol, 3,4-dimethylphenol (internal standard, IS), dodecanoic acid, 4-hydroxy-3-methoxybenzaldehyde (vanillin), and acetaldehyde were purchased from Fluka (Buchs, Switzerland); ethyl isobutyrate, ethyl 3-methylbutyrate, isoamyl acetate, 5-methyl-2-hexanol (IS), 2-methyl-1-butanol, 4-methyl-2-pentanol (IS), eugenol, 4-methylguaiacol, 4-ethylguaiacol, 4-methylsyringol, 4-allyl-syringol, and acetovanillone were purchased from Aldrich (Steinheim, Germany); syringol, 3-methyl-1-butanol, and 2-methyl-1-propanol from Sigma-Aldrich (Steinheim, Germany); malic acid diethyl ester was obtained from TCI (Zwijndrecht, Belgium).

Volatile Compound Analysis

The analysis of volatile composition of the aged wine spirits was focused on several compounds previously identified as odorants through olfactometry analysis (Caldeira et al., 2008), including major and minor volatile compounds. Additionally, the contents of acetaldehyde and ethyl acetate were determined.

Quantification of Higher Alcohols, Acetaldehyde, and Ethyl Acetate

The major volatile compounds, encompassing the higher alcohols (2-methyl-1-propanol, 2 methyl-1-butanol + 3-methyl-1-butanol), ethyl acetate, and acetaldehyde, were quantified by applying GC-FID to the distillate, prepared according to the official method (OIV, 2019), and using the conditions of a validated method described in a previous work (Granja-Soares et al., 2020). For quantification, the response factor between the internal standard and the standard analytes was determined based on the analysis of hydroalcoholic solutions of standards under the same chromatographic conditions.

Quantification *of Minor* Volatile Compounds

The analysis of minor volatile (*trans*-2-hexen-1-ol, linalool, 2-phenylethanol, butanoic acid, hexanoic acid, isovaleric acid, dodecanoic acid, ethyl isobutyrate, isobutyl acetate, ethyl butyrate, ethyl isovalerate, isoamyl acetate, ethyl hexanoate, ethyl octanoate, diethyl malate, acetic acid, guaiacol, 4-methyl-guaiacol, eugenol, syringol, 4-methyl-syringol, 4-allyl-syringol, vanillin, and acetovanillone) compounds followed the validated methodology (Caldeira et al., 2004) with the modifications introduced by Caldeira et al. (2010). Their quantification was performed based on calibration curves established by analysing hydroalcoholic solutions of standards under similar conditions.

Identification of Volatile Compounds by GC/MS

The identification of the analysed compounds of the distillates and of the extract samples at GC-FID was confirmed by using a GC mass spectrometer (Magnum, Finnigan Mat, San Jose CA) equipped with a polyethylene glycol fused silica capillary column (30 m length, 0.25 mm internal diameter, and 0.25 µm film thickness; HP-INNOWax of J&W Scientific Folsom CA, USA). The injector, operating in split mode with a 1:60 split ratio, and transfer line were set to 250 °C. Helium was used as the carrier gas at a pressure of 83 kPa, and the oven operated with a temperature program of 3.5 °C/min from 35 °C (held isothermally for 6 min) to 55 °C, followed by 7.5 °C/min to 130 °C, and then 5 °C/min to 210 °C, maintaining this temperature for 30 min. Manual injection of the samples was performed with a volume of 0.4 µL. Mass spectra were acquired under full scan at electron impact mode at 70 eV, with the *m/z* range of 40–340. Compound identification was achieved by comparing all mass spectra obtained with those from the NIST data system library. Additionally, identification was confirmed by comparing GC retention times and mass spectra with those of pure standard compounds.

Sensory Analysis

The aged wine spirit samples, diluted to an alcoholic strength of 40% vol., were evaluated by a tasting panel consisting of eight trained tasters. These tasters assessed 16 orthonasal aroma attributes (alcohol, fruity, vanilla, wood, rancid, spicy, caramel, toasted, dried fruits, smoke, coffee, sweet, green, glue, caoutchouc, and tails) and 12 gustatory attributes (sweetness, smooth, burning, astringency, roughness, bitter, body, unctuous, flavour evolution, flavour complexity, retronasal aroma, and flavour persistency) using a structured scale ranging from 0 (no perception) to 5 (strongest perception). These attributes were previously generated by the taster group as described in a previous work (Granja-Soares et al., 2020). All tasters provided informed consent, signifying their voluntary involvement in the sensory sessions conducted within this study. The sensory assessments were performed, at each sampling point, in accordance to the ethical guidelines ratified by the INIAV executive board.

At each sampling point, 24 samples, randomly distributed across three sensory sessions of eight samples each, were assessed by the tasters. This sample set included 20 samples from each sampling point (5 experimental ageing modalities × 4 replicates) and four repetitions included in each session to assess the reliability of the panel, as previously described (Granja-Soares et al., 2020). The sensory sessions were conducted in the morning, between 10:00 a.m. and 12:00 a.m., at the tasting room of the INIAV – Polo de Inovação de Dois Portos, Portugal. The tasting room was equipped with individual white booths for each taster. Additionally, a water glass was provided in each booth for palate cleansing between samples. Standard tasting glasses (ISO 3591, 1977) containing 30 mL of aged wine spirit, coded with three random digits, were presented to the tasters in a balanced order to eliminate first-order carryover effects (Macfie et al., 1989). The taster panel assessed the intensity of all attributes and also rated the overall quality of the aged spirits on a scale from 0 to 20. The sensory data were collected using the Tastel software (ABT Informatique, Rouvroy-sur-Marne, France).

