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Changes in the Chemical Composition of Plum Distillate During Maturation with Oak Chips under Different Conditions

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Summary

This study investigates the effect of ageing on the qualitative and quantitative composition of plum distillate in contact with oak wood chips. Maturation was performed with lightly toasted French oak (*Quercus sessiflora* and *Quercus robur*) chips or oak chips made from fragments of empty barrels that had been used for ageing cognac. The effects of oak chip dose, process temperature, ageing system (static or circulatory) and ultrasound treatment were assessed. Maturation of plum distillate samples with oak chips resulted in higher levels of extractable organics (including tannins) and colour changes, which were correlated with the type and dose of oak chips, and the conditions of maturation. The content of sugars such as glucose, xylose and arabinose also increased, depending on the conditions and type of oak chips. Degradation of lignin resulted in liberation of sinapaldehyde, syringaldehyde, coniferaldehyde and vanillin, with intensities depending on the applied parameters. In terms of volatiles, decreases in the concentration of higher alcohols and aliphatic aldehydes were observed in the majority of maturation experiments, while concentrations of furanic aldehydes increased depending on the type and dose of oak chips, as well as on the conditions of maturation. The quantities of esters such as ethyl acetate decreased in the majority of experimental variants, whereas concentrations of ethyl caproate, ethyl caprylate and ethyl caprate increased gradually. Some phenols and lactones were detected in all matured samples, with the lowest levels found in the samples aged with oak chips made from cognac barrels.

Key words: plum distillate, maturation, plum brandy colour, volatile compounds, phenolic compounds, lactones

Introduction

Plum brandy (slivovitz) is the spirit prepared from varieties of plum (*Prunus* sp.), originating from the Balkan Peninsula. This beverage is also quite well known in Central Europe (Hungary, Poland, Czech Republic, Slovakia and Romania), and similar plum brandies are produced in Germany, Austria (Zwetschgenwasser), France (eau de vie de prune), and Switzerland (Pflümli wasser). Poland also has a long tradition of making slivovitz. One of the more recognized such products is Śliwowica Łącka,

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which is produced by spontaneous fermentation of Węgierka Zwykła plums grown in a submontane region of Poland with specific climatic and soil conditions (*1*).

The quality of plum brandy is influenced by many factors, including the soil and climate, the characteristics of the used fruit varieties and the employed technological procedures (*2*). Plum varieties with their distinctive aromatic compounds give to the distillates the features of a local character (*3*).

In view of the fact that new distillates are characterized with inharmonious taste and sharp odour, the ageing in wooden casks is the most important, final step of production process of several beverages (cognac, whisky, brandy, calvados, mescal and wine). It is also common to age the plum brandies in wooden barrels (*4*).

Ageing, also known as maturation, is one of the most important and also costly factors that influence the quality of distilled beverages, such as cognac, whisky, brandy and calvados. While probably serving originally only as a means of storage and transportation, time spent in an oak cask is now seen as fundamental to the finished taste and aroma (*5*). During maturation, spirit beverages develop a distinctive aroma and flavour that are appreciated by consumers. Ageing not only improves the sensory attributes of distilled beverages, but also causes changes in the content of total polyphenols, as well as developing other important health properties, such as increased antioxidant capacity (*6,7*).

The oak (*Quercus* sp.) is by far the most widely used wood in the manufacture of barrels for ageing wines and spirits. In Europe, the oak species *Quercus petraea* (syn. *Quercus sessilis*) and *Quercus robur* (syn. *Quercus pedunculata*) are used.

In America, the oak species *Quercus alba* is widely used, the wood of which is richer in lactones than the European species (*8*). A common practice in cooperage, regarded as the most important technological step, is toasting the oak wood. High temperatures modify the physical structure of the wood, which is crucial for facilitating the shaping of the staves. More importantly, however, its chemical composition undergoes significant changes due to thermal degradation of the oak wood polymers (*4*).

The type of cask and the duration of maturation are two variables equally important for quality of old plum brandies. Both parameters of ageing in production of whisky and cognac are defined according to the international or local regulation for the product name (*5*). According to the tradition in European countries, brandy is kept for at least two years (sometimes for several decades) in different wooden casks, but the local regulation for brandies does not define the ageing parameters. They depend a lot on the local production practice (*9*).

The process of ageing changes the colour and flavour of the maturing spirit and reduces both its volume and the alcoholic content. The time required for satisfactory maturation varies, depending on the characteristics of the raw distillate, the origins of the wood, the size and treatment of the cask and the environment in which the spirit is matured. Taste and aroma are influenced by changes in the composition and concentration of compounds. These changes may be caused by direct extraction of wood compounds, by decomposition of wood macromolecules and extraction of the products into the distillate, by reactions between wood components and the constituents of the raw distillate, by reactions involving only extractable wood components (*10*), by oxidoreductions, esterifications, Maillard reactions, polymerizations or condensations (*11*).

In addition to positive impact on the quality, the ageing of brandy in wooden barrels has a disadvantage, increased costs. Wooden barrels are expensive, and difficult to clean and maintain. Evaporation of ethanol from wooden barrels is higher than from stainless steel tanks. If the quality of the wood and the workmanship are poor, the quality of the spirits will be affected and evaporation losses will increase. Furthermore, barrels lose their extractable substances after a few years and must be replaced (*12*). Costs rise in proportion to the length of the ageing period, as a consequence of tied up capital.

Treatment with oak chips, especially charred or toasted chips, is known to speed up the process of brandy ageing. The simplest method for adding wood-derived compounds is to use oak chips. Oak chips are increasingly used in the maturation of brandies (*5*). The use of oak chips is part of traditional good cellar practice, it is legally permitted and commonplace in commercial production of wines (*12*). Oak chips of various sizes, pre-treated in different ways, are now commercially available and are used to compensate for the low levels of extractable components in old barrels, which may nevertheless be used to provide a barrel-like environment. Alternatively, renewable stacks of planks placed in large steel maturation tanks may be used in the production of brandies (*5*). The use of alternative solutions to the traditional barrels, such as oak wood fragments, may be a rapid and economical method of ageing treatment (*13*). Nowadays, there is no legislation applied to the ageing of spirits in contact with oak wood fragments. There are some works on brandy aged in contact with wood fragments, on cider brandy, on aged rum and on Brandy de Jerez (*14–16*). The composition of the final product is mainly influenced by the botanical and geographical origin of the used oak, but also important during accelerated ageing are: the oak fragment size, the amount of applied wood and the toasting level (light, medium or high) (*17,18*).

Oak extracts or chips contain very high levels of wood-derived compounds, which may even exceed those in barrels, in particular phenolic and furanic compounds (*19*). However, ageing using oak chips may be considered incomplete, as the oxygen that penetrates through the staves during the long maturation period has an important influence from a chemical and sensory standpoint. Different mechanisms have been proposed, whereby oxygen plays a part in the formation of compounds such as phenolic compounds derived from lignin (*20*), methylketones (*21*) or acetaldehyde and acetic acid (*22*).

This study set out to evaluate the effects of the type and dose of oak chips and maturation conditions on the changes in the qualitative and quantitative composition of plum distillate, using lightly toasted French oak wood chips and oak wood chips made from barrels used for ageing cognac.

Materials and Methods

Raw material

The raw material used was a plum distillate with an alcoholic strength by volume (ASV) of 77 %, produced on an industrial scale by a Polish factory, +H2O Sp. z o. o. (Chociszew near Lodz, Poland). The raw material used for the production of distillate were plums var. Węgierka Zwykła purchased from a Polish fruit processing factory (ROLFOODS, Ziewanice near Lodz, Poland). The plum pulp for alcoholic fermentation was prepared from the stoned plums (with the addition of 10 %, by mass, comminuted stones), supplemented with $(NH_4)_2HPO_4$ (0.2 g per kg of fruit pulp) as a nitrogen source for the yeast.

Fermentation was initiated using dry wine yeast *Saccharomyces bayanus* (Fermentis, Lesaffre, Marcq-en-Baroeul, France) in the mass fraction of 0.3 g per kg of plum pulp, with an alcohol tolerance of up to 18 % (by volume) and a wide fermentation temperature range (10–35 °C). The fermentation was conducted at 18 °C for 10 days. The distillation was done in a one-column continuous apparatus (F.P.H.U. SPO-CHEM, Myślenice, Poland).

Maturation of plum distillate samples was performed using lightly toasted French oak chips (produced from two varieties of French oak wood, *Quercus sessiflora* and *Quercus robur*) and French oak chips made from fragments of emptied barrels used for ageing cognac (Ares Trading SA, Warsaw, Poland). The average chip size was 7.5 mm×10 mm.

Experimental design

The scope of the experiments included assessment of the following factors: the type (see above) and dose of oak chips (3, 5 or 7 g/L), the temperature of fermentation (18–20, 35 or 45 °C) and the system of ageing (static, circulatory or ultrasound treatment). The effect of treatment with ultrasound was assessed using a SONOPULS HD 2200 homogenizer (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) in continuous mode, set to 50 % amplitude (ultrasound power 400 W, 24 kHz).

In all experiments, the plum distillate (77 %) was diluted with deionized water to ASV of 55 %. Next, 1.5 L of distillate was transferred into 2-litre lab bottles (made from clear soda glass) with a wide mouth and a ground glass stopper for an air-tight seal, and different amounts of oak chips were added. The prepared samples were aged for 12 months in a dark place under various conditions.

The variants for ageing were as follows: lightly toasted French oak chips, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week) (variant A), French oak chips obtained after ageing of cognac, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week) (variant B), lightly toasted French oak chips, 35 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 3 months, then matured under the same conditions as variant A) (variant C), lightly toasted French oak chips, 45 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 6 months, then matured under

the same conditions as variant A) (variant D), lightly toasted French oak chips, 45 °C, a circulation system (samples heated and stirred simultaneously for 2 h per day, 5 days per week, for 6 months, then matured under the same conditions as variant A) (variant E), and lightly toasted French oak chips, 20 °C with ultrasound treatment (50 % amplitude for 20 min per day, 5 days per week, for 3 months, then matured under the same conditions as variant A) (variant F).

Samples were prepared in triplicate for each variant, therefore 12 bottles were prepared, *i.e.* 3 bottles with plum distillate were aged for 3 months, 3 bottles for 6 months, 3 bottles for 9 months and 3 bottles for 12 months. Distillates were analyzed at the start of the ageing process and after 3, 6, 9 and 12 months. The analytes were: global parameters, such as alcoholic strength by volume (ASV in %), total extract content (g/L), colour according to CIELab chromatic parameters and concentrations of phenolic compounds (mg/L), tannins as pyrogallol (mg/L), sugars (mg/L), major and minor volatiles (mg/L of absolute alcohol). The control sample for each variant was the plum distillate before ageing.

Distillation process

The preparation of matured plum distillate samples for analyses consisted of separation of ethanol and other volatiles from the extracted compounds in a digital distilling unit model Super Dee (Gibertini Elettronica S.r.l., Novate Milanese, MI, Italy). Prior to distillation, 100 mL of sample were transferred to the glass flask of the distilling apparatus and then diluted with 50 mL of deionized water. In the next step, distillation was carried out to separate ethyl alcohol, water and other volatile compounds (distillate) from nonvolatile compounds (extract). Volatile compounds (approx. 90 mL) were condensed and collected in a separate volumetric flask (100 mL) and then distilled water was added to reach the same volume as the original sample (*i.e.* 100 mL). This sample was used for the determination of ASV and major volatile compounds.

The distillation residue, *i.e.* extract fraction, was cooled and quantitatively transferred to a 100-mL flask, and then distilled water was added to reach the same volume as the original sample, *i.e.* 100 mL. This sample was used for the concentration determination of total extract, phenolic compounds, tannins and sugars.

Alcoholic strength by volume

Ethanol concentration (*i.e.* alcoholic strength by volume; ASV) was measured with a hydrometer (thermoalcoholometer, Conbest Sp. z o.o., Cracow, Poland) using the percentage scale (by volume of ethanol) at 20 °C. If the distillate temperature diverged from 20 °C, then a correction was applied, using alcoholometric tables (*23*).

Total extract

Total extract content in the extracted fraction obtained after separation of ethanol and other volatiles, as described above, was determined following the methodology recommended in alcohol industry (*24*).

Colour according to CIELab chromatic parameters

Colour measurements were made using a Chroma Meter CR-5 (Konica Minolta, Osaka, Japan). The results were expressed in Commission Internationale d´Eclairage (CIE) L^*, a^* and b^* colour space coordinates (*25*). These parameters defined: L^* indicates lightness (white (100) to black (0)), a^* coordinate (red (+100) to green (−100)), b^* coordinate (yellow (+100) to blue (−100)). These parameters were measured using the CIE 1964 Standard Observer (10° visual field) and the CIE standard illuminant D65 as references (*9,25*). The total differences in colour (ΔE^*) between the tested samples (after maturation) and the standard sample (before maturation) were calculated according to the following equation (*26*):

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

HPLC-DAD analysis of phenolic compounds

Phenolic compounds were analyzed using a modified method described by dos Anjos *et al.* (*27*) on an Agilent 1260 Infinity Binary LC system (Agilent Technologies, Palo Alto, CA, USA), consisting of a binary pump, a vacuum degasser, an autosampler, a thermostated column compartment and a diode array detector (DAD) with a 10-mm max-light flow cell. The samples were filtered through 0.45 µm PES (polyethersulfone) membrane filters (Sigma-Aldrich, Saint Louis, MO, USA) before injection, and separations were performed using an Agilent-Zorbax Eclipse XDB-C18 (4.6 mm×250 mm, 5 µm) column connected to an Agilent-Zorbax Eclipse XDB-C18 4-Pack (4.6 mm× 12.5 mm, 5 µm) pre-column. The mobile phase consisted of 2 % acetic acid in water (solvent A) and methanol/water/acetic acid 70:28:2 (solvent B). The samples were eluted according to the following gradient: 0 to 25 min 0–40 % B, 25 to 40 min 40–55 % B, 40 to 43 min 55–60 % B, 43 to 50 min 60–100 % B, 50 to 55 min 100–0 % B and 55 to 60 min 0 % B.

Phenolic compounds were eluted at a 0.8 mL/min flow rate and the temperature was set to 30 °C. Absorbance was measured at 280 nm and the injection volume was 20 µL. The compounds coniferaldehyde, sinapaldehyde, syringaldehyde and vanillin were identified using reference standards, while quantification was performed using external calibration curves with standards. The recovery studies for the quantified compounds were observed in the range of 80–120 %. The relative standard deviations (RSDs) were less than 2.6 %.

Analysis of tannins

The content of tannins was determined in matured samples (once the volatiles, including ethanol and others, had been removed) and expressed as pyrogallol in mg/L, following the methodology described in the European Pharmacopoeia 5.0 (*28*).