Statistical Analysis

Data obtained from both sensory and chemical analyses at each sampling time during the bottle storage underwent a two-way analysis of variance (ANOVA) to examine the influence of two factors: the first factor was the ageing technology (traditional and alternative), and the second factor was the storage time in the bottle. The Cochran test was applied to determine variance homogeneity, and when statistically significant effects were detected ($p < 0.05$), the mean comparison Fisher test was used. Additionally, the results were evaluated using multidimensional analysis, specifically heatmap clustering. This visualization technique allows identifying patterns and relationships within large datasets by grouping similar data points into clusters and representing them with colour gradients. Such analysis facilitates the identification of trends and correlations, thereby assisting in data interpretation. The calculations were performed using STATISTICA/vs7 from StatSoft, Inc. (2004) and Microsoft Excel 2019 MSO (16.0.10380.20037).

Results and Discussion

Effect of Bottle Storage on the Volatile Composition of Aged Wine Spirits

The ANOVA results for volatile compounds of the aged wine spirits during bottle storage are presented in Table 1 and Online Resource 1 (Table OR1a and Table OR1b). The statistical analysis revealed a significant effect of bottle storage on the concentrations of 17 volatile compounds, while 20 compounds were significantly influenced by the ageing technology. Furthermore, an interaction effect between the two factors was observed for three compounds. In terms of bottle storage, the majority of compounds, including ethyl isobutyrate, ethyl isovalerate, ethyl octanoate, diethyl malate, butanoic acid, 4-methylguaiacol, eugenol, acetaldehyde, and ethyl acetate, showed an increasing trend over time. Conversely, the amounts of isobutyl acetate, isoamyl acetate, ethyl hexanoate, hexanoic acid, isovaleric acid, dodecanoic acid, and 4-methylsyringol decreased over time (Fig. 2 and Table OR1a). These findings indicate ongoing reactions within the bottle, resulting in changes in volatile compound contents over time. Interestingly, the majority of the identified volatile compounds that tend to increase in concentration over the bottle storage period are ethyl esters, which have a longer alkyl chain on the carbonyl side. The differences in reactivity of ethyl esters, with respect to the alkyl chain on the carbonyl side, are mostly due to electronic and steric factors. Bulkier and more electron-donating alkyl groups reduce the ester's reactivity by lowering the electrophilicity of the carbonyl carbon and increasing steric hindrance. Since the methyl or ethyl substituent groups are small, with weaker inductive donating effect and less steric hindrance, methyl and ethyl acetates are fairly reactive, which may justify its consumption throughout the bottling time (Table 1 and Fig. 2).

Most of the analysed alcohols, aliphatic and aromatic alcohols, were not affected by bottle storage, with the exception of linalool (Table 1). These findings are substantially in line with previous studies of Qiao and Sun (2015) and Matias-Guiu et al. (2020), which found no significant differences in these compounds during glass bottle storage of Fenjiu and fruit spirits, respectively.

Table 1 Effect of bottle storage time and ageing modality on the content of volatile compounds (mg/L) in wine spirits samples—summary of ANOVA analysis and means comparison tests performed

	Bottle storage (months)			Ageing modality					Interaction
Volatile compounds	0	6	12	B	N	O15	O30	O60	
Linalool	0.193 ± *0.053*a	0.313 ± *0.017*b	0.300 ± *0.008*b	0.251 ± *0.090*a	0.261 ± *0.082*a	0.260 ± *0.072*a	0.287 ± *0.034*b	0.283 ± *0.027*b	s
Butanoic acid	0.834 ± *0.087*a	0.743 ± *0.161*a	1.089 ± *0.252*b	-	-	-	-	-	ns
Hexanoic acid	2.401 ± *0.195*b	2.199 ± *0.125*a	2.103 ± *0.109*a	-	-	-	-	-	ns
Isovaleric acid	1.608 ± *0.589*b	1.270 ± *0.074*a	1.301 ± *0.099*a	1.574 ± *0.507*b	1.569 ± *0.471*b	1.422 ± *0.326*ab	1.209 ± *0.128*a	1.190 ± *0.149*a	ns
Dodecanoic acid	0.652 ± *0.058*b	0.572 ± *0.062*a	0.581 ± *0.048*a	0.663 ± *0.039*b	0.597 ± *0.034*a	0.586 ± *0.057*a	0.558 ± *0.078*a	0.603 ± *0.076*a	ns
Ethyl 2-methylpropanoate	0.703 ± *0.165*b	0.787 ± *0.046*a	0.805 ± *0.086*a	0.896 ± *0.119*b	0.684 ± *0.093*a	0.752 ± *0.135*a	0.718 ± *0.049*a	0.775 ± *0.059*a	ns
2-Methyl-1-propylacetate	0.741 ± *0.787*b	0.532 ± *0.703*a	< QL	0.978 ± *0.829*b	0.960 ± *0.768*b	0.183 ± *0.312*a	< QL	< QL	ns
Ethyl isovalerate	0.122 ± *0.038*a	0.092 ± *0.125*a	0.241 ± *0.031*b	0.169 ± *0.061*ab	0.202 ± *0.083*b	0.119 ± *0.105*a	0.137 ± *0.123*ab	0.131 ± *0.122*ab	ns
3-Methyl-1-buthyl acetate	2.391 ± *0.169*b	2.074 ± *0.120*a	2.041 ± *0.144*a	2.351 ± *0.242*c	2.130 ± *0.120*ab	2.124 ± *0.161*ab	2.052 ± *0.248*a	2.186 ± *0.207*b	s
Ethyl hexanoate	1.742 ± *0.112*b	1.621 ± *0.132*a	1.619 ± *0.076*a	1.777 ± *0.172*b	1.575 ± *0.060*a	1.669 ± *0.047*ab	1.609 ± *0.095*a	1.672 ± *0.104*ab	ns
Ethyl octanoate	2.063 ± *0.311*b	3.370 ± *0.511*a	3.645 ± *0.603*a	3.392 ± *0.981*b	2.680 ± *0.647*a	3.336 ± *0.908*b	2.654 ± *0.816*a	3.069 ± *0.834*ab	ns
Malic acid diethyl ester	< QL	< QL	1.188 ± *0.333*	0.567 ± *0.892*b	0.351 ± *0.562*a	0.345 ± *0.536*a	0.395 ± *0.617*a	0.321 ± *0.502*a	–
Acetic acid	278.4 ± *79.6*b	246.5 ± *52.3*a	249.2 ± *57.6*a	376.2 ± *41.6*b	220.8 ± *6.4*a	231.6 ± *13.9*a	231.5 ± *11.3*a	230.1 ± *19.7*a	s
Guaiacol	-	-	-	0.091 ± *0.030*a	0.528 ± *0.052*b	0.518 ± *0.036*b	0.479 ± *0.051*b	0.522 ± *0.053*b	ns
4-Methyl-guaiacol	0.234 ± *0.101*a	0.265 ± *0.094*b	0.286 ± *0.108*b	0.077 ± *0.053*a	0.307 ± *0.053*b	0.315 ± *0.053*b	0.298 ± *0.053*b	0.312 ± *0.064*b	ns
Eugenol	0.285 ± *0.039*a	0.286 ± *0.031*a	0.326 ± *0.032*b	0.348 ± *0.025*c	0.276 ± *0.041*a	0.282 ± *0.024*ab	0.304 ± *0.033*b	0.287 ± *0.020*ab	ns
Syringol	-	-	-	0.247 ± *0.049*a	1.447 ± *0.118*b	1.478 ± *0.100*b	1.458 ± *0.063*b	1.431 ± *0.090*b	ns
4-Methyl-syringol	1.037 ± *0.365*b	0.936 ± *0.318*a	0.935 ± *0.323*a	0.342 ± *0.026*a	1.065 ± *0.032*b	1.167 ± *0.068*c	1.126 ± *0.099*c	1.147 ± *0.092*c	ns
Vanillin	-	-	-	7.778 ± *0.425*b	5.485 ± *0.726*a	7.527 ± *0.375*b	7.267 ± *0.245*b	7.807 ± *0.523*b	ns
Acetovanillone	-	-	-	0.360 ± *0.042*a	0.838 ± *0.062*b	0.827 ± *0.076*b	0.834 ± *0.031*b	0.889 ± *0.070*b	ns
Acetaldehyde	58.67 ± *5.48*a	62.07 ± *3.94*b	68.29 ± *5.65*c	60.99 ± *4.14*ab	59.70 ± *10.60*a	64.85 ± *2.37*c	65.25 ± *6.92*c	64.28 ± *4.64*bc	ns
Ethyl acetate	390.9 ± *48.7*a	408.9 ± *72.3*ab	419.5 ± *76.9*b	520.7 ± *40.2*b	360.4 ± *42.6*a	384.0 ± *15.1*a	372.1 ± *26.1*a	395.1 ± *20.9*a	ns

Fisher test: for each compound, means within the same line followed by different letters are significantly different ($p < 0.05$); ns for $p \geq 0.05$; *QL*, quantification limit

Linalool, a remarkable monoterpene compound found in *Vitis vinifera* grape juice and wines (Kersh et al., 2023; Tarasov et al., 2021), may be influenced by fermenting and distilling conditions (Canonico et al., 2023; Tian et al., 2022). However, linalool persists in the distillate after the wood ageing step and has been identified as a key odorant compound in both freshly distilled Cognac and aged wine spirits, contributing to floral and citrus notes (Caldeira et al., 2008; Ferrari et al., 2004). However, the observed increase in linalool content during bottle storage (Table 1) contradicts the findings from other studies; indeed, a decrease in linalool levels in Madeira rums during bottle storage has been reported by Aguiar et al. (2021). Further research is necessary to elucidate this discrepancy. Moreover, the increase in linalool content varied among samples from different ageing modalities (Table OR1b), with a more pronounced increase observed in samples aged in wooden barrels (B), those aged with staves with low level of micro-oxygenation (O15), and those aged with staves and nitrogen (N). Evidence exists that geraniol identified in chestnut staves (Fernández de Simón et al., 2014) can isomerize and to generate linalool (Fajdek-Bieda et al., 2023), which may explain the increase in linalool concentration observed during the first 6 months of bottle storage.