HPLC analysis of sugars

The contents of glucose, xylose and arabinose in the samples of aged plum distillate were determined in the extract (obtained as described above), using an Infinity 1260 HPLC system equipped with a refractive index de-

tector (RID; Agilent Technologies, Palo Alto) in a Hi-Plex Ca column (7.7 mm×300 mm, 8 µm; Agilent Technologies). Column temperature was maintained at 80 °C. HPLC grade water was used as a mobile phase at a flow rate of 0.6 mL/min with a sample volume of 20 µL. Prior to analysis, all samples were filtered through 0.45-µm PES (polyethersulfone) membranes (*29*). The recovery studies for the quantified compounds were in the range of 92–107 %. The RSDs were less than 6.2 %.

GC-FID analysis of major volatile compounds

Analysis of the major volatile compounds was performed using gas chromatograph (GC) model Agilent 7890A (Agilent Technologies, Santa Clara, CA, USA), equipped with a flame-ionization detector (FID), a split/splitless injector and a capillary column (HP-Innowax, 60 m×0.32 mm inner diameter×0.50 µm film thickness; Agilent Technologies, Santa Clara). Each sample (1 µL) previously diluted with deionized water to ASV=40 % was injected directly onto the GC column using an autosampler in split mode (1:45). The temperature of the injector and FID was kept at 250 °C. The oven temperature program was as follows: 40 °C initial, 6-minute hold; 2 °C/min to 80 °C; 5 °C/min to 210 °C final, 5-minute hold. The flow rate of the carrier gas (helium) through the column was 2 mL/min. The volatile compounds (acetaldehyde, ethyl acetate, methanol, propanol, isobutanol, butanol, amyl alcohols, pentanol, hexanol and 2-phenylethanol) were identified using reference GC standards and then quantified with standard calibration curves, using 4-heptanone as the internal standard. All analyses were performed in triplicate. The obtained data were analysed using Mass-Hunter software (Agilent Technologies, Santa Clara).

SPME-GC-MS analysis of minor volatile compounds

Chromatographic analysis of the minor volatile compounds in the distillates was performed using a GC apparatus (Agilent 7890A; Agilent Technologies, Santa Clara) with a mass spectrometer (Agilent MSD 5975C; Agilent Technologies, Santa Clara). Solid phase microextraction (SPME) was used to extract the volatile compounds, with subsequent analysis using gas chromatography-mass spectrometry (GC-MS).

SMPE conditions were as follows: a 5-mL plum distillate sample (previously diluted to ASV=20 %) mixed with the internal standard (4-heptanone at a concentration of 45 mg/mL of absolute alcohol) was placed in a 20-mL amber headspace vial and capped tightly. Carboxene/divinylbenzene/polydimethylsiloxane fibre (DVB/CAR/PDMS, 50/30 µm, 1 cm fibre length; Supelco, Bellefonte, PA, USA) was used in the analyses of all samples. The vial was equilibrated at 50 °C for 15 min, after which SPME fibre was introduced. Extraction was continued for 15 min at 50 °C. The compounds were then desorbed for 5 min at 250 °C in splitless mode. GC-MS analysis was performed. After each injection, the fibre was conditioned at 250 °C for 10 min. All headspace SPME extractions were performed in triplicate.

SPME-GC-MS conditions were as follows: a capillary column (VF-WAX MS, 60 m×0.32 mm i.d.×0.50 µm film thickness; Agilent Technologies, Santa Clara) was used to separate the compounds. The GC oven temperature pro-

gram was as follows: 35 °C initial, 6-minute hold; 2 °C/min to 80 °C; 10 °C/min to 250 °C final, 5-minute hold. The flow rate of the carrier gas (helium) was 1.2 mL/min. The MS run parameters were as follows: ion source temperature 230 °C, transfer line temperature 260 °C, quadrupole temperature 150 °C and ionization energy 70 eV. Detection was carried out in scan mode over a range of m/z=29–289. The volatile components were identified based on a comparison of their mass spectra with those registered in the NIST MS library (NIST 98.1 and the Wiley Registry of Mass Spectral Data, 8th edition) (*30*). Their retention indices (RI) were also compared with reference compounds and data in the literature (*31,32*).

The minor volatile compounds were quantified with an internal standard method using MassHunter software (Agilent Technologies, Santa Clara). All gas chromatography standards were purchased from Sigma-Aldrich and were of GC purity. Standard solutions were prepared using anhydrous ethanol (Sigma-Aldrich) as the solvent and stored at 4 °C.

The recovery studies for the quantified compounds were in the range of 90–107 %. The determined RSDs of intra- and interday precision were lower than 20 %.

Statistical analysis

For the data evaluation, three-way (major volatile compounds) and two-way (phenolic compounds, tannins, sugars and CIELab) analysis of variance (ANOVA) with a significance level p≤0.05 were used, followed by Tukey's *post-hoc* test when significant effect occurred. Statistical analyses were performed using Statistica software v. 10 (Statsoft, Tulsa, OK, USA).

SPME-GC-MS data of minor volatile compounds in aged plum distillate samples were subjected to principal component analysis (PCA) using XLSTAT software v. 2017.4 (Addinsoft, New York, NY, USA).

Results and Discussion

Changes in alcoholic strength by volume and total extract content

The process of ageing of spirit beverages in oak wood barrels is associated with changes in the physical and chemical parameters of the maturing spirit (*5*). One such change is a decline in the volume and the alcoholic strength by volume (ASV) of a new spirit with ASV higher than approx. 50 %, as a consequence of the preferential evaporation of ethanol through the oak wood. Ullage increases at higher temperatures and lower humidity, as evaporation intensifies.

In this study, plum distillate with an ASV=55 % was stored in tightly closed bottles with oak wood chips. The loss of alcohol was not expected. Unfortunately, measurements taken in successive maturation periods indicated that the ASV had decreased (see Table 1). No significant effect of dose of oak chips on ethyl alcohol content was found in all maturation variants. The ethanol loss ranged from 0.48 to 0.88 % (by volume) and was affected by ageing system, as well as by temperature and time of maturation. When assessing the effect of ageing temperature on

the ethanol content changes, the statistically significant differences were observed (variants A, C and D). The lower volume fractions were found in distillates matured at 35 and 45 °C (variants C and D respectively) than in those aged at 18–20 °C (variant A). Moreover, time×temperature (variants A, C and D) and time×ageing system (variants D and E, and variants A and F) interactions were found. The application of agitation in the ageing systems such as ultrasound treatment and circulation may cause ethanol evaporation (Table 1). Lower ASVs could be further attributed to the penetration of ethanol into the porous structure of the oak chips during maturation.

Changes in the composition of aged spirits can also be related to the extraction of wood constituents (*33*). In our study, the maturation of plum distillate samples with oak chips resulted in an increase in total extract content in all experiments (Table 1). During ageing experiments, the effect of five factors on extract content changes was studied, as well as interactions among them. Extract levels in tested samples of matured plum distillate were significantly affected by the dose and type of oak chips, temperature and time of maturation, and ageing system, as well as by interactions among them. Only the interaction oak chips×ageing system (variants A and F) did not significantly affect extract content. The highest dynamics of extraction took place in the initial 3 months of maturation and decreased over the subsequent months. The values for this parameter were correlated with the dose and type of oak chips, as well as with the maturation conditions. After 12 months, samples aged with oak chips from cognac barrels were characterized by up to approx. 34 % lower extract content than those aged with fresh oak chips. Moreover, it was observed that higher temperatures during maturation led to higher levels of extractable organic content in the tested samples. Periodic circulation of the oak chips in the plum distillate also led to significant further increases in extract content.

CIELab chromatic parameters of aged plum distillate samples

Fresh plum distillates obtained after distillation are colourless. Changes in colour were a visible effect of ageing, resulting from the decomposition of polyphenols extracted from the oak wood chips (*6*). The colour of aged alcoholic beverages is very complex, and difficult to describe and compare. The CIElab method was used for more specific and objective determination of colour, measuring two colour coordinates, a^* and b^*, as well as a psychometric index of lightness, L^* (*34*). The data of CIELab chromatic parameters and results of two-way ANOVA are shown in Table 2.

The results of our study indicate that the type of oak chip (lightly toasted, not having been used previously for maturation, or made from barrels used for cognac maturation), the dose of wood chips and the conditions of ageing each had an effect on CIElab parameters. Also interactions of dose of lightly toasted French oak chips×temperature and dose of lightly toasted French oak chips×ageing system (static and circulation) combined with agitation were observed.

The values for lightness (L^*) decreased with increasing doses of both lightly toasted and oak chips after ageing of

Table 1. Changes in alcoholic strength by volume (ASV) and total extract content during maturation of plum distillate samples with oak chips

Parameter	Maturation variant	Control sample	$[m$(oak chip)$/V$(plum distillate)$]/$(g/L)											
			3				5				7			
			t(maturation)/month											
			3	6	9	12	3	6	9	12	3	6	9	12
ASV/%	A	55.43 ±0.05	55.35 ±0.02	55.15 ±0.02	55.05 ±0.03	54.95 ±0.02	55.45 ±0.02	55.05 ±0.02	55.00 ±0.03	54.95 ±0.02	55.30 ±0.02	55.15 ±0.02	55.05 ±0.03	54.85 ±0.02
	B	55.43 ±0.05	55.32 ±0.02	55.12 ±0.02	54.82 ±0.02	54.62 ±0.02	55.22 ±0.03	55.12 ±0.02	54.72 ±0.02	54.62 ±0.02	55.12 ±0.02	54.90 ±0.02	54.70 ±0.02	54.62 ±0.02
	C	55.43 ±0.05	55.15 ±0.02	54.95 ±0.02	54.75 ±0.01	54.75 ±0.02	55.15 ±0.02	55.05 ±0.02	54.85 ±0.01	54.75 ±0.02	55.05 ±0.01	54.95 ±0.02	54.80 ±0.01	54.75 ±0.02
	D	55.43 ±0.05	55.12 ±0.02	54.92 ±0.15	54.72 ±0.15	54.72 ±0.15	55.15 ±0.02	54.90 ±0.01	54.72 ±0.02	54.72 ±0.01	55.05 ±0.02	54.92 ±0.02	54.80 ±0.01	54.72 ±0.02
	E	55.43 ±0.05	55.05 ±0.03	54.82 ±0.02	54.62 ±0.02	54.62 ±0.01	55.05 ±0.03	54.72 ±0.02	54.65 ±0.02	54.62 ±0.01	54.90 ±0.03	54.75 ±0.02	54.65 ±0.02	54.62 ±0.01
	F	55.43 ±0.05	54.85 ±0.02	54.65 ±0.02	54.55 ±0.02	54.55 ±0.02	54.85 ±0.02	54.70 ±0.02	54.60 ±0.02	54.55 ±0.02	54.78 ±0.02	54.65 ±0.02	54.60 ±0.02	54.55 ±0.02
γ(total extract)/(g/L)	A	n.d.	0.75 ±0.05	1.25 ±0.07	1.50 ±0.05	1.55 ±0.05	1.25 ±0.05	1.75 ±0.05	2.25 ±0.05	2.35 ±0.05	1.75 ±0.05	2.15 ±0.05	2.35 ±0.05	2.50 ±0.05
	B	n.d.	0.45 ±0.02	1.15 ±0.02	1.25 ±0.03	1.35 ±0.05	0.65 ±0.05	1.35 ±0.03	1.55 ±0.05	1.55 ±0.03	0.75 ±0.03	1.35 ±0.05	1.65 ±0.05	1.80 ±0.05
	C	n.d.	0.90 ±0.05	1.35 ±0.05	1.75 ±0.05	2.00 ±0.05	1.45 ±0.05	1.75 ±0.05	2.05 ±0.05	2.65 ±0.05	1.75 ±0.05	2.15 ±0.05	2.75 ±0.05	2.94 ±0.05
	D	n.d.	1.28 ±0.05	1.55 ±0.05	1.95 ±0.05	2.15 ±0.05	1.48 ±0.05	1.95 ±0.05	2.30 ±0.05	2.86 ±0.05	1.80 ±0.05	2.28 ±0.05	2.80 ±0.05	2.95 ±0.05
	E	n.d.	1.25 ±0.05	1.60 ±0.05	1.90 ±0.05	2.10 ±0.05	1.65 ±0.05	1.85 ±0.05	2.25 ±0.05	2.22 ±0.05	1.85 ±0.05	2.35 ±0.05	2.85 ±0.05	3.05 ±0.05
	F	n.d.	0.75 ±0.05	1.15 ±0.07	1.30 ±0.05	1.60 ±0.05	1.15 ±0.05	1.65 ±0.05	2.05 ±0.05	2.25 ±0.05	1.65 ±0.05	2.05 ±0.05	2.35 ±0.05	2.40 ±0.05

Results of three-way ANOVA

Parameter	Effect of dose of lightly toasted French chips, temperature and time of maturation, and interaction among them (variants A, C and D, static maturation system)							Effect of dose of lightly toasted French chips, ageing system with agitation and time of maturation, and interaction among them (variants D and E, maturation at 45 °C)							Effect of dose and type of oak chips and time of maturation, and interaction among them (variants A and B, static maturation system, at 20 °C)							Effect of dose of lightly toasted French chips, ageing system with agitation and time of maturation, and interaction among them (variants A and F, maturation at 20 °C)						
	DC	T	MT	DC × T	DC × MT	T × MT	DC × T × MT	DC	AS	MT	DC × AS	DC × MT	AS × MT	DC × A × MT	DC	TC	MT	DC × TC	DC × MT	TC × MT	DC × TC × MT	DC	AS	MT	DC × AS	DC × MT	AS × MT	DC × AS × MT
ASV/%	ns	***	***	ns	ns	***	ns	ns	***	***	ns	**	**	ns	ns	ns	***	ns	*	***	*	ns	***	***	ns	ns	***	ns
γ(total extract)/(g/L)	***	***	***	***	***	***	***	***	**	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	ns	***	***	**

A=lightly toasted French oak chips, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week), B=French oak chips obtained after ageing of cognac, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week), C=lightly toasted French oak chips, 35 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 3 months, then matured under the same conditions as variant A), D=lightly toasted French oak chips, 45 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 6 months, then matured under the same conditions as variant A), E=lightly toasted French oak chips, 45 °C, a circulation system (samples heated and stirred simultaneously for 2 h per day, 5 days per week, for 6 months, then matured under the same conditions as variant A), F=lightly toasted French oak chips, 20 °C with ultrasound treatment (50 % amplitude for 20 min per day, 5 days per week, for 3 months, then matured under the same conditions as variant A); n.d.=not determined

DC=dose of oak chips (3, 5 and 7 g/L), T=temperature of maturation (18–20, 35 and 45 °C), MT=maturation time (0, 3, 6, 9 and 12 months), AS=ageing system with agitation (variants A and D=static, variant E=circulation, variant F=ultrasound treatment), TC=type of oak chips (lightly toasted French chips or French chips after ageing of cognac); ***p<0.001, **p<0.01, *p<0.05, ns=not significant

Table 2. CIELab chromatic parameters of plum distillate samples aged over 12 months with oak wood chips under various conditions

CIELab chromatic parameter	[m(oak chip)/V(plum distillate)]/(g/L)																	
	3						5						7					
	Maturation variant																	
	A	B	C	D	E	F	A	B	C	D	E	F	A	B	C	D	E	F
L^*	91.09 ±0.55	94.05 ±0.65	90.49 ±0.48	89.64 ±0.45	89.52 ±0.48	90.80 ±0.43	88.70 ±0.45	91.05 ±0.62	87.49 ±0.52	86.85 ±0.42	86.52 ±0.40	87.80 ±0.43	86.24 ±0.44	88.70 ±0.39	85.62 ±0.40	82.65 ±0.35	81.87 ±0.32	85.95 ±0.41
ΔL^*	−6.59 ±0.54	−3.63 ±0.64	−7.19 ±0.47	−8.04 ±0.45	−8.16 ±0.46	−6.88 ±0.41	−8.98 ±0.43	−6.63 ±0.62	−10.19 ±0.50	−10.83 ±0.40	−11.16 ±0.39	−9.88 ±0.41	−11.44 ±0.42	−8.98 ±0.37	−12.06 ±0.39	−15.03 ±0.34	−15.81 ±0.31	−11.73 ±0.40
a^*	−2.83 ±0.03	−1.47 ±0.01	−2.80 ±0.02	−2.73 ±0.02	−2.02 ±0.01	−2.53 ±0.02	−2.58 ±0.02	−1.27 ±0.02	−2.35 ±0.02	−2.22 ±0.02	−2.02 ±0.02	−2.33 ±0.02	−0.57 ±0.02	−0.58 ±0.02	0.19 ±0.02	0.22 ±0.02	0.24 ±0.02	−0.44 ±0.03
Δa^*	−2.69 ±0.02	−1.33 ±0.02	−2.66 ±0.02	−2.59 ±0.02	−1.88 ±0.01	−2.39 ±0.02	−2.44 ±0.02	−1.13 ±0.02	−2.21 ±0.02	−2.08 ±0.02	−1.88 ±0.02	−2.19 ±0.02	−0.43 ±0.02	−0.44 ±0.02	0.33 ±0.02	0.36 ±0.02	0.38 ±0.02	−0.30 ±0.02
b^*	26.42 ±0.12	11.48 ±0.05	30.55 ±0.08	36.64 ±0.06	36.87 ±0.06	27.50 ±0.04	37.20 ±0.05	21.48 ±0.03	31.79 ±0.03	36.64 ±0.03	37.87 ±0.04	27.50 ±0.02	46.55 ±0.06	37.20 ±0.04	47.23 ±0.05	47.66 ±0.05	47.72 ±0.04	45.78 ±0.04
Δb^*	25.99 ±0.12	11.05 ±0.05	30.12 ±0.08	36.21 ±0.06	36.44 ±0.06	27.07 ±0.04	36.77 ±0.05	21.05 ±0.03	31.36 ±0.03	36.21 ±0.03	37.44 ±0.04	27.07 ±0.02	46.12 ±0.06	36.77 ±0.04	46.8 ±0.05	47.23 ±0.05	47.29 ±0.04	45.35 ±0.05
ΔE	26.94 ±0.15	11.71 ±0.06	31.08 ±0.09	37.18 ±0.08	37.39 ±0.08	28.03 ±0.05	37.93 ±0.07	22.10 ±0.03	33.05 ±0.04	37.85 ±0.04	39.11 ±0.06	28.89 ±0.03	47.52 ±0.08	37.85 ±0.05	48.33 ±0.06	49.57 ±0.07	49.86 ±0.08	46.84 ±0.05

Results of two-way ANOVA

	Effect of dose of lightly toasted French chips and temperature, and interaction between them (variants A, C and D, static maturation system)			Effect of dose of lightly toasted French chips and ageing system with agitation, and interaction between them (variants D and E, maturation at 45 °C)			Effect of dose and type of oak chips, and interaction between them (variants A and B, static maturation system, at 20 °C)			Effect of dose of lightly toasted French chips and ageing system with agitation, and interaction between them (variants A and F, maturation at 20 °C)		
	DC	T	DC × T	DC	AS	DC × AS	DC	TC	DC × TC	DC	AS	DC × AS
L^*	***	***	**	***	*	*	***	***	ns	***	*	ns
ΔL^*	***	***	***	***	*	ns	***	***	ns	***	*	ns
a^*	***	***	***	***	***	***	***	***	***	***	***	***
Δa^*	***	***	***	***	***	***	***	***	***	***	***	***
b^*	***	***	***	***	***	***	***	***	***	***	***	***
Δb^*	***	***	***	***	***	***	***	***	***	***	***	***
ΔE	***	***	***	***	***	***	***	***	***	***	***	***

A–E=see Table 1, standard sample (plum distillate sample before maturation): $L^*_{standard}$=97.68, $a^*_{standard}$=−0.14, $b^*_{standard}$=0.43

DC=dose of oak chips (3, 5 and 7 g/L), T=temperature of maturation (18–20, 35 and 45 °C), AS=ageing system with agitation (variants A and D=static, variant E=circulation, variant F=ultrasound treatment), TC=type of oak chips (lightly toasted French chips or French chips after ageing of cognac); ***p<0.001, **p<0.01, *p<0.05, ns=not significant

cognac. Higher temperatures also resulted in lower values of L^*, associated with extraction of polyphenols (9). The samples matured with cognac barrel oak chips showed higher values of L^* (greater lightness) than analogous samples aged with fresh oak chips. This confirms the poorer composition of chips made from oak barrels previously used for storing cognac than lightly toasted French oak chips.

It is interesting to note that negative values of parameter a^* were found in the majority of samples. Positive values of this coordinate were obtained only for samples aged with 7 g/L of lightly toasted oak chips, periodically heated to 35 or 45 °C. In contrast, parameter b^* was positive in all variants, with the lowest value detected in the sample with oak chips after cognac ageing. Parameter b^* showed statistically significant differences depending on

a dose and type of oak chips (variants D and E). Significant effect of interaction between these two factors was also found. The addition of lightly toasted oak chips in a dose of 7 g/L of plum distillate resulted in higher value of parameter b^* than of samples matured with oak chips after cognac ageing. The use of circulation as well as ultrasound treatment during maturation had a significant effect on the higher value of parameter b^* than of analogous samples matured in static system.

Treatment with ultrasound in the first 3 months of maturation had no positive effect on the colour of the plum brandy. Based on the values of a^* and b^*, all the samples can be described as yellow, with slight hints of green or red. The values of ΔE^*, *i.e.* the total colour difference in relation to the control sample, were calculated using the same coordinates discussed above. All evaluated

factors had significant effect on Δ*E** values. The lowest Δ*E** values were observed for plum brandy samples aged with 3 g/L of oak chips (especially with barrel oak chips after cognac ageing), which increased with increasing dose of oak chips. Moreover, higher values of this factor were observed in samples aged with lightly toasted fresh oak chips. Application of periodical heating and circulation during maturation also had strong effect on higher values of Δ*E**. Pecić *et al.* (*9*), who tested the effect of maturation conditions on the sensory and antioxidant properties of Serbian plum brandies matured in casks for between 10 and 47 years, obtained significantly lower values of *L** and higher values of parameter *a**. Only the values of co-ordinate *b** were similar to those obtained in our study.

Changes in concentrations of phenolic compounds

Oak wood tissue is mostly made of polysaccharides such as cellulose, hemicelluloses and lignin, and is impregnated with tannins and small amounts of lipids (*35*). The main extractive components of wood are hydrolysable tannins and volatile compounds (*36*). Degradation of lignin in hydroalcoholic solutions results in the liberation of benzoic (vanillin, vanillic acid, syringaldehyde and syringic acid) and cinnamic (coniferaldehyde and sinapaldehyde) derivatives, which are the important phenolics for aroma and taste in aged brandies (*37*). The statistically significant effect of time×type of chips interaction was found only for vanillin content (p<0.05). After 12 months of maturation, in samples aged with cognac barrel oak chips, the vanillin concentration was lower than in the analogous samples aged with lightly toasted French oak chips (Fig. 1). No significant differences were observed for other phenolic compounds in plum distillate samples maturated with the two types of oak chips (p>0.05). In turn, temperature of maturation had statistically significant effect on the contents of all determined phenolic compounds (p<0.05). The largest increase was observed of sinapaldehyde content. After 3 months of maturation its concentration reached (14.48±0.72) mg/L (in distillate aged with lightly toasted fresh oak chips, heated periodically to 45 °C). Further changes in sinapaldehyde content were much less dynamic, and after 6 months sinapaldehyde concentrations were observed to decrease as a consequence of oxidation to syringaldehyde (*20*).

The conditions of maturation had a significant effect on the contents of all phenolic compounds determined in the study. Periodic circulation was found to improve coniferyl aldehyde and vanillin contents, which increased (p<0.05), in comparison with samples aged in static system. Treatment with ultrasound did not result in the release of more phenolics, with the exception of sinapaldehyde. The highest sinapaldehyde content after 12 months of maturation was found in the sample treated with ultrasound, probably as a consequence of the inhibition of oxidation to syringaldehyde by the ultrasound. Use of cognac barrel oak chips resulted in significantly lower concentrations of phenolic compounds, which is indicative of natural partial wood exhaustion.

Changes in concentrations of tannins

Tannins (soluble polyphenols with a molecular mass of 500–3000 Da) are among the most abundant constitu-
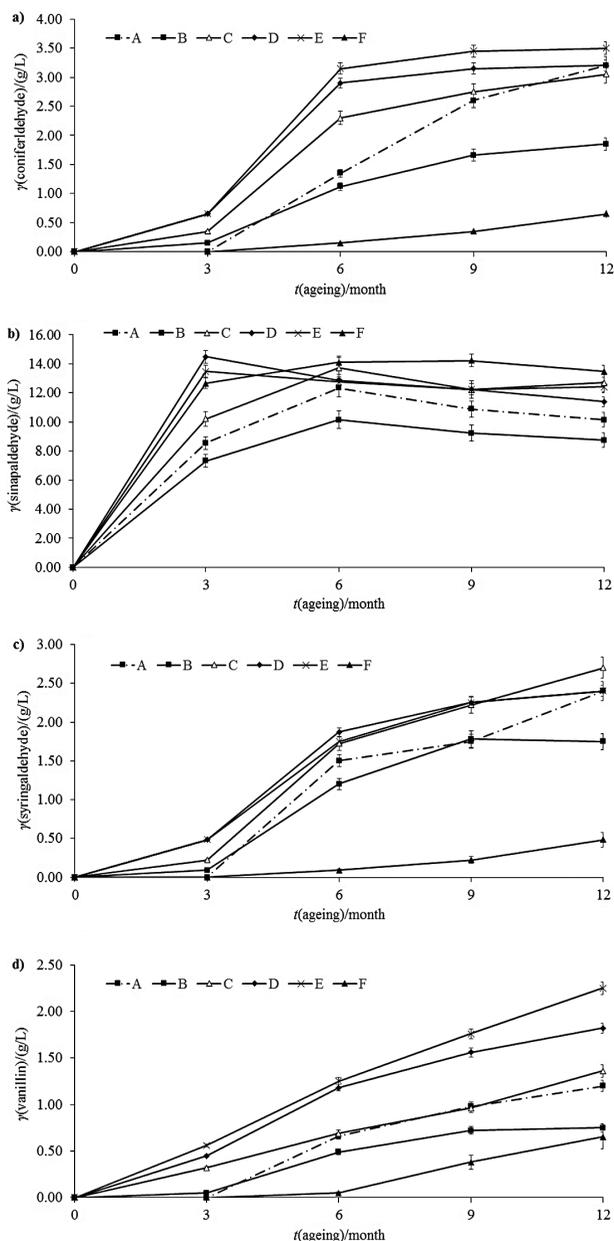

Fig. 1. Changes in concentrations of phenolic compounds: a) coniferaldehyde, b) sinapaldehyde, c) syringaldehyde, and d) vanillin during maturation of plum distillate samples under various conditions with added oak chips (5 g/L).
Designation of maturation variants:
A=lightly toasted French oak chips, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week), B=French oak chips obtained after ageing of cognac, 18–20 °C, a static system (samples were stirred for only 3 min per day, 5 days per week), C=lightly toasted French oak chips, 35 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 3 months, then matured under the same conditions as variant A), D=lightly toasted French oak chips, 45 °C, a static system (samples were stirred for only 3 min per day and then heated for 2 h per day, 5 days per week, for 6 months, then matured under the same conditions as variant A), E=lightly toasted French oak chips, 45 °C, a circulation system (samples heated and stirred simultaneously for 2 h per day, 5 days per week, for 6 months, then matured under the same conditions as variant A), F=lightly toasted French oak chips, 20 °C with ultrasound treatment (50 % amplitude for 20 min per day, 5 days per week, for 3 months, then matured under the same conditions as variant A)

ents of oak wood that are extracted into alcoholic beverages during ageing (*38*). Tannins can be classified according to their chemical structures into hydrolysable and condensed tannins. During ageing in oak barrels, the hydrolysable tannins extracted from oak wood due to the action of ethanol can be condensed with other compounds, such as polysaccharides and polypeptides (*39*).