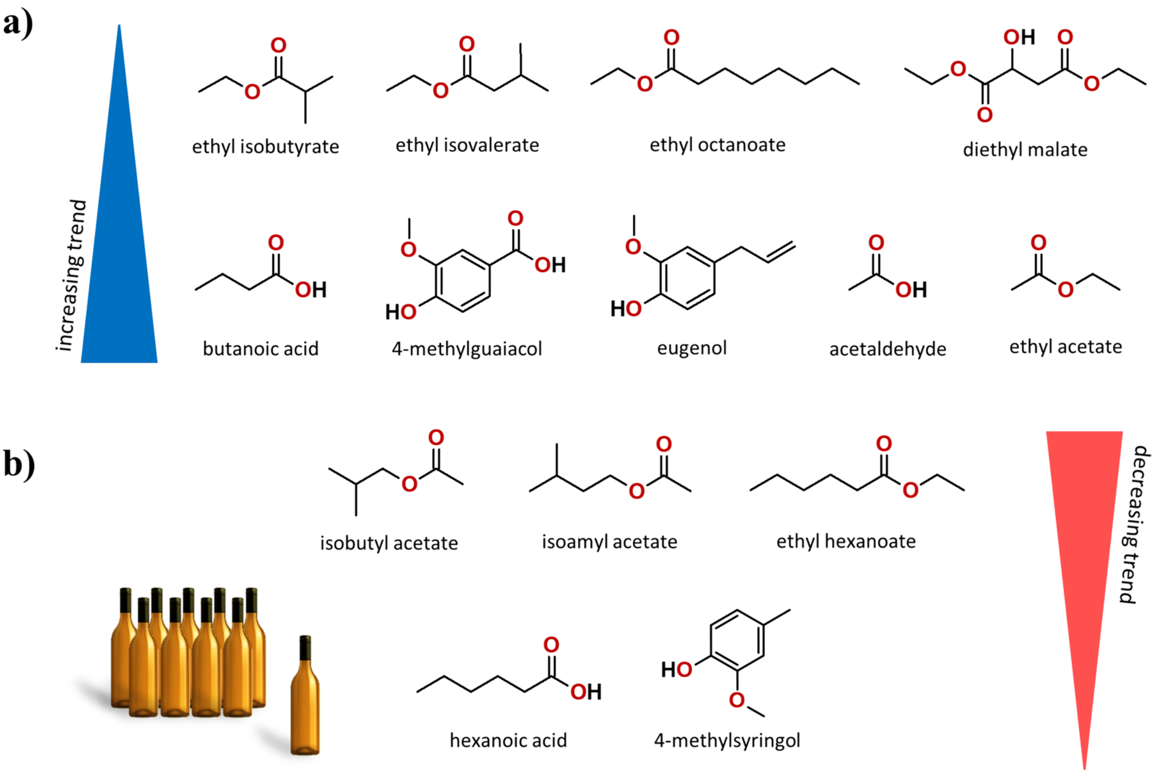


Fig. 2 Structures of the most representative: **a** volatile compounds with an increasing concentration trend and **b** volatile compounds with a decreasing concentration trend over bottle storage time

Acetaldehyde is the primary aldehyde found in freshly distilled wine spirits, whose content depends significantly on the distillation technique (Matias-Guiu et al., 2020). During wood ageing of alcoholic beverages, acetaldehyde can be formed through the oxidation of ethanol and subsequently oxidize further to produce acetic acid (Reazin et al., 1976). During bottle storage, a significant increase in acetaldehyde levels was observed (Table 1). By the end of the bottle storage period (12 months), the acetaldehyde levels of all wood ageing modalities were similar. These findings suggest that within the bottle, ethanol oxidation may be more vigorous than acetaldehyde oxidation, with these reactions potentially influenced by pre-bottling micro-oxygenation treatments. Similar observations were made by Ugliano et al. (2012) with red wines, in which post-bottling oxygen consumption was affected by pre-bottling micro-oxygenation conditions, consequently impacting the evolution of some sulphur volatile compounds. The increase in acetaldehyde during bottle storage (Table 1) was in line with the changes reported for bottled wine storage (Ugliano, 2013) but differs from the findings for grape marc spirits (Flouros et al., 2003), in which no significant modifications were reported for this compound.

The majority of the analysed acids were influenced by bottle storage, showing a tendency to decrease over time. An exception is butanoic acid, which maintains unchanged levels during the first six months of bottle storage before increasing by the twelfth month. These outcomes are consistent with those of Diéguez et al. (2002) on grape marc spirits who also observed a clear decrease in organic acid levels over time. Similarly, a study by Valaer and Frazier (1936) on whiskey storage over 4 years in bottles revealed a decrease in acids quantified by a global chemical method. Additionally, in the bottle storage of red wine, a significant decrease in all acids was found, except for butanoic acid (Pérez-Prieto et al., 2003). Fatty acids, originating from fermentation, may vary in freshly distilled wine spirits due to fermentation conditions and distillation with lees and usually present unpleasant odour notes (Ferrari et al., 2004). Watts and Butzke (2003) proposed a mechanism for ketone formation from fatty acid oxidation during wood ageing. On the other hand, acetic acid exists in freshly distilled wine spirits (Ferrari et al., 2004), but its content may change during wood ageing due to extraction from the wood, ethanol oxidation, and participation in ethyl acetate formation (Reazin et al., 1976). Hence, the decreasing trend in acid levels detected in this study suggests their involvement in oxidation and esterification reactions during bottle storage, which could be important from a sensory perspective as most acids are associated with unpleasant olfactory notes (Caldeira et al., 2008; Ferrari et al., 2004).

Ethyl acetate increased over the bottle storage, as for acetic acid, suggesting the prevalence of esterification reactions with the participation of this acid, as established by Reazin et al. (1976). This is supported by the decreasing concentration of acetic acid during bottle storage, indicating that it was consumed. A more detailed analysis of other identified volatile esters, which are primarily associated with pleasant fruity odours (Caldeira et al., 2008; Ferrari et al., 2004), reveals two trends: some esters contents, such as ethyl isobutyrate, ethyl isovalerate, ethyl octanoate, and diethyl malate, increase during bottle storage, while others, like isobutyl acetate, isoamyl acetate, and ethyl hexanoate, decrease over time. Similar trends were observed in Fenjiu distillates during a 3-year bottle storage period (Qiao & Sun, 2015). These results reveal the occurrence of esterification reactions, as seen with ethyl acetate, alongside ester hydrolysis, which has been proposed to explain the decrease in ester levels observed in bottled wines (Pati et al., 2020; Ramey & Ough, 1980; Ugliano, 2013). Indeed, acetate esters of higher alcohols generally hydrolyse more rapidly than ethyl fatty acid esters in both wine and model solutions (Ramey & Ough, 1980).

The increase in ethyl esters was expected, as ethanol and acetic acid are the most common reactive substrates in wine spirits and, in a slightly acidic environment, they can react with one another or with other acids, to produce ethyl acetate, in the case of ethanol, and other esters via esterification. The interconversion of ethanol, acetaldehyde, acetic acid, and ethyl acetate is well-known and could influence the flavour development and production of alcoholic beverages. Oxidation reactions can produce acetaldehyde and acetic acid, while esterification and hydrolysis reactions include the generation and breakdown of ethyl acetate. When minority substrates with carboxylic acid and alcohol functional groups combine with one another, these same esterification mechanisms might produce additional esters at lower concentrations. These other esters, while limited in quantity when compared to ethyl acetate, can also increase in concentration throughout the bottle storage period and will add different kinetic and flavour properties depending on the complexity of the aliphatic chain. Ester formation is a dynamic process that continuously takes place. During the process of transesterification, subsequently synthesised esters have the ability to react with an alcohol and produce a different ester compound. Table 1 and Fig. 2 disclose that the reactivity/stability of these esters varied; the three compounds that showed a concentration decrease were those with the most sterically hindered or long alkyl chains: isobutyl acetate, isoamyl acetate, and ethyl hexanoate.