During the maturation of plum distillate samples, increases in tannin content were observed. The increases were correlated both with the maturation time (maximum growth occurred in the first half of the year) and with the dose of added oak chips (p<0.05) (data shown only for 5 g/L of oak chips; see Fig. 2). Using so-called maderization, heating the samples to 35 °C caused an increase in tannins content (p<0.05), but further heating to 45 °C did not affect any changes (p>0.05). In samples aged with cognac barrel oak chips, the tannin concentration was approx. 50 % lower than in the analogous samples aged with lightly toasted fresh oak chips (Fig. 2) (p<0.05). One of ageing systems used in our study, *i.e.* periodic circulation, resulted in greater extraction of tannins, with concentrations after 6 months of ageing reaching approx. 80 % of the values determined after 12 months (p<0.05), while periodical treatment with ultrasound did not cause higher extraction of tannins from oak wood (p>0.05). Tao *et al.* (*40*) studied the release of oak-related compounds from oak chips during wine ageing. They used 25-kHz ultrasound waves to intensify the mass transfer of phenolics from oak chips into a model wine. Their results showed that acoustic energy density did not affect the total phenolic yield significantly, whereas increased transfer of phenolics was observed with temperature increases during sonication.
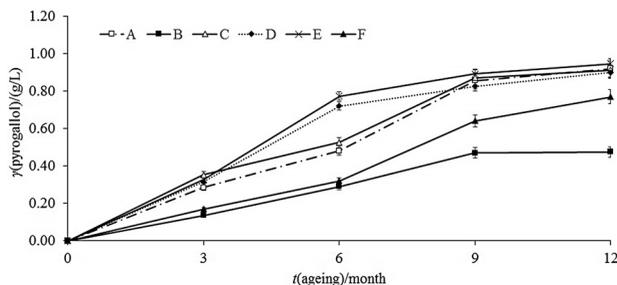

Fig. 2. Changes in the concentrations of tannins during maturation of plum distillate samples under various conditions with added oak chips (5 g/L). For designation of maturation variants A–F see Fig. 1

Changes in concentrations of sugars

The thermal degradation of cellulose and hemicelluloses that occurs during the toasting of oak wood, as well as in the processes of hydrolysis or ethanolysis which take place during ageing of hydroalcoholic solutions with oak wood leads to the release of various compounds with lower molecular mass (*5,40*). These include sugars such as glucose, xylose and arabinose (*41*). Glucose is one of the main sugars in aged whiskies (*22*). During the maturation of plum distillate samples, the liberation of sugars from oak wood chips was observed. The changes are presented in Fig. 3, using the results obtained from samples aged with a medium dose of oak chips as an example, *i.e.* 5 g/L
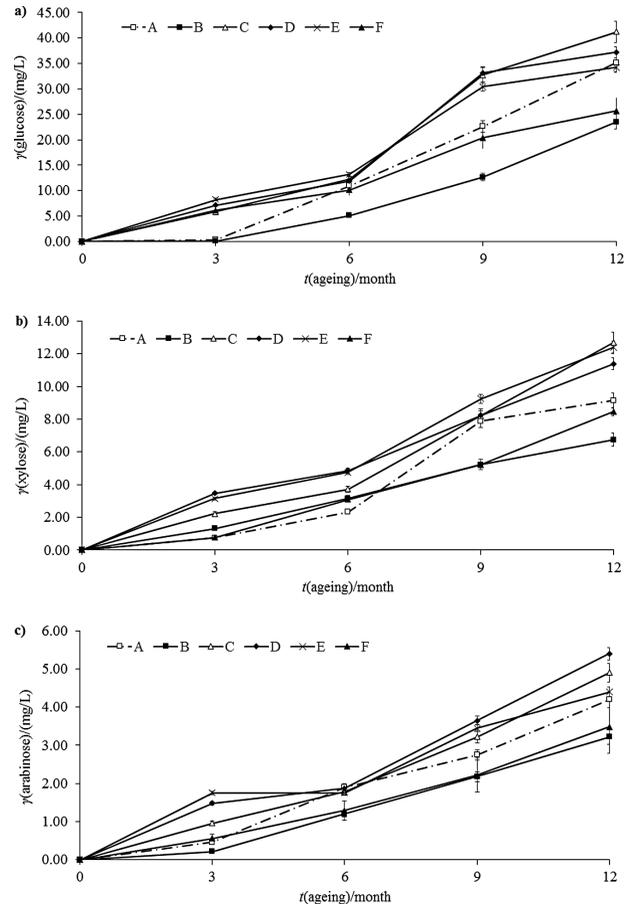

Fig. 3. Changes in the concentrations of sugars: a) glucose, b) xylose, c) arabinose during maturation of plum distillate samples under various conditions with added oak chips (5 g/L). For designation of maturation variants A–F see Fig. 1

of plum distillate. The changes in sugar content in samples aged with 3 and 7 g/L of oak chips (data not shown) were analogous to those observed with 5 g/L. The significantly different concentrations of sugars in maturated distillates were affected by all evaluated factors of maturation process (*i.e.* temperature, time and ageing system) as well as the interactions among them (p>0.001). During the first 3 months of maturation at 20 °C, no significant changes in glucose content were observed (p>0.05), while significant effect was found for time×temperature interaction (p<0.001). No clear differences were detected over subsequent months between the contents of sugars (p> 0.05) in the samples aged at 35 or 45 °C. After 12 months, glucose was present in the highest concentrations, of up to (41.20±2.47) mg/L, xylose concentration reached (12.70± 0.64) mg/L and the arabinose concentration was up to (5.43±0.03) mg/L. Periodical heating of aged plum distillate samples to 35 and 45 °C caused a decrease in glucose concentration, in comparison with trials maturated at room temperature. Moreover, in samples heated periodically to 45 °C (variants D and E), intense liberation of all sugars was observed (p>0.05) during maturation, but application of periodical circulation (variant E) did not cause an increase (p>0.05) in glucose, arabinose and xylose concentrations, in comparison with the same sample maturated in static system (variant D).

The type of chips and time of maturation were also used to evaluate changes of sugar content in matured distillate samples (variants A and B). With extended time of maturation, the sugar content increased (p<0.05), while differences between the types of chips were found only for arabinose (p<0.05).

Changes in concentrations of major volatile compounds

During maturation of alcoholic beverages, as well as the extraction and degradation of oak wood components, changes in the volatile compound content occur (37). Volatiles such as carbonyl compounds, esters, higher alcohols, organic acids and others (42), which are by-products of fermentation, are subjected to oxidation, reduction, esterification and transesterification during maturation (37).

Aldehydes and ketones, known as carbonyl compounds, are usually considered to have a negative influence on the quality of spirits. The concentrations of carbonyl compounds in agricultural distillates depend on the quality of the raw materials, their chemical composition, the conditions under which technological processes take place, the type of yeast species/strain involved in the fermentation (43) and the extent of microbial contamination (44). The quality of fruit distillates may also vary depending on the distillation conditions and the type of equipment used. The above-mentioned factors explain the differences in concentrations of volatile compounds (i.e. methanol, acetaldehyde, fusel alcohols and others) in the plum distillate used in our experiments when comparing with other published works (2,9,45).

In our study, decreases in acetaldehyde (Tables 3 and 4) were observed in all maturation variants (with the exception of the samples treated with ultrasound). These changes were affected mainly by the time, but not by the conditions of maturation. Increases in acetaldehyde content were observed in the samples treated with ultrasound, in comparison with distillate samples maturated in static system (variants A and F). These samples were also characterized by the largest decrease in ethanol content (Table 1). The lack of significant differences between the content of acetaldehyde in samples aged at 20 or 35 °C (variants A and C) can be interpreted as a consequence of the partial oxidation of ethanol at elevated temperatures (45). Mangas et al. (46) report that throughout maturation, acetaldehyde can be produced directly from ethanol, and its interaction with ethanol results in the production of acetal. This was confirmed by the results for acetaldehyde concentration revealed in our study (Table 3). Samples aged with cognac barrel oak chips had significantly higher concentrations of acetaldehyde than those aged with fresh lightly toasted oak chips, but no significant effect of the type of chips×dose of chips interaction was found (Tables 3 and 4).

Concentrations of isobutyraldehyde and isovaleraldehyde decreased with different intensities depending on the conditions of ageing. The application of periodical heating accelerated the decrease in aldehyde concentrations, especially isobutyraldehyde and isovaleraldehyde. However, the statistically significant changes of isobutyraldehyde were observed only in samples heated to 45 °C compared to those heated to 35 °C. Moreover, the effect of

neither the type nor the dose of oak chips on the content of aldehydes could be clearly interpreted.

One of the heterocyclic aldehydes that occur in distillates is furfural, which is mainly formed through the dehydration of pentoses at elevated temperatures (47). Furfural is produced during distillation involving Maillard reactions (48). Furanic compounds may also be produced by the degradation of sugars during the wood toasting (49). The only furanic compound detected in the fresh distillate was furfural. Periodical increase of the temperature, circulation, and in particular increasing the dose of oak wood chips resulted in higher furfural content in all of the samples (Table 3). The type of oak chips was also a significant factor. Furfural was more abundant in samples aged with lightly toasted fresh oak chips than in those aged with oak chips made from cognac barrels.

Esters form an important group of flavour compounds found in spirits, mostly comprising ethyl esters of monocarboxylic acids. Ethyl acetate is quantitatively the most important component in the ester fraction, usually accounting for over 50 % of the total. Many short-chain esters, such as isobutyl acetate, ethyl 3-methylbutyrate, ethyl n-butyrate, 2-methylbutyl acetate and 3-methylbutyl acetate, have fairly strong odours (50). Ethyl esters are formed during fermentation, and their concentrations increase during ageing (51). Plum distillates also contain esters of fatty acids C6–C10 (52). Several studies on distillates aged in oak have shown variations in these compounds during ageing, with distinct processes responsible in each case, such as hydrolysis, esterification or transesterification between compounds contained in the distillate and/or wood, or directly extracted from the wood (37). In our study, the predominant ester in the analysed plum distillate was ethyl acetate. The level of ethyl acetate was found to decrease over the course of maturation affected by the maturation time and temperature, but no interaction among these factors was observed. On the other hand, in samples treated with ultrasound, ethyl acetate content increased progressively during ageing (Table 5). This was in agreement with the findings of an earlier study (37). It is noteworthy that the changes in the concentration of this ester were not associated with either the dose or type of oak chips or with the maturation conditions (variants with static and circulation ageing systems). Only in samples aged with 7 g/L of lightly toasted oak chips, elevated temperatures were observed to have an effect on lowering the concentration of ethyl acetate after 12 months of maturation. Rodríguez Madrera et al. (53) suggest that its high initial concentration in fresh distillate causes partial hydrolysis of ethyl acetate, which increases as the acetic acid concentration rises (reflected in the amount of volatile acidity). Acetic acid may be extracted from wood, as already noted, or formed through the oxidation of ethanol (22).

Isoamyl acetate was significantly less reactive than ethyl acetate. Concentrations of isoamyl acetate did not change significantly over the 12-month period of maturation. There was no significant effect of all tested factors on its concentration. Rodríguez Madrera et al. (37) compared ageing methods in stainless steel vessels with oak wood staves of different origins and traditional ageing in oak wood barrels, revealing decreases in isoamyl acetate con-

Table 3. Changes in the concentrations of major carbonyl compounds during maturation of plum distillate samples with oak chips under various conditions

Compound	Maturation variant	Control sample	[m(oak chip)/V(plum distillate)]/(g/L)											
			3				5				7			
			t(maturation)/month											
			3	6	9	12	3	6	9	12	3	6	9	12
			γ/(mg/L of absolute alcohol)											
Acetaldehyde	A	69.37 ±3.25	63.92 ±1.15	57.26 ±1.21	49.75 ±1.32	47.96 ±1.15	61.92 ±1.25	56.26 ±1.15	49.96 ±1.22	50.16 ±0.75	57.93 ±1.05	53.55 ±0.82	50.96 ±0.76	52.81 ±0.69
	B	69.37 ±3.25	65.70 ±1.05	63.70 ±0.95	60.65 ±1.12	61.70 ±0.75	66.25 ±1.15	60.70 ±0.68	60.15 ±1.12	60.85 ±1.05	65.22 ±0.85	66.70 ±1.05	63.24 ±0.72	60.48 ±0.84
	C	69.37 ±3.25	60.13 ±1.35	60.89 ±0.75	58.13 ±1.35	55.13 ±0.75	60.66 ±1.32	57.89 ±0.55	56.42 ±0.85	53.22 ±0.48	60.82 ±0.75	58.25 ±0.84	57.13 ±0.66	53.22 ±0.75
	D	69.37 ±3.25	61.13 ±1.26	55.13 ±0.82	54.18 ±0.69	51.13 ±0.66	63.22 ±0.85	60.13 ±0.74	56.18 ±0.62	53.13 ±0.39	60.40 ±0.66	54.22 ±0.74	51.13 ±0.66	50.05 ±0.66
	E	69.37 ±3.25	60.31 ±0.55	62.31 ±0.45	56.31 ±1.12	52.31 ±0.55	58.42 ±0.55	53.22 ±0.39	52.35 ±0.26	50.26 ±0.72	60.96 ±0.55	54.31 ±0.66	52.31 ±0.55	52.31 ±0.70
	F	69.37 ±3.25	65.37 ±1.42	70.37 ±3.15	73.37 ±1.15	75.37 ±1.42	67.22 ±0.72	71.53 ±0.82	70.12 ±1.15	72.26 ±0.86	70.25 ±1.33	72.37 ±0.82	71.45 ±1.36	72.32 ±1.15
Isobutyraldehyde	A	5.15 ±0.15	4.86 ±0.07	4.24 ±0.06	4.05 ±0.05	3.82 ±0.05	4.78 ±0.07	4.22 ±0.03	4.15 ±0.05	4.09 ±0.05	4.86 ±0.07	4.24 ±0.06	4.05 ±0.05	3.93 ±0.03
	B	5.15 ±0.15	4.97 ±0.08	4.63 ±0.05	4.15 ±0.05	4.04 ±0.04	4.92 ±0.05	4.55 ±0.03	4.21 ±0.05	4.16 ±0.04	5.03 ±0.07	4.61 ±0.05	4.15 ±0.05	4.04 ±0.04
	C	5.15 ±0.15	4.45 ±0.04	4.05 ±0.04	3.78 ±0.03	3.61 ±0.03	4.48 ±0.04	4.07 ±0.03	3.72 ±0.03	3.55 ±0.04	4.65 ±0.04	4.15 ±0.03	3.85 ±0.03	3.55 ±0.03
	D	5.15 ±0.15	4.40 ±0.05	4.15 ±0.06	3.65 ±0.04	3.50 ±0.04	4.45 ±0.05	4.18 ±0.03	3.71 ±0.03	3.43 ±0.04	4.40 ±0.05	4.26 ±0.06	3.65 ±0.04	3.50 ±0.04
	E	5.15 ±0.15	4.50 ±0.05	4.17 ±0.03	4.00 ±0.04	3.46 ±0.04	4.55 ±0.05	4.26 ±0.03	3.92 ±0.04	3.22 ±0.03	4.39 ±0.04	4.16 ±0.03	3.72 ±0.03	3.46 ±0.04
	F	5.15 ±0.15	4.75 ±0.05	4.44 ±0.04	4.25 ±0.05	3.88 ±0.05	4.68 ±0.05	4.40 ±0.04	4.32 ±0.03	3.82 ±0.03	4.82 ±0.05	4.34 ±0.04	4.27 ±0.05	3.83 ±0.05
Isovaleraldehyde	A	1.22 ±0.02	1.05 ±0.02	1.00 ±0.02	0.95 ±0.02	0.87 ±0.02	1.10 ±0.02	0.95 ±0.02	0.95 ±0.02	0.92 ±0.02	1.11 ±0.02	0.97 ±0.02	0.95 ±0.02	1.00 ±0.02
	B	1.22 ±0.02	1.13 ±0.02	1.02 ±0.02	0.85 ±0.02	0.78 ±0.02	1.15 ±0.02	1.05 ±0.02	0.95 ±0.02	0.97 ±0.02	1.10 ±0.02	1.05 ±0.02	0.97 ±0.02	0.93 ±0.02
	C	1.22 ±0.02	1.05 ±0.02	0.97 ±0.02	0.92 ±0.02	0.83 ±0.02	1.02 ±0.02	0.96 ±0.02	0.93 ±0.02	0.85 ±0.02	1.11 ±0.02	0.98 ±0.02	0.93 ±0.02	0.85 ±0.02
	D	1.22 ±0.02	1.08 ±0.02	1.02 ±0.01	0.95 ±0.02	0.95 ±0.01	1.06 ±0.02	0.97 ±0.01	0.94 ±0.02	0.90 ±0.02	1.03 ±0.02	0.92 ±0.01	0.90 ±0.02	0.90 ±0.02
	E	1.22 ±0.02	1.15 ±0.01	1.12 ±0.02	1.15 ±0.02	1.17 ±0.01	1.12 ±0.02	1.05 ±0.02	0.98 ±0.01	0.95 ±0.01	1.07 ±0.02	0.95 ±0.02	0.98 ±0.01	0.95 ±0.01
	F	1.22 ±0.02	1.22 ±0.02	1.24 ±0.02	1.20 ±0.02	1.25 ±0.02	1.15 ±0.02	1.10 ±0.02	1.10 ±0.02	1.15 ±0.02	1.17 ±0.02	1.15 ±0.02	1.15 ±0.02	1.17 ±0.02
Furfural	A	3.46 ±0.06	3.48 ±0.05	3.52 ±0.06	3.55 ±0.05	3.57 ±0.06	3.65 ±0.04	3.82 ±0.04	4.25 ±0.05	4.57 ±0.03	4.28 ±0.05	5.12 ±0.04	5.55 ±0.05	5.80 ±0.05
	B	3.46 ±0.06	3.48 ±0.05	3.43 ±0.05	3.36 ±0.05	3.48 ±0.04	3.58 ±0.05	3.63 ±0.05	3.76 ±0.04	3.95 ±0.03	3.55 ±0.05	3.75 ±0.05	3.96 ±0.05	3.90 ±0.05
	C	3.46 ±0.06	3.58 ±0.05	3.76 ±0.04	4.06 ±0.05	4.21 ±0.04	3.78 ±0.05	3.96 ±0.04	4.26 ±0.03	4.76 ±0.03	4.15 ±0.05	4.72 ±0.04	5.23 ±0.04	5.66 ±0.04
	D	3.46 ±0.06	3.66 ±0.05	3.95 ±0.04	4.25 ±0.05	4.45 ±0.05	4.05 ±0.03	4.22 ±0.03	4.55 ±0.05	4.85 ±0.04	4.36 ±0.05	4.95 ±0.04	5.27 ±0.05	5.80 ±0.05
	E	3.46 ±0.06	3.94 ±0.05	4.26 ±0.05	4.55 ±0.04	4.78 ±0.05	4.12 ±0.05	4.26 ±0.05	4.60 ±0.04	4.78 ±0.04	4.64 ±0.04	5.26 ±0.05	5.72 ±0.04	5.87 ±0.05
	F	3.46 ±0.06	3.45 ±0.05	3.60 ±0.04	3.65 ±0.05	3.70 ±0.05	3.55 ±0.04	3.60 ±0.04	3.68 ±0.03	4.26 ±0.05	3.95 ±0.05	4.27 ±0.04	4.65 ±0.05	4.65 ±0.03

A–E=see Table 1

Table 4. Results of three-way ANOVA for major volatile compound content in plum distillate samples after maturation with oak chips under various conditions

Compound	Effect of dose of lightly toasted French chips, temperature and time of maturation, and interaction among them (variants A, C and D, static maturation system)							Effect of dose of lightly toasted French chips, ageing system with agitation and time of maturation, and interaction among them (variants D and E, maturation at 45 °C)							Effect of dose and type of oak chips, and time of maturation, and interaction among them (variants A and B, static maturation system, at 20 °C)							Effect of dose of lightly toasted French chips, ageing system with agitation and time of maturation, and interaction among them (variants A and F, maturation at 20 °C)						
	DC	T	MT	DC×T	DC×MT	T×MT	DC×T×MT	DC	AS	MT	DC×AS	DC×MT	AS×MT	DC×AS×MT	DC	TC	MT	DC×TC	DC×MT	TC×MT	DC×TC×MT	DC	AS	MT	DC×AS	DC×MT	AS×MT	DC×AS×MT
Acetaldehyde	*	***	*	*	***	***	ns	***	ns	***	***	*	ns	***	ns	***	***	ns	**	***	***	ns	***	***	ns	ns	***	***
Isobutyraldehyde	ns	***	***	ns	ns	***	ns	ns	*	***	*	***	***	ns	ns	***	***	ns	**	***	ns	ns	ns	***	ns	ns	***	ns
Isovaleraldehyde	ns	***	***	***	***	***	***	***	***	***	***	***	***	***	***	*	***	***	***	***	***	***	***	***	***	***	***	***
Furfural	***	***	***	***	***	***	*	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***	***
Ethyl acetate	ns	***	***	ns	ns	*	ns	ns	*	***	ns	ns	ns	ns	ns	ns	***	ns	ns	ns	ns	ns	***	***	ns	**	***	ns
Isoamyl acetate	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
Ethyl hexanoate	ns	***	***	ns	ns	***	ns	ns	*	***	**	***	***	***	ns	***	***	ns	ns	***	*	***	***	***	**	***	***	**
Ethyl octanoate	ns	ns	***	ns	ns	ns	ns	***	***	***	**	***	***	***	ns	ns	***	ns	ns	ns	ns	ns	***	***	ns	ns	**	ns
Ethyl decanoate	ns	***	***	*	ns	***	ns	ns	ns	***	ns	ns	ns	ns	ns	***	***	ns	ns	***	ns	***	ns	***	*	ns	ns	*
Methanol	***	***	***	***	***	***	**	***	***	***	ns	***	***	*	***	ns	***	ns	*	ns	ns	***	***	***	***	ns	***	ns
1-Propanol	*	***	***	***	***	***	***	***	*	***	ns	***	*	ns	***	***	***	***	***	***	*	***	***	***	***	**	***	***
1-Butanol	***	ns	***	ns	***	ns	ns	ns	ns	***	ns	ns	ns	ns	ns	***	***	ns	ns	***	ns	**	***	***	**	**	***	*
2-Methyl-1-propanol	***	***	***	***	***	**	**	***	***	***	***	*	***	***	***	***	***	ns	***	***	ns	ns	ns	***	**	**	***	*
2-Methyl-1-butanol	***	***	***	**	***	***	*	**	ns	***	***	**	ns	*	ns	***	***	ns	ns	***	**	ns	**	***	ns	***	***	ns
3-Methyl-1-butanol	ns	***	***	ns	ns	***	ns	***	ns	***	ns	**	ns	ns	ns	***	***	**	ns	***	ns	***	ns	***	ns	**	*	ns
Hexanol	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
2-Phenylethanol	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns

DC=dose of oak chips (3, 5 and 7 g/L), T=temperature of maturation (18–20, 35 and 45 °C), MT=maturation time (0, 3, 6, 9, and 12 months), AS=ageing system with agitation (variants A and D=static, variant E=circulation, variant F=ultrasound treatment), TC=type of oak chips (lightly toasted French chips or French chips after ageing of cognac); ***p <0.001, **p<0.01, *p<0.05, ns=not significant

tent of up to approx. 30 % in relation to control samples. The application of micro-oxygenation resulted in more intense changes than traditional ageing.

Ethyl esters of medium chain fatty acids, i.e. hexanoic, octanoic, decanoic and dodecanoic acids, are of particular interest, on account of their relatively high levels in fermented beverages and spirits and the fruity odours they give to products (37). In our study, the concentrations of ethyl caproate, ethyl caprylate and ethyl caprate were found to gradually increase in all samples. The observed changes were affected by maturation time. Significant effect was found for time×temperature interaction, for compounds such as ethyl hexanoate and ethyl decanoate (Table 4). Unfortunately, no clear correlation was found with the type or dose of oak chips, or with maturation conditions. The only exception were the samples matured with cognac barrel oak chips, in which the concentration of ethyl caprate was significantly higher than in the distillate matured with lightly toasted oak chips. It may be supposed that the elevated concentration of this compound (described as having a 'fruity, reminiscent of cognac' smell) originated from the porous oak barrels, which would have been impregnated with cognac aroma compounds, which were then liberated during maturation of the plum distillate.

Methanol is an undesirable compound in spirit distillates. While methanol does not directly affect the flavour of the distillate, it is subjected to restrictive controls, owing to its high toxicity (54). In our study, the concentration of methanol was observed to decrease in the majority of samples (Tables 4 and 6), as a consequence of sorption and esterification reactions (46). Only treatment with ultrasound led to a significant increase in methanol content in the first 3 months of ageing, after which there were no significant further changes. During treatment with ultrasound, methanol may be released from oak wood chips principally by demethylation of lignin, hemicellulose or xylans (40,55). The obtained results revealed no significant effect of the type of oak chips on methanol concentration (Table 6).

Table 5. Changes in the concentrations of major esters during maturation of plum distillate samples with oak chips under various conditions