Regarding the volatile compounds originating from the wood, different effects of bottle storage were observed (Table 1). No significant influence was detected for vanillin levels, which is in accordance with previous findings (Oliveira-Alves et al., 2022), nor for acetovanillone, syringol, and 4-allylsyringol levels. However, guaiacyl phenols (guaiacol, 4-methylguaiacol, eugenol) and 4-methylsyringol contents were notably affected by bottle storage time (Table 1). The former compounds exhibited an increase over time, while the latter showed a tendency to decrease (Table 1). These effects were consistent across all ageing modalities, as no interaction between the two factors was detected (Table 1). Fernandéz de Simón et al. (2006) reported similar findings for red wines, observing a slight increase in guaiacol and eugenol levels during the first year of bottling followed by a decrease in the second year.

Effect of the Ageing Technology on the Volatile Composition After 1 Year of Bottle Storage

The ageing technologies exerted a significant influence on the content of several volatile compounds, which is in agreement with outcomes obtained for these wine spirits prior to bottling (Caldeira et al., 2021). Indeed, the results shown in Table 1 and Table OR1a show that the distinctions induced by the ageing technologies remained even after 1 year of bottle storage.

Hence, significantly lower contents of volatile phenols, including guaiacol, 4-methylguaiacol, syringol, and 4-methylsyringol, as well as acetovanillone, were found in wine spirit samples aged in chestnut wooden vessels (B) (Table 1). Conversely, for eugenol and vanillin, lower levels were detected in samples aged with nitrogen (N), with the highest amounts observed in those aged in wooden vessels (B) and those subjected to the highest micro-oxygenation flow (O60) (Table 1). However, no effect of the ageing technology was detected on 4-allylsyringol content (Table 1 and Table OR1a). Similar effects were observed for syringol, guaiacol, and acetovanillone in wines aged using different ageing technologies after 10 years of bottle storage (del Alamo-Sanza et al., 2019).

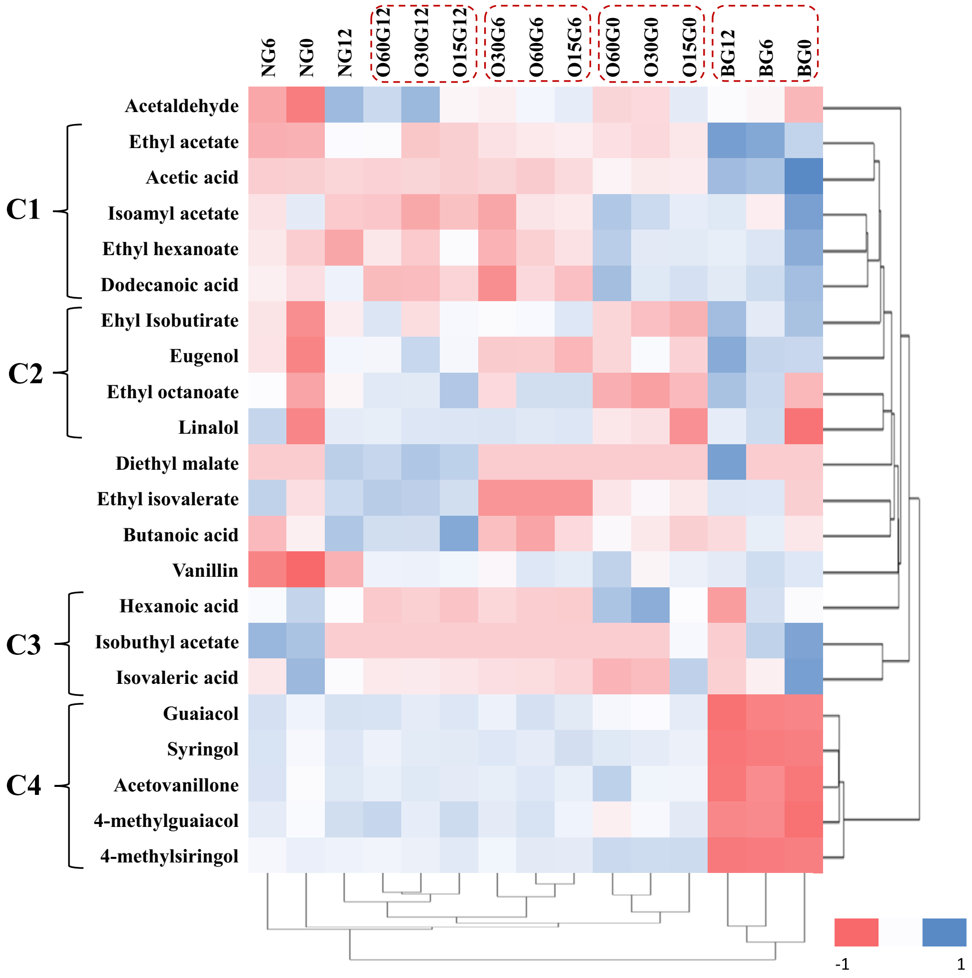
For other volatile compounds originating from the distillate, such as ethyl hexanoate, ethyl acetate, acetaldehyde, isobutyl acetate, and acetic acid, a significant effect of wood ageing technology was detected (Table 1, Table OR1a, and Table OR1b). Wine spirits aged in wooden barrels (B) had significantly higher contents of ethyl hexanoate, ethyl acetate, and isobutyl acetate. These samples, along with those aged under nitrogen (N), showed the lowest levels of acetaldehyde (Table 1). Such findings are also in agreement with those obtained for these wine spirits after 1 year of wood ageing (Caldeira et al., 2021). Conversely, a significant effect of the wood ageing technology was observed on the isovaleric acid, dodecanoic acid, ethyl isobutyrate, ethyl isovalerate, ethyl octanoate, and diethyl malate contents, which differ from previous findings reported by Caldeira et al. (2021). It suggests that the changes occurring in the bottle, as aforementioned, potentiated the discrimination induced by the ageing technologies. For acids and the majority of esters, significantly higher amounts were determined in wine spirits aged in wooden barrels (B). These outcomes differ from the results obtained in red wines (del Alamo-Sanza et al., 2019; Roussis et al., 2013). In fact, although the ageing process is similar, the alcoholic beverages being aged are quite different, and therefore, the phenomena occurring during the aging of red wine (Jarauta et al., 2005) and wine spirit (Canas et al., 2022) are not exactly the same. When wine is placed in a barrel, it contains low ethanol content, many non-volatile compounds such as sugars and several phenolic compounds (Lisanti et al., 2021), and volatile compounds (Jarauta et al., 2005). Conversely, young spirits have high ethanol content, water, and volatile compounds (Caldeira et al., 2010). Moreover, the rate of oxygen transmission, which corresponds to the oxygen reaching the alcoholic beverage through the joints between the staves and through the wood, depends on factors such as species and geographic origin, among others (del Alamo-Sanza et al., 2017). These differences could explain the discrepancies between the results in this manuscript and those of cited works (del Alamo-Sanza et al., 2019; Roussis et al., 2013).