Compound	Maturation variant	Control sample	[m(oak chip)/V(plum distillate)]/(g/L)											
			3				5				7			
			t(maturation)/month											
			3	6	9	12	3	6	9	12	3	6	9	12
			γ/(mg/L of absolute alcohol)											
Ethyl acetate	A	132.20 ±5.25	127.20 ±3.48	125.20 ±3.26	116.20 ±3.50	108.70 ±3.33	129.20 ±3.82	122.20 ±3.15	114.20 ±3.33	108.82 ±3.12	125.20 ±3.33	119.20 ±3.45	116.20 ±3.50	115.46 ±3.65
	B	132.20 ±5.25	130.20 ±4.50	124.20 ±3.35	118.20 ±2.25	114.87 ±3.15	132.20 ±3.42	126.20 ±3.15	116.20 ±2.42	109.87 ±3.15	127.20 ±3.50	122.20 ±2.78	115.48 ±2.25	111.87 ±3.15
	C	132.20 ±5.25	122.20 ±2.36	113.20 ±3.27	107.20 ±2.25	105.20 ±2.68	126.20 ±2.22	115.20 ±2.27	110.20 ±2.72	108.20 ±2.48	120.20 ±2.19	116.20 ±3.15	109.20 ±2.25	105.44 ±2.85
	D	132.20 ±5.25	124.22 ±3.25	112.20 ±2.25	109.20 ±2.75	106.55 ±2.28	120.37 ±3.52	114.27 ±2.29	106.20 ±2.55	105.55 ±2.27	124.22 ±3.25	112.20 ±2.42	109.20 ±2.75	106.55 ±2.28
	E	132.20 ±5.25	123.51 ±3.42	116.20 ±3.25	111.20 ±2.65	108.65 ±2.77	124.55 ±3.17	115.20 ±3.19	114.09 ±2.35	107.77 ±2.77	121.42 ±3.26	113.66 ±3.77	110.77 ±2.49	105.65 ±2.48
	F	132.20 ±5.25	184.20 ±6.22	178.20 ±4.66	176.20 ±4.25	172.44 ±5.33	180.55 ±4.65	178.20 ±4.15	173.20 ±3.25	172.44 ±3.33	164.20 ±6.22	175.20 ±4.82	173.45 ±4.27	172.44 ±5.33
Isoamyl acetate	A	23.71 ±0.98	23.51 ±0.64	23.62 ±0.68	24.32 ±0.74	23.70 ±0.39	23.66 ±0.68	23.91 ±0.78	23.15 ±0.47	22.95 ±0.98	23.55 ±0.68	24.15 ±0.88	23.71 ±0.72	24.34 ±0.98
	B	23.71 ±0.98	23.31 ±0.58	23.54 ±0.76	23.77 ±0.78	24.34 ±0.58	23.77 ±0.68	23.56 ±0.91	23.79 ±0.78	24.22 ±0.68	23.63 ±0.66	23.51 ±0.58	24.35 ±0.69	24.66 ±0.58
	C	23.71 ±0.98	23.55 ±0.64	23.67 ±0.58	24.59 ±0.68	24.66 ±0.49	23.31 ±0.68	23.85 ±0.73	23.77 ±0.68	24.39 ±0.76	23.77 ±0.68	23.66 ±0.77	23.75 ±0.85	23.82 ±0.76
	D	23.71 ±0.98	23.76 ±0.68	24.15 ±0.84	23.94 ±0.85	23.82 ±0.58	23.55 ±0.77	23.76 ±0.84	24.15 ±0.68	23.42 ±0.77	23.65 ±0.88	23.94 ±0.82	23.77 ±0.59	23.61 ±0.68
	E	23.71 ±0.98	23.77 ±0.78	24.72 ±0.84	23.77 ±0.69	23.61 ±0.65	23.74 ±0.88	24.27 ±0.76	23.92 ±0.94	23.54 ±0.68	23.76 ±0.68	24.72 ±0.74	23.56 ±0.69	23.39 ±0.68
	F	23.71 ±0.98	24.42 ±0.88	24.33 ±0.76	23.77 ±0.82	23.39 ±0.59	23.92 ±0.94	23.76 ±0.82	23.52 ±0.94	23.39 ±0.48	24.55 ±0.93	23.97 ±0.74	23.73 ±0.88	23.50 ±0.77
Ethyl hexanoate	A	16.25 ±0.02	16.44 ±0.02	16.57 ±0.02	16.74 ±0.03	16.97 ±0.03	16.51 ±0.02	16.59 ±0.02	16.77 ±0.03	16.89 ±0.03	16.47 ±0.02	16.66 ±0.02	16.78 ±0.03	16.97 ±0.05
	B	16.25 ±0.02	16.37 ±0.03	16.62 ±0.02	16.66 ±0.02	16.79 ±0.02	16.32 ±0.02	16.59 ±0.02	16.61 ±0.02	16.82 ±0.03	16.41 ±0.03	16.65 ±0.02	16.69 ±0.02	16.79 ±0.02
	C	16.25 ±0.02	16.57 ±0.03	16.78 ±0.03	16.95 ±0.03	17.16 ±0.03	16.59 ±0.03	16.84 ±0.03	16.91 ±0.03	17.23 ±0.03	16.62 ±0.03	16.85 ±0.03	16.93 ±0.03	17.10 ±0.03
	D	16.25 ±0.02	16.61 ±0.03	16.74 ±0.03	16.80 ±0.02	16.96 ±0.03	16.66 ±0.03	16.79 ±0.03	16.84 ±0.02	16.89 ±0.03	16.54 ±0.03	16.77 ±0.03	16.84 ±0.02	16.92 ±0.03
	E	16.25 ±0.02	16.55 ±0.02	16.75 ±0.02	16.93 ±0.03	16.88 ±0.03	16.62 ±0.02	16.71 ±0.02	16.83 ±0.03	16.84 ±0.03	16.51 ±0.02	16.77 ±0.02	16.85 ±0.03	16.94 ±0.03
	F	16.25 ±0.02	16.17 ±0.03	16.27 ±0.02	16.52 ±0.03	16.73 ±0.03	16.20 ±0.02	16.34 ±0.02	16.45 ±0.02	16.70 ±0.01	16.22 ±0.02	16.46 ±0.02	16.75 ±0.02	16.73 ±0.02
Ethyl octanoate	A	2.84 ±0.03	3.15 ±0.03	3.47 ±0.04	3.66 ±0.04	3.71 ±0.04	3.05 ±0.03	3.52 ±0.03	3.61 ±0.04	3.75 ±0.04	3.22 ±0.03	3.50 ±0.04	3.65 ±0.04	3.70 ±0.04
	B	2.84 ±0.03	3.36 ±0.03	3.64 ±0.04	3.84 ±0.04	3.90 ±0.04	3.39 ±0.03	3.54 ±0.04	3.77 ±0.04	3.75 ±0.04	3.16 ±0.03	3.43 ±0.04	3.74 ±0.04	3.90 ±0.04
	C	2.84 ±0.03	3.44 ±0.04	3.81 ±0.04	3.84 ±0.04	3.95 ±0.04	3.49 ±0.04	3.75 ±0.04	3.86 ±0.04	3.90 ±0.04	3.34 ±0.04	3.69 ±0.04	3.85 ±0.04	3.87 ±0.04
	D	2.84 ±0.03	3.28 ±0.03	3.84 ±0.04	3.91 ±0.04	3.44 ±0.04	3.18 ±0.03	3.81 ±0.04	3.96 ±0.04	3.44 ±0.04	3.37 ±0.03	3.85 ±0.04	3.91 ±0.04	3.86 ±0.04
	E	2.84 ±0.03	2.98 ±0.03	3.66 ±0.03	3.84 ±0.03	3.85 ±0.03	3.08 ±0.03	3.54 ±0.03	3.88 ±0.03	3.90 ±0.03	3.28 ±0.03	3.66 ±0.03	3.84 ±0.03	3.93 ±0.03
	F	2.84 ±0.03	2.74 ±0.03	2.88 ±0.03	3.14 ±0.03	3.35 ±0.03	2.80 ±0.03	2.86 ±0.03	3.25 ±0.03	3.39 ±0.03	2.90 ±0.03	2.88 ±0.03	3.26 ±0.03	3.45 ±0.03
Ethyl decanoate	A	13.30 ±0.15	13.26 ±0.15	13.75 ±0.18	14.12 ±0.22	14.29 ±0.25	13.42 ±0.15	13.70 ±0.17	14.06 ±0.16	14.15 ±0.16	13.40 ±0.15	13.83 ±0.17	14.30 ±0.25	14.79 ±0.23
	B	13.30 ±0.15	13.69 ±0.17	14.30 ±0.25	15.30 ±0.23	15.86 ±0.25	13.63 ±0.15	14.34 ±0.16	15.42 ±0.19	15.80 ±0.15	13.55 ±0.15	14.72 ±0.22	15.25 ±0.23	15.88 ±0.22
	C	13.30 ±0.15	13.64 ±0.16	13.82 ±0.15	14.07 ±0.18	14.18 ±0.18	13.72 ±0.16	13.99 ±0.16	14.12 ±0.18	14.20 ±0.22	13.55 ±0.16	13.80 ±0.15	14.13 ±0.18	14.18 ±0.17
	D	13.30 ±0.15	13.50 ±0.15	13.87 ±0.14	14.30 ±0.15	14.47 ±0.17	13.68 ±0.15	13.96 ±0.18	14.35 ±0.18	14.47 ±0.17	13.52 ±0.15	13.83 ±0.16	14.39 ±0.18	14.69 ±0.21
	E	13.30 ±0.15	13.66 ±0.13	14.17 ±0.15	14.37 ±0.16	14.52 ±0.20	13.78 ±0.15	13.98 ±0.17	14.33 ±0.16	14.57 ±0.18	13.49 ±0.15	13.77 ±0.16	14.29 ±0.19	14.58 ±0.20
	F	13.30 ±0.15	13.43 ±0.16	13.89 ±0.18	14.30 ±0.15	14.76 ±0.16	13.19 ±0.15	13.47 ±0.16	13.84 ±0.17	14.56 ±0.21	13.62 ±0.17	13.94 ±0.15	14.09 ±0.15	14.56 ±0.13

A−E=see Table 1

Table 6. Changes in the concentrations of alcohols during maturation of plum distillate samples with oak chips under various conditions

Compound	Maturation variant	Control sample	[m(oak chip)/V(plum distillate)]/(g/L)											
			3				5				7			
			t(maturation)/month											
			3	6	9	12	3	6	9	12	3	6	9	12
			γ/(mg/L of absolute alcohol)											
Methanol	A	115.8 ±1.6	114.5 ±1.2	114.3 ±1.2	105.9 ±0.9	103.7 ±0.8	115.2 ±1.2	114.7 ±1.2	108.9 ±0.9	105.5 ±0.6	115.5 ±1.3	114.6 ±1.1	107.9 ±0.8	105.6 ±0.8
	B	115.8 ±1.6	114.5 ±1.2	114.3 ±1.2	105.9 ±0.9	103.7 ±0.8	115.2 ±1.2	114.7 ±1.2	108.9 ±0.9	105.5 ±0.6	115.5 ±1.3	114.6 ±1.1	107.9 ±0.8	108.3 ±0.8
	C	115.8 ±1.6	110.5 ±1.2	104.3 ±1.2	105.9 ±0.9	103.7 ±0.8	111.5 ±1.2	107.3 ±1.2	105.6 ±0.9	103.3 ±0.8	110.5 ±1.2	104.3 ±1.2	105.9 ±0.9	101.7 ±0.8
	D	115.8 ±1.6	108.5 ±1.2	102.3 ±1.2	98.9 ±0.9	93.7 ±0.8	110.5 ±1.2	108.3 ±1.2	103.9 ±0.9	96.7 ±0.8	111.5 ±1.2	102.3 ±1.2	98.9 ±0.9	95.6 ±0.8
	E	115.8 ±1.6	108.9 ±1.2	97.3 ±1.2	97.9 ±0.9	94.2 ±0.8	110.9 ±1.2	105.3 ±1.2	102.9 ±0.9	92.7 ±0.8	108.9 ±1.2	97.3 ±1.2	97.9 ±0.9	95.3 ±0.8
	F	115.8 ±1.6	120.5 ±1.2	119.3 ±1.2	120.9 ±1.5	120.7 ±1.8	123.2 ±1.2	124.7 ±1.2	124.9 ±1.9	123.5 ±1.6	125.2 ±1.3	125.7 ±1.5	124.9 ±1.9	125.5 ±2.6
1-Propanol	A	397.3 ±3.4	395.9 ±3.8	394.8 ±4.5	356.1 ±6.8	337.3 ±7.2	389.7 ±6.4	356.2 ±3.7	340.8 ±6.6	329.6 ±4.7	393.5 ±6.4	372.2 ±6.5	350.3 ±7.1	348.8 ±5.4
	B	397.3 ±3.4	398.9 ±5.2	387.8 ±6.1	376.1 ±6.8	357.3 ±6.2	390.7 ±6.6	376.2 ±5.5	364.8 ±6.6	349.6 ±5.3	383.5 ±5.4	366.2 ±4.3	357.3 ±5.1	354.6 ±4.8
	C	397.3 ±3.4	378.9 ±4.5	356.8 ±4.5	349.1 ±6.8	343.5 ±5.2	379.7 ±6.4	366.2 ±5.3	352.8 ±6.6	339.6 ±4.7	379.5 ±7.4	372.2 ±6.5	350.3 ±7.1	346.3 ±6.3
	D	397.3 ±3.4	375.6 ±3.7	364.8 ±4.5	356.1 ±6.8	337.3 ±7.2	382.7 ±6.4	359.2 ±5.7	340.8 ±6.6	329.6 ±4.7	393.5 ±7.4	377.2 ±8.2	363.3 ±7.7	348.2 ±5.4
	E	397.3 ±3.4	370.9 ±5.2	354.8 ±4.5	346.1 ±6.8	335.8 ±6.7	374.9 ±56.1	359.8 ±4.3	343.1 ±5.5	339.6 ±4.4	383.5 ±6.4	376.2 ±6.5	350.3 ±7.1	348.5 ±5.9
	F	397.3 ±3.4	390.8 ±4.5	376.1 ±5.4	362.3 ±5.2	359.7 ±5.4	393.6 ±4.7	382.1 ±5.8	369.3 ±6.7	355.7 ±7.4	392.4 ±4.7	374.1 ±6.3	364.8 ±6.6	355.8 ±5.3
1-Butanol	A	6.35 ±0.16	6.32 ±0.15	5.11 ±0.13	4.86 ±0.12	4.80 ±0.13	6.18 ±0.15	5.15 ±0.13	4.96 ±0.12	4.84 ±0.13	6.14 ±0.14	5.22 ±0.11	4.96 ±0.12	4.77 ±0.14
	B	6.35 ±0.16	6.21 ±0.15	5.86 ±0.13	5.54 ±0.12	5.33 ±0.14	6.30 ±0.15	5.86 ±0.13	5.54 ±0.12	5.33 ±0.14	6.18 ±0.15	5.86 ±0.12	5.54 ±0.13	5.33 ±0.12
	C	6.35 ±0.16	5.42 ±0.14	5.21 ±0.13	4.77 ±0.13	4.80 ±0.13	5.35 ±0.14	5.11 ±0.12	4.91 ±0.12	4.84 ±0.13	5.37 ±0.15	5.16 ±0.13	4.76 ±0.12	4.70 ±0.13
	D	6.35 ±0.16	5.29 ±0.15	5.08 ±0.13	4.76 ±0.12	4.65 ±0.13	5.34 ±0.13	4.88 ±0.12	4.76 ±0.11	4.66 ±0.11	5.49 ±0.15	5.11 ±0.13	4.73 ±0.12	4.66 ±0.11
	E	6.35 ±0.16	5.34 ±0.12	5.17 ±0.10	4.72 ±0.09	4.75 ±0.11	5.26 ±0.12	4.93 ±0.12	4.76 ±0.1	4.69 ±0.08	5.22 ±0.11	4.93 ±0.09	4.81 ±0.08	4.80 ±0.13
	F	6.35 ±0.16	6.12 ±0.11	5.66 ±0.11	4.85 ±0.09	4.76 ±0.11	6.32 ±0.15	6.11 ±0.12	5.24 ±0.09	4.86 ±0.08	6.14 ±0.13	5.56 ±0.09	5.37 ±0.09	4.84 ±0.09
2-Methyl-1-propanol	A	483.3 ±5.8	475.3 ±5.5	469.8 ±5.6	466.7 ±4.6	441.6 ±5.2	478.3 ±5.3	463.4 ±4.8	454.4 ±4.5	431.5 ±5.1	477.5 ±5.5	471.9 ±5.2	448.4 ±4.8	425.6 ±5.1
	B	483.3 ±5.8	480.3 ±5.5	479.8 ±4.8	476.7 ±4.9	469.6 ±3.5	478.3 ±4.8	473.4 ±3.6	464.4 ±3.8	460.5 ±3.3	479.5 ±3.8	475.9 ±3.6	463.4 ±4.5	457.6 ±3.8
	C	483.3 ±5.8	465.5 ±3.7	459.8 ±3.8	446.7 ±3.5	438.6 ±3.7	463.3 ±3.5	460.4 ±3.8	454.4 ±2.8	441.5 ±2.6	472.5 ±3.8	464.9 ±3.5	453.4 ±3.8	445.6 ±3.5
	D	483.3 ±5.8	465.3 ±2.8	459.2 ±3.5	456.3 ±2.7	443.6 ±2.7	458.7 ±2.8	443.4 ±2.6	434.4 ±2.7	430.5 ±2.5	457.5 ±2.6	451.9 ±3.2	448.6 ±3.3	425.6 ±2.7
	E	483.3 ±5.8	478.3 ±4.2	466.5 ±3.5	460.9 ±2.8	441.6 ±3.1	468.8 ±2.4	463.4 ±2.7	458.4 ±2.8	439.4 ±3.2	479.7 ±3.8	472.6 ±2.4	458.4 ±2.8	455.6 ±4.2
	F	483.3 ±5.8	469.3 ±54.2	456.5 ±3.5	457.9 ±3.7	451.6 ±3.4	471.8 ±3.4	465.4 ±3.2	456.6 ±3.4	444.5 ±3.2	473.7 ±3.8	462.6 ±2.4	455.6 ±3.2	452.6 ±3.2

Table 6. – continued

Compound	Maturation variant	Control sample	[m(oak chip)/V(plum distillate)]/(g/L)											
			3				5				7			
			t(maturation)/month											
			3	6	9	12	3	6	9	12	3	6	9	12
			γ/(mg/L of absolute alcohol)											
2-Methyl-1-butanol	A	579.0 ±6.5	570.0 ±5.7	565.6 ±5.5	552.2 ±6.1	504.2 ±4.9	572.3 ±5.3	558.4 ±4.7	542.8 ±4.5	523.3 ±4.6	571.8 ±4.7	561.6 ±4.6	548.8 ±4.5	497.3 ±5.7
	B	579.0 ±6.5	574.0 ±6.7	572.3 ±6.5	562.6 ±6.3	544.2 ±4.9	576.0 ±6.5	570.3 ±5.5	566.1 ±5.3	542.2 ±5.4	570.0 ±5.7	568.3 ±6.5	562.6 ±5.3	548.2 ±4.9
	C	579.0 ±6.5	564.0 ±5.5	555.6 ±5.5	522.1 ±6.1	494.2 ±4.5	554.0 ±4.7	536.6 ±4.2	525.1 ±4.1	491.3 ±4.5	560.0 ±5.1	535.6 ±4.5	524.1 ±5.1	488.4 ±4.5
	D	579.0 ±6.5	567.6 ±5.7	552.3 ±5.3	531.2 ±5.1	488.2 ±4.5	564.2 ±4.7	546.6 ±4.2	532.1 ±4.6	502.2 ±5.5	552.0 ±4.8	537.2 ±4.5	529.3 ±4.6	483.5 ±4.5
	E	579.0 ±6.5	564.6 ±5.2	550.8 ±5.2	526.3 ±5.1	505.2 ±5.5	552.0 ±5.3	542.6 ±4.8	528.6 ±45.1	496.4 ±4.5	566.0 ±5.10	545.6 ±4.5	529.1 ±4.8	491.2 ±4.2
	F	579.0 ±6.5	555.0 ±4.7	550.6 ±4.5	542.1 ±4.1	524.2 ±4.9	552.3 ±4.3	548.4 ±3.7	536.8 ±3.5	529.3 ±3.1	551.8 ±4.7	550.6 ±4.6	548.8 ±4.5	517.2 ±3.7
3-Methyl-1-butanol	A	1573.3 ±18.4	1560.3 ±15.6	1545.8 ±15.4	1510.2 ±14.3	1507.8 ±13.9	1571.2 ±15.4	1495.7 ±14.7	1489.3 ±14.3	1461.2 ±13.7	1565.8 ±14.4	1532.3 ±14.7	1487.9 ±14.5	1488.2 ±13.8
	B	1573.3 ±18.4	1566.3 ±14.7	1549.5 ±13.4	1522.2 ±15.4	1529.8 ±18.4	1571.3 ±14.7	1559.5 ±14.6	1536.2 ±13.5	1527.8 ±13.4	1568.3 ±13.7	1533.5 ±13.9	1522.2 ±13.4	1529.8 ±12.6
	C	1573.3 ±18.4	1549.3 ±13.6	1530.8 ±14.6	1518.2 ±13.3	1513.8 ±13.5	1521.6 ±13.4	1496.5 ±13.2	1477.3 ±13.7	1466.8 ±13.3	1549.8 ±13.4	1530.8 ±13.6	1482.5 ±13.5	1478.5 ±13.6
	D	1573.3 ±18.4	1550.8 ±13.9	1541.2 ±14.6	1521.6 ±13.8	1521.8 ±13.7	1527.2 ±13.5	1490.7 ±12.7	1469.5 ±12.6	1460.6 ±12.5	1538.5 ±12.6	1530.8 ±12.3	1488.5 ±12.7	1476.6 ±12.6
	E	1573.3 ±18.4	1544.3 ±13.2	1538.8 ±13.5	1522.7 ±14.1	1525.8 ±13.2	1524.3 ±13.4	1488.5 ±13.2	1469.6 ±12.2	1455.6 ±12.3	1549.2 ±13.1	1535.5 ±13.4	1486.5 ±12.7	1480.2 ±11.8
	F	1573.3 ±18.4	1552.3 ±15.4	1546.8 ±15.4	1532.2 ±14.3	1517.8 ±13.9	1541.2 ±15.4	1525.7 ±14.7	1490.3 ±14.3	1468.2 ±13.7	1549.8 ±14.6	1532.3 ±14.7	1512.9 ±14.5	1498.2 ±13.8
1-Hexanol	A	45.90 ±2.23	45.23 ±2.55	44.75 ±2.48	44.36 ±2.36	44.28 ±2.29	45.29 ±2.45	44.75 ±2.33	44.36 ±2.15	44.43 ±2.42	45.63 ±2.35	45.25 ±2.28	44.62 ±2.38	44.33 ±2.12
	B	45.90 ±2.23	45.72 ±2.28	45.30 ±2.45	45.12 ±2.22	44.75 ±2.55	45.72 ±2.28	45.30 ±2.45	45.12 ±2.22	44.75 ±2.55	45.72 ±2.28	45.30 ±2.45	45.12 ±2.22	44.75 ±2.55
	C	45.90 ±2.23	45.12 ±2.45	44.42 ±2.28	44.06 ±2.22	44.18 ±2.40	44.79 ±2.30	44.55 ±2.30	44.36 ±2.25	44.23 ±2.22	44.63 ±2.25	44.25 ±2.30	44.12 ±2.18	44.33 ±2.12
	D	45.90 ±2.23	44.66 ±2.35	44.35 ±2.27	44.15 ±2.22	44.28 ±2.33	44.69 ±2.45	44.75 ±2.23	44.36 ±2.25	44.13 ±2.12	44.63 ±2.35	44.25 ±2.28	44.02 ±2.15	44.13 ±2.12
	E	45.90 ±2.23	44.66 ±2.35	44.35 ±2.27	44.15 ±2.22	44.28 ±2.33	44.69 ±2.45	44.75 ±2.23	44.36 ±2.25	44.13 ±2.12	44.63 ±2.35	44.25 ±2.28	44.02 ±2.15	44.13 ±2.12
	F	45.90 ±2.23	45.13 ±2.25	44.85 ±2.15	44.36 ±2.22	44.28 ±2.29	45.19 ±2.22	44.65 ±2.15	44.36 ±2.15	44.43 ±2.42	44.83 ±2.35	44.25 ±2.28	44.02 ±2.15	44.12 ±2.12
2-Phenyl ethanol	A	53.75 ±3.28	53.42 ±2.95	53.25 ±2.82	52.76 ±2.22	52.67 ±2.35	53.32 ±2.59	53.52 ±2.28	52.92 ±2.41	52.76 ±2.53	53.63 ±2.65	53.25 ±2.48	52.76 ±2.22	52.33 ±2.15
	B	53.75 ±3.28	53.44 ±2.66	53.45 ±2.36	53.35 ±2.12	52.86 ±2.35	53.60 ±2.42	53.54 ±2.61	53.27 ±2.27	52.68 ±2.53	53.60 ±2.42	53.15 ±2.16	52.76 ±2.27	52.86 ±2.45
	C	53.75 ±3.28	53.28 ±2.64	53.27 ±2.62	52.72 ±2.33	52.63 ±2.12	52.78 ±2.46	53.36 ±2.26	52.66 ±2.23	52.63 ±2.12	52.96 ±2.33	53.17 ±2.12	52.72 ±2.22	52.67 ±2.34
	D	53.75 ±3.28	52.42 ±2.35	52.25 ±2.47	52.33 ±2.22	52.27 ±2.15	52.62 ±2.27	52.34 ±2.33	52.18 ±2.05	52.64 ±2.32	52.53 ±2.41	52.36 ±2.17	52.64 ±2.27	52.46 ±2.15
	E	53.75 ±3.28	52.42 ±2.42	52.25 ±2.48	52.26 ±2.35	52.12 ±2.05	52.55 ±2.34	52.13 ±2.22	52.36 ±2.15	53.12 ±2.26	53.12 ±2.16	52.65 ±2.05	52.34 ±1.95	52.17 ±2.12
	F	53.75 ±3.28	53.44 ±2.76	52.81 ±2.55	52.55 ±2.26	52.27 ±2.05	53.63 ±2.67	53.42 ±2.27	52.79 ±2.14	52.47 ±2.19	53.13 ±2.33	52.81 ±2.27	52.59 ±2.26	52.14 ±2.32

A–E=see Table 1

The higher alcohols, *i.e.* 1-propanol, 2-methyl-1-propanol, 2-butanol, 1-butanol, 2-propen-1-ol, 2-methyl-1--butanol, 3-methyl-1-butanol, 1-hexanol and 2-phenylethanol are considered part of the aromatic skeleton of fruit distillates (*56*). A decrease in the concentration of higher alcohols was detected in the majority of samples, compared to the initial value in the fresh spirit (Table 6). The results of statistical analysis of the effect of maturation time showed that in samples aged at 20 °C, the changes in concentrations of higher alcohols were statistically insignificant during the first 6 months of the process. In the subsequent 6 months, the content of higher alcohols decreased significantly, on average by 15 %, compared to the control. Samples heated periodically (both to 35 and 45 °C) showed a more regular downward trend, but overall the decrease was not significantly greater than in samples aged at 20 °C. The type of used oak had a significant effect on final higher alcohol concentration, especially on 2-methyl-1-propanol, 2-methyl-1-butanol and 3-methyl-1-butanol, which were higher in samples aged with cognac barrel oak chips (Tables 4 and 6).

Rodríguez Madrera *et al.* (*37*) list evaporation, esterification, oxidation and sorption as causes that may explain the decrease in higher alcohols during ageing. Our results suggest that sorption plays an important role in the evolution of these volatiles. Sorption is especially connected with the type of oak chips used and the different porosities of the wood. The concentrations of 1-hexanol and 2-phenyl ethanol in the plum distillate samples did not show statistically significant changes during ageing, probably as a result of the lower reactivity of alcohols with increasing molecular mass (*57*).

Changes in concentrations of minor volatile compounds

An analysis of minor volatiles was also performed and presented in Table 7. To enable differentiation between plum distillate samples, principal component analysis (PCA) was conducted using as variables the concentration of minor volatile compounds (Figs. 4–10), which were significantly different (ANOVA, $p < 0.05$).

Table 7. Concentration of minor volatile compounds in matured plum distillate samples

Compound	γ/(mg/L of absolute alcohol)						
	Control sample	A	B	C	D	E	F
Acetate esters							
Methyl thiolacetate	0.007±0.001	0.005±0.001	0.006±0.001	0.006±0.001	0.006±0.001	0.008±0.001	0.008±0.001
Propyl acetate	0.007±0.001	0.004±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.002±0.001
Ethyl thiolacetate	0.017±0.001	0.010±0.001	0.017±0.001	0.007±0.001	0.007±0.001	n.d.	0.005±0.001
2-Methyl-1-butyl acetate	0.202±0.010	0.194±0.010	0.190±0.010	0.184±0.009	0.184±0.009	0.187±0.009	0.211±0.011
Linalyl acetate	0.011±0.001	0.001±0.001	0.005±0.001	0.005±0.001	0.006±0.001	0.007±0.001	n.d.
Nerolidyl acetate	0.027±0.001	0.028±0.001	0.023±0.001	0.028±0.001	0.027±0.001	0.025±0.001	0.016±0.001
2,3-Dihydrofarnesyl acetate	0.001±0.001	0.001±0.000	0.001±0.000	0.001±0.000	0.001±0.000	0.007±0.001	0.001±0.000
2-Methylpropyl acetate	0.023±0.001	0.030±0.002	0.028±0.001	0.027±0.001	0.029±0.002	0.031±0.002	0.032±0.002
Pentyl acetate	0.279±0.014	0.276±0.014	0.278±0.014	0.283±0.014	0.239±0.012	0.273±0.014	0.152±0.008
Octyl acetate	0.004±0.001	0.002±0.001	0.003±0.001	0.003±0.001	0.003±0.001	0.003±0.001	0.003±0.001
2-Phenylethyl acetate	0.093±0.010	0.082±0.010	0.070±0.008	0.078±0.008	0.073±0.006	0.081±0.007	0.212±0.011
Decyl acetate	0.016±0.001	0.014±0.001	0.013±0.001	0.013±0.001	0.013±0.001	0.010±0.001	0.017±0.001
Dodecyl acetate	0.006±0.001	0.005±0.001	0.004±0.001	0.004±0.001	0.005±0.001	0.004±0.001	0.007±0.001
Other esters							
Ethyl propanoate	0.036±0.002	0.048±0.002	0.038±0.002	0.041±0.002	0.043±0.002	0.044±0.002	0.045±0.002
Ethyl 2-methylpropanoate	0.382±0.019	0.246±0.012	0.239±0.012	0.289±0.015	0.269±0.014	0.278±0.014	0.158±0.008
3-Methyl-1-butyl acetate	0.002±0.001	0.009±0.001	0.007±0.001	0.009±0.001	0.009±0.001	0.020±0.001	0.018±0.001
Ethyl butanoate	0.240±0.012	0.252±0.013	0.247±0.012	0.260±0.013	0.260±0.013	0.254±0.013	0.255±0.013
Ethyl 2-butenoate	0.006±0.001	0.008±0.001	0.007±0.001	0.007±0.001	0.007±0.001	0.011±0.001	0.004±0.001
Ethyl 2-methylbutanoate	0.147±0.007	0.182±0.009	0.178±0.009	0.198±0.010	0.198±0.010	0.205±0.010	0.184±0.009
Ethyl 3-methylbutanoate	0.309±0.015	0.369±0.018	0.355±0.018	0.400±0.020	0.400±0.020	0.417±0.021	0.386±0.019
Ethyl pentanoate	0.029±0.002	0.002±0.001	0.001±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.003±0.001
Methyl pentanoate	0.004±0.001	0.003±0.001	0.002±0.001	0.003±0.001	0.003±0.001	0.001±0.001	0.002±0.001
Ethyl 2-methyllactate	n.d.	0.011±0.001	0.010±0.001	0.006±0.001	0.006±0.001	0.008±0.001	0.008±0.001
3-Methyl-1-butyl propanoate	0.020±0.001	0.012±0.001	0.025±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.009±0.001
3-Methyl-1-butyl isobutyrate	0.021±0.001	0.020±0.001	0.017±0.001	0.015±0.001	0.018±0.001	0.019±0.001	0.018±0.001
2-Methyl-pentyl propanoate	0.038±0.002	0.004±0.001	0.003±0.001	0.003±0.001	0.003±0.001	0.004±0.001	0.003±0.001
Ethyl 2-hexenoate	0.001±0.000	0.002±0.001	0.002±0.001	n.d.	0.002±0.001	0.003±0.001	0.002±0.001