Multivariate Approach of Volatile Composition Results

Cluster heatmaps were also made to ascertain whether the analysed variables could distinguish the pre-established groups (such as wood ageing technology and bottle storage) (Fig. 3).

Only the variables with a significant effect in the ANOVA analysis were considered for this approach. Wine spirit differentiation seemed to be primarily influenced by the ageing process, with samples aged in wooden barrels (B) positioned opposite those treated with nitrogen (N). However, regarding the samples from micro-oxygenation (O15, O30, and O60), the differentiation was based on the duration of storage in bottle. The more relevant variables, to the differentiation of traditional ageing technology versus alternative one, were volatile phenols (guaiacol, 4-methylguaiacol, syringol, and 4-methylsyringol), exhibiting a negative correlation,

Fig. 3 Cluster heatmap of volatile compounds present in aged wine spirits according to the modalities under study



whereas acetic acid and ethyl acetate were positively correlated. Some of these discriminating variables, namely, volatile phenols, could be used in the quality control of ageing technologies through rapid analysis techniques (Anjos et al., 2022). Samples stored for longer durations in bottle exhibit stronger associations with ethyl octanoate, diethyl malate, linalool, and acetaldehyde, showing an increasing trend with time in the bottle.

The diethyl malate, ethyl isovalerate, and butanoic acid present a considerable evolution in the micro-oxygenation samples O30 compared to the others. Compounds in cluster C1 and C2 are also important to discriminate samples.

Although the duration of storage in bottle seemed to affect several volatile compounds, changes induced by different ageing technologies remain even after this stage in bottle (Fig. 3). Similar behaviour has been reported for red wines by del Alamo-Sanza et al. (2019).

The variation in average concentrations of volatile compounds during bottle storage leads to the conclusion that some chemical reactions take place inside the bottle and may be influenced by the oxygen level. The observed changes in volatile compound contents cannot be attributed to a single cause but rather to a variety of chemical and physical phenomena. These include competing mechanisms, hydrolysis reactions that produce acids, oxidation that results in the formation of esters, oxygen exposure, compounds present in the ageing environment, and adsorption of volatile substances. However, given the scarcity of data in the literature about the reaction mechanisms involving these compounds, further in-depth study, that includes controlling of available oxygen, is needed for a better understanding of the bottle storage effects.

Effect of Bottle Storage on the Sensory Profile of Aged Wine Spirits

The ANOVA applied to the sensory data revealed that bottle storage had a significant influence on 12 sensory attributes, whereas the ageing technologies affected six attributes (Table 2 and Online Resource 2 Table OR2). Attributes such as rancid, flavour evolution, and flavour complexity were positively correlated with overall quality, while alcohol, burning astringency, and bitterness were negatively correlated (Granja-Soares et al., 2020). These attributes tended to increase over the 1-year bottle storage period (Table 2). Conversely, sweetness and green intensity tended to decrease over time, while the coffee attribute exhibited an initial increase followed by a decrease. During the storage in the bottle, the overall quality tended to increase, at least in the first 6 months, and then stabilize (Table 2).

These results suggest that the potential reactions occurring inside the bottle favoured the quality of the beverage, and this tendency was not influenced by the ageing technology, as no interaction between the two factors was found (Table 2 and Table OR2). To analyse this further, it is necessary to cross-check these results with the evolution of volatile species in previous subsections. Ethyl esters, for example, exhibit less intermolecular interactions than carboxylic acids as the hydrogen atom is not directly linked to the oxygen atom. As a result, ethyl esters tend to be more

Table 2 Effect of bottle storage time and ageing modality on sensory attributes of wine spirits samples—summary of ANOVA analysis and means comparison tests performed

Sensory attribute	Bottle storage (months)			Ageing modality					Interaction
	0	6	12	B	N	O15	O30	O60	
Alcohol	2.0 ± *0.0* ab	1.9 ± *0.3* a	2.2 ± *0.4* b	-	-	-	-	-	ns
Rancid	0.0 ± *0.0* a	0.0 ± *0.0* a	0.6 ± *0.5* b	-	-	-	-	-	ns
Dried fruits	-	-	-	2.2 ± *0.3* b	1.6 ± *0.5* a	2.1 ± *0.6* b	1.9 ± *0.6* ab	1.8 ± *0.4* ab	ns
Coffee	0.5 ± *0.7* ab	0.8 ± *0.6* b	0.2 ± *0.5* a	-	-	-	-	-	ns
Green	0.2 ± *0.3* b	0.0 ± *0.0* a	0.0 ± *0.0* a	0.0 ± *0.0* b	0.2 ± *0.3* b	0.0 ± *0.0* a	0.0 ± *0.0* a	0.0 ± *0.0* a	ns
Sweetness	2.8 ± *0.4* b	2.2 ± *0.2* a	2.3 ± *0.5* a	-	-	-	-	-	ns
Smooth	-	-	-	3.2 ± *0.3* b	3.1 ± *0.4* b	3.1 ± *0.2* b	2.5 ± *0.5* a	3.1 ± *0.3* b	ns
Burning	2.2 ± *0.4* a	2.3 ± *0.5* a	2.7 ± *0.4* b						ns
Astringency	2.1 ± *0.3* b	1.6 ± *0.7* a	2.3 ± *0.5* b	-	-	-	-	-	ns
Bitter	1.5 ± *0.5* a	1.9 ± *0.7* b	2.2 ± *0.3* b	-	-	-	-	-	ns
Flavour evolution	2.8 ± *0.4* a	3.2 ± *0.5* b	3.4 ± *0.4* b	-	-	-	-	-	ns
Flavour complexity	2.8 ± *0.4* a	3.2 ± *0.2* b	3.4 ± *0.3* b	-	-	-	-	-	ns
Retronasal aroma	-	-	-	3.3 ± *0.4* ab	2.9 ± *0.3* a	3.6 ± *0.5* b	3.4 ± *0.5* b	3.2 ± *0.4* ab	ns
Overall quality	13.7 ± *0.9* a	14.5 ± *0.8* b	14.3 ± *0.5* b	14.5 ± *0.4* b	13.8 ± *0.9* a	14.7 ± *0.6* b	14.1 ± *1.1* ab	14.4 ± *0.6* ab	ns

Fisher test: for each sensory attribute, means within the same line followed by different letters are significantly different ($p < 0.05$); ns for $p \geq 0.05$

volatile than their corresponding carboxylic acids, making their odorous qualities more detectable. As a result, when comparing a molecule with a carboxylic acid group to its ethyl ester, the ethyl ester is expected to be more volatile, which could explain the increased complexity and flavour evolution ascribed by the panel of tasters for the samples stored in bottles for 12 months (Table 2). To our knowledge, there are no published results on the impact of bottle storage on the sensory properties of distilled beverages, which hinders the discussion of the results presented in this study. Only a limited number of studies have been conducted on white and red wines, which have shown an improvement in overall quality for storage periods of 18 and 15 months, respectively (Liu et al., 2016; Vázquez-Pateiro et al., 2020).

Effect of Ageing Technology on the Sensory Profile of Wine Spirits After 1 Year of Bottle Storage

At the end of 1 year of bottle storage, the ageing technology seems to have a more subdued effect than that detected prior to bottling, as reported in a previous study (Caldeira et al., 2021). These results (Table 2) suggest that the favourable effects of bottle storage previously mentioned may have mitigated the sensory discrimination introduced by the ageing technology. However, the separation in terms of overall appreciation remains, with wine spirits produced in the absence of oxygen (N) showing significantly lower values and spirits from traditional ageing technology (B) and alternative ageing technologies with micro-oxygenation (O15 and O60) presenting significantly higher values. In the alternative technology, the O30 modality exhibited the lowest overall quality. These findings may be assigned to distinctions observed in attributes such as dried fruits, smoothness, and retronasal aroma, aligning consistently with those identified prior to bottling (Caldeira et al., 2021).

Multivariate Approach of Sensory Results

Hierarchical clustering analysis was also conducted on the sensory analysis results, using only the variables that proved to be significantly influenced by the factors under study. The heatmap obtained (Fig. 4) shows that sample separation was mainly determined by the bottle storage time, with a clear

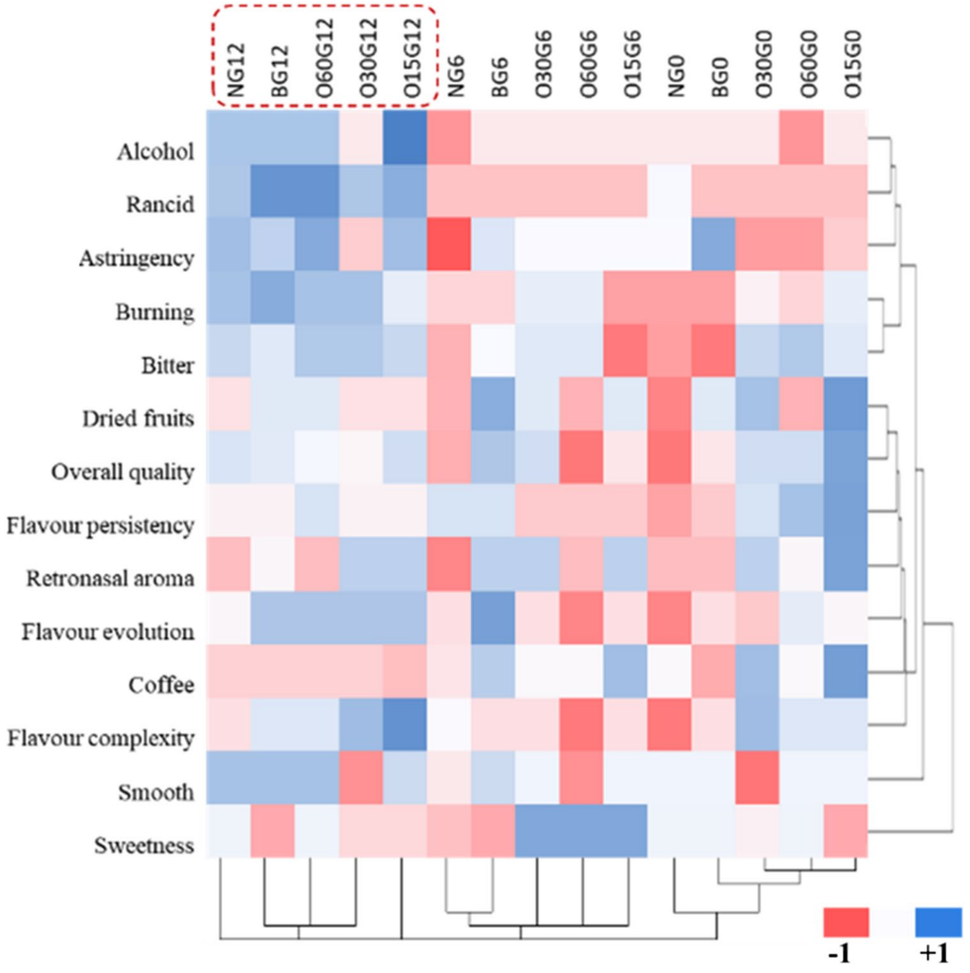


Fig. 4 Cluster heatmap of sensory attributes in aged wine spirits according to the modalities under study

distinction between the beginning (samples G0) and the end of this stage (samples G12).

Conclusions

This study presents novel and crucial information on the effect of bottle storage on volatile compounds, shedding light on their significant impact on the sensory attributes of wine spirits. The comprehensive sampling of wine spirits with several ageing modalities, including chestnut staves and micro-oxygenation, facilitated the identification of volatile species that are particularly susceptible to longer bottle storage periods, thereby pivotal for ensuring product quality and customer satisfaction.

Statistical analysis of the results revealed a significant effect of bottle storage time on the levels of various odorant compounds, such as volatile phenols (4-allylsyringol, 4-methylsyringol, eugenol, and 4-methylguaiacol), acids (acetic, butanoic, hexanoic, and dodecanoic acids), and esters (ethyl isobutanoate, isobutyl acetate, ethyl isovalerate, isoamyl acetate, ethyl hexanoate, ethyl octanoate, and diethyl malate), while others (primarily alcohols) remained unaffected. Changes were observed in the contents of these compounds, and some of them were influenced by the ageing technology; the ageing technology notably influenced the levels of 20 odorant compounds. Multivariate analysis highlighted the differentiation between samples from traditional ageing technology and those produced with alternative ageing technologies, with a slight separation based on bottle storage time.

In terms of sensory results, a significant effect of bottle storage time on several sensory attributes were noted, whereas the ageing technology influenced only four attributes (dried fruits, green, smooth, and retronasal aroma). The overall quality of the samples improved with increased bottle storage time.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11947-024-03573-z.

Acknowledgements The authors thanks João Pedro Catela, Nádia Santos, Manuela Gomes, Eugénia Gomes, and Inês Antunes from Adega Cooperativa da Lourinhã, José Abílio Gonçalves and Sérgio Gonçalves from Tanoaria J. M. Gonçalves, Pedro Rodrigues and Diogo Rodrigues from AZ3Oeno Portugal, Deolinda Mota, Silvia Lourenço and João Amaral from INIAV-Polo de Inovação de Dois Portos as well as A. Pedro Belchior for their technical support. Additionally, gratitude is extended to all the tasters who participated as volunteers in the sensory sessions. The authors also thank MED (https://doi.org/https://doi.org/10.54499/UIDB/05183/2020; https://doi.org/https://doi.org/10.54499/UIDP/05183/2020), CHANGE (https://doi.org/https://doi.org/10.54499/LA/P/0121/2020), CERNAS-IPCB (https://doi.org/https://doi.org/10.54499/UIDP/00681/2020), CEF (https://doi.org/https://doi.org/10.54499/UIDB/00239/2020), TERRA (https://doi.org/https://doi.org/10.54499/LA/P/0092/2020) and CQE (https://doi.org/https://doi.org/10.54499/UIDP/00100/2020)

Author Contribution Ilda Caldeira: funding acquisition, conceptualization, investigation, methodology, validation, software, formal analysis, supervision, writing-original draft preparation, writing—review and editing; Ofélia Anjos: funding acquisition, conceptualization, investigation, methodology, validation, software, formal analysis, supervision, writing-original draft preparation, writing—review and editing; Cláudia Vitória: methodology, software, formal analysis, writing—review and editing; Sheila Alves: formal analysis, writing—review and editing; Tiago A. Fernandes: funding acquisition, conceptualization, writing—original draft preparation, writing—review and editing; Sara Canas: funding acquisition, project administration, conceptualization, methodology, investigation, writing—review and editing; Sofia Catarino: funding acquisition, conceptualization, writing—review and editing. All authors reviewed the manuscript.

Funding This study was supported by the research Project OXYREBRAND, funded by National Funds through FCT—Foundation for Science and Technology under Project POCI-01–0145-FEDER-027819 (PTDC/OCE-ETA/27819/2017). Additionally, this work was funded by National Funds through FCT—Foundation for Science and Technology through the Projects UIDB/05183/2020 (https://doi.org/10.54499/UIDB/05183/2020; https://doi.org/10.54499/UIDP/05183/2020 and https://doi.org/10.54499/LA/P/0121/2020) [MED&CHANGE]; UID/AGR/04129/2020, DL 57/2016/CP1382/CT0025 [LEAF]; UIDB/00681/2020 (https://doi.org/10.54499/UIDP/00681/2020) [CERNAS-IPCB]; UIDB/00239/2020 (DOI 10.54499/UIDB/00239/2020) [CEF]; UIDB/00100/2020 (DOI 10.54499/UIDP/00239/2020) [TERRA], PTDC/QUI-QAN/32242/2017, UIDP/00100/2020[CQE] and LA/P/0056/2020 (https://doi.org/10.54499/LA/P/0056/2020), as well as contract CEECIND/02725/2018/CP1572/CT006 (10.544.CEECIND/02725/2018/CP1572/CT0006 (to Tiago A. Fernandes).

Data Availability No datasets were generated or analysed during the current study.

Declarations

Conflict of Interest The authors declare no competing interests.

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