Table 7. – continued

Compound	γ/(mg/L of absolute alcohol)						
	Control sample	A	B	C	D	E	F
Ethyl 2-furancarboxylate	0.010±0.001	0.016±0.001	0.025±0.001	0.014±0.001	0.015±0.001	0.002±0.001	0.002±0.001
3-Methyl-1-butyl butyrate	0.014±0.001	0.015±0.001	0.014±0.001	0.015±0.001	0.015±0.001	0.002±0.001	0.002±0.001
Ethyl 2,4-hexadienoate	0.007±0.001	0.032±0.002	0.104±0.005	0.042±0.002	0.055±0.003	0.005±0.001	0.013±0.001
Ethyl heptanoate	0.370±0.019	5.548±0.277	0.387±0.019	4.090±0.205	3.090±0.155	4.015±0.201	0.453±0.023
Methyl octanoate	0.006±0.001	0.041±0.002	0.034±0.002	0.056±0.003	0.046±0.002	0.006±0.001	0.004±0.001
Ethyl hexanoate	6.351±0.318	6.221±0.311	6.315±0.316	6.932±0.347	6.932±0.347	6.432±0.322	6.440±0.322
Ethyl 4-hexenoate	11.849±0.593	5.817±0.291	6.139±0.307	5.915±0.296	6.779±0.339	5.915±0.296	6.191±0.310
Ethyl 3-hexenoate	0.057±0.003	0.041±0.002	0.042±0.002	0.036±0.002	0.038±0.002	0.035±0.002	0.037±0.002
Ethyl benzoate	0.128±0.006	1.026±0.051	1.105±0.055	1.129±0.056	1.091±0.055	1.139±0.057	0.184±0.009
Diethyl butanedioate	n.d.	0.098±0.005	0.085±0.004	0.084±0.004	0.093±0.005	0.083±0.004	0.051±0.003
Butyl hexanoate	0.008±0.001	0.008±0.001	0.009±0.001	0.009±0.001	0.006±0.001	0.008±0.001	0.009±0.001
Ethyl octanoate	0.097±0.195	0.086±0.001	0.066±0.001	0.076±0.001	0.083±0.204	0.110±0.206	0.006±0.001
4-Octenoic acid ethyl ether	0.013±0.001	0.009±0.001	0.198±0.010	0.009±0.001	0.013±0.001	0.012±0.001	0.015±0.001
Methyl 2-hydroxybenzoate	0.004±0.001	0.002±0.001	0.005±0.001	0.004±0.001	0.005±0.001	0.005±0.001	0.007±0.001
Phenyl octadecanoate	0.002±0.001	0.027±0.001	n.d.	0.029±0.002	0.029±0.001	0.027±0.001	0.052±0.003
3-Methylbutyl octanoate	0.045±0.002	0.041±0.002	0.040±0.002	0.039±0.002	0.041±0.002	0.040±0.002	0.041±0.002
Propyl octanoate	0.009±0.0004	0.006±0.001	0.006±0.001	0.006±0.001	0.006±0.001	0.006±0.001	0.006±0.001
Ethyl nonanoate	22.30±1.12	21.36±1.07	23.46±1.17	22.35±1.12	23.61±1.18	20.21±1.01	22.16±1.11
Methyl decanoate	0.015±0.001	0.077±0.004	n.d.	0.014±0.001	0.012±0.001	0.010±0.001	0.011±0.001
2-Methylpropyl octanoate	0.075±0.004	0.064±0.003	0.059±0.003	0.063±0.003	0.062±0.003	0.055±0.003	0.063±0.003
2-Methylbutyl 2-furancarboxylate	0.005±0.001	0.069±0.003	n.d.	0.003±0.001	0.004±0.001	0.003±0.001	0.002±0.001
Ethyl decanoate	6.43±0.322	6.109±0.306	5.951±0.298	5.921±0.296	6.321±0.316	5.885±0.294	6.537±0.327
Ethyl 5-methylnonanoate	0.005±0.001	0.006±0.001	0.005±0.001	0.007±0.001	0.007±0.001	0.006±0.001	0.008±0.001
2-Methylbutyl octanoate	0.224±0.011	0.191±0.010	2.802±0.140	0.175±0.009	0.185±0.009	0.153±0.008	0.190±0.010
Ethyl 3-phenyl-2-propenoate	0.001±0.001	0.012±0.001	0.001±0.001	0.024±0.001	0.023±0.001	0.025±0.001	n.d.
Propyl decanoate	0.016±0.001	0.013±0.001	0.011±0.001	0.011±0.001	0.009±0.001	0.009±0.001	0.012±0.001
Butyl 9-decenoate	0.002±0.001	0.021±0.001	0.016±0.001	0.017±0.001	0.017±0.001	0.013±0.001	0.002±0.001
2-Methylpropyl decanoate	0.235±0.012	0.187±0.009	0.175±0.009	0.155±0.008	0.185±0.009	0.147±0.007	0.186±0.010
Pentyl nonanoate	0.004±0.001	0.022±0.001	0.002±0.001	0.003±0.001	0.003±0.001	0.002±0.001	0.023±0.001
Methyl dodecanoate	0.979±0.049	0.066±0.003	0.086±0.004	0.047±0.002	0.052±0.003	0.049±0.003	0.074±0.004
Ethyl dodecanoate	3.695±0.185	3.475±0.174	3.395±0.170	3.391±0.170	3.594±0.180	3.181±0.159	3.783±0.189
Isopropyl dodecanoate	0.004±0.001	0.005±0.001	0.003±0.001	0.004±0.001	0.005±0.001	0.006±0.001	0.036±0.002
Butyl 9-decenoate	0.018±0.001	0.025±0.001	0.013±0.001	0.013±0.001	0.014±0.001	0.014±0.001	0.244±0.012
3-Methylbutyl decanoate	1.061±0.053	0.989±0.050	0.967±0.048	0.914±0.046	1.014±0.051	0.843±0.042	1.002±0.050
1-Methylbutyl decanoate	0.385±0.019	0.347±0.017	0.318±0.016	0.298±0.015	0.331±0.017	0.268±0.013	0.351±0.018
2-Phenylethyl hexanoate	0.005±0.001	0.005±0.001	0.002±0.001	0.003±0.002	0.003±0.001	0.002±0.001	0.003±0.001
Propyl dodecanoate	0.004±0.001	0.005±0.001	0.004±0.001	0.004±0.002	0.003±0.001	0.003±0.001	0.002±0.001
Ethyl tridecanoate	0.004±0.001	0.003±0.001	0.002±0.001	0.003±0.001	0.002±0.001	0.003±0.001	0.002±0.001
Isobutyl dodecanoate	0.041±0.002	0.037±0.002	0.030±0.002	0.023±0.001	0.030±0.002	0.036±0.002	0.029±0.001
Ethyl heptadecanoate	0.001±0.001	0.004±0.001	0.003±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.001±0.001
Octyl octanoate	0.006±0.001	0.009±0.001	0.005±0.001	0.006±0.001	0.005±0.001	0.004±0.001	0.003±0.001
Ethyl tetradecanoate	0.647±0.032	0.429±0.022	0.371±0.019	0.389±0.019	0.399±0.020	0.406±0.020	0.445±0.022
Isopropyl tetradecanoate	0.028±0.001	0.004±0.001	0.001±0.001	0.002±0.001	0.003±0.001	0.002±0.001	0.002±0.001
3-Methylbutyl dodecanoate	0.109±0.005	0.134±0.007	0.127±0.006	0.132±0.007	0.125±0.006	0.091±0.005	0.155±0.008
2-Methylbutyl dodecanoate	0.038±0.002	0.040±0.002	0.048±0.002	0.041±0.002	0.041±0.002	0.029±0.002	0.075±0.004
2-Phenylethyl dodecanoate	0.009±0.001	0.010±0.001	0.008±0.001	0.011±0.001	0.009±0.001	0.014±0.001	0.011±0.001
Ethyl 9-octadecenoate	0.003±0.001	0.001±0.000	0.002±0.001	0.002±0.001	0.034±0.002	0.011±0.001	0.003±0.001
Ethyl pentadecanoate	0.010±0.001	0.008±0.001	0.007±0.001	0.008±0.001	0.008±0.001	0.009±0.001	0.007±0.001

Table 7. – continued

Compound	γ/(mg/L of absolute alcohol)						
	Control sample	A	B	C	D	E	F
Methyl hexadecanoate	0.001±0.000	0.001±0.000	0.001±0.000	0.001±0.001	0.001±0.0001	0.308±0.015	0.003±0.001
Isobutyl tetradecanoate	0.001±0.000	0.001±0.000	0.001±0.000	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001
Ethyl 9-hexadecenoate	0.010±0.000	0.007±0.001	0.007±0.001	0.005±0.001	0.005±0.001	0.006±0.001	0.015±0.001
Ethyl hexadecanoate	0.439±0.022	0.349±0.018	0.309±0.015	0.316±0.016	0.328±0.016	0.303±0.015	0.661±0.033
Isopropyl hexadecanoate	0.002±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.003±0.001	0.004±0.001
Isoamyl dodecanoate	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001
Ethyl heptadecanoate	0.001±0.001	0.033±0.002	0.003±0.001	0.003±0.001	0.002±0.001	0.002±0.001	0.013±0.001
Isooctyl dodecanoate	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.007±0.001	0.002±0.001
Dibutyl decanedioate	0.001±0.001	0.018±0.001	0.014±0.001	0.019±0.001	0.018±0.001	0.030±0.002	0.006±0.001
Acetals							
Formaldehyde diethyl acetal	0.019±0.001	0.179±0.009	0.229±0.012	0.217±0.012	0.189±0.010	0.260±0.013	0.140±0.007
Acetaldehyde ethyl propyl acetal	n.d.	0.007±0.001	0.005±0.001	0.011±0.001	0.012±0.001	0.015±0.001	0.002±0.001
Isobutyraldehyde diethyl acetal	n.d.	0.017±0.001	0.007±0.001	0.015±0.001	0.018±0.001	0.017±0.001	0.010±0.001
Valeraldehyde diethyl acetal	n.d.	0.012±0.001	0.049±0.003	0.051±0.003	0.052±0.003	0.047±0.002	0.009±0.001
Acetaldehyde ethyl amyl acetal	0.037±0.002	0.041±0.002	0.035±0.002	0.185±0.009	0.186±0.009	0.277±0.014	0.132±0.007
Octanal diethyl acetal	0.001±0.000	0.001±0.000	0.001±0.000	0.002±0.000	0.001±0.000	0.002±0.000	0.010±0.001
Aldehydes							
Hexanal	0.025±0.001	0.035±0.002	0.023±0.001	0.033±0.002	0.034±0.002	0.034±0.002	0.038±0.002
Benzaldehyde	0.888±0.044	0.817±0.041	0.114±0.006	0.094±0.005	0.043±0.002	0.077±0.004	0.126±0.006
5-Hydroxymethylfurfural	n.d.	0.420±0.021	0.228±0.011	0.532±0.027	0.568±0.028	0.558±0.028	0.163±0.008
Benzeneacetaldehyde	0.007±0.001	0.008±0.001	0.005±0.001	0.006±0.001	0.006±0.001	0.008±0.001	0.006±0.001
p-Methylbenzaldehyde	n.d.	0.022±0.001	0.012±0.001	0.014±0.001	0.013±0.001	0.011±0.001	n.d.
Nonanal	0.747±0.037	0.601±0.030	0.633±0.032	0.604±0.030	0.608±0.031	0.677±0.034	0.625±0.031
Cinnamaldehyde	0.012±0.001	0.042±0.002	0.023±0.001	0.045±0.002	0.040±0.002	0.042±0.002	0.026±0.001
Decanal	0.078±0.004	0.038±0.002	0.078±0.004	0.093±0.005	0.096±0.005	0.160±0.008	0.100±0.005
trans-2-Dodecenal	0.068±0.003	0.038±0.002	0.058±0.003	0.049±0.002	0.059±0.003	0.051±0.003	0.040±0.002
Dodecanal	0.006±0.001	0.012±0.001	0.020±0.001	0.024±0.001	0.040±0.002	0.029±0.002	0.039±0.002
Tetradecanal	0.051±0.003	0.048±0.002	0.046±0.002	0.044±0.002	0.047±0.002	0.034±0.002	0.069±0.003
Ketones							
2-Pentanone	0.008±0.001	0.024±0.001	0.095±0.005	0.042±0.002	0.037±0.002	0.041±0.002	0.022±0.001
2-Methyl-3-hexanone	0.006±0.001	0.007±0.001	0.004±0.001	0.008±0.001	0.007±0.001	0.007±0.001	0.005±0.001
2-Butanone	n.d.	0.003±0.001	0.002±0.001	0.004±0.001	0.004±0.001	0.004±0.001	0.003±0.001
4-Heptanone	1.401±0.070	1.371±0.069	1.354±0.068	1.462±0.073	1.382±0.069	1.436±0.072	0.856±0.043
3-Octanone	0.021±0.001	0.031±0.002	0.021±0.001	0.027±0.001	0.029±0.001	0.029±0.002	0.035±0.002
3,5,5-Trimethyl-2-cyclohexen-1-one	0.001±0.001	0.002±0.001	0.001±0.001	0.002±0.001	0.002±0.001	0.002±0.001	0.001±0.001
2-Nonanone	0.044±0.002	0.163±0.008	0.365±0.018	0.210±0.011	0.425±0.021	0.456±0.023	0.237±0.012
4-Undecanone	0.053±0.003	0.048±0.002	0.037±0.002	0.045±0.002	0.048±0.002	0.043±0.002	0.021±0.001
2-Tridecanone	0.239±0.012	0.111±0.006	0.109±0.006	0.099±0.005	0.087±0.004	0.087±0.004	0.135±0.007
Alcohols							
3-Ethyl-2-methyl-1-penten-3-ol	0.009±0.001	0.025±0.001	0.028±0.001	0.036±0.002	0.031±0.002	0.034±0.002	0.022±0.001
4-Methyl-1-pentanol	0.018±0.001	0.013±0.001	0.009±0.001	0.007±0.001	0.005±0.001	0.006±0.001	0.008±0.001
3-Methyl-1-pentanol	0.029±0.002	0.037±0.002	0.035±0.002	0.023±0.001	0.013±0.001	0.036±0.002	0.012±0.001
Heptanol	0.005±0.001	0.009±0.001	0.009±0.001	0.007±0.001	0.007±0.001	0.016±0.001	0.013±0.001
3-Octanol	3.179±0.159	3.538±0.177	3.825±0.191	3.796±0.190	3.365±0.168	3.756±0.188	4.098±0.205
Benzyl alcohol	0.056±0.003	0.022±0.001	0.041±0.002	0.019±0.001	0.022±0.001	0.016±0.001	0.008±0.001
β-Ethenyl-benzeneethanol	0.004±0.001	0.006±0.001	0.005±0.001	0.005±0.001	0.006±0.001	0.006±0.001	0.005±0.001
1-Octanol	0.034±0.002	0.039±0.002	0.012±0.001	0.035±0.002	0.044±0.002	0.062±0.003	0.046±0.002
2-Methyl-3-furanthiol	0.029±0.002	0.023±0.001	0.030±0.002	0.023±0.001	0.021±0.001	0.025±0.001	0.023±0.001

Table 7. – continued

Compound	γ/(mg/L of absolute alcohol)						
	Control sample	A	B	C	D	E	F
1-Dodecanol	0.943±0.047	1.233±0.062	1.057±0.053	0.763±0.038	0.461±0.023	0.499±0.025	0.465±0.0232
Citronellol	0.022±0.001	0.027±0.001	0.033±0.002	0.035±0.002	0.032±0.002	0.042±0.002	0.024±0.001
1-Decanol	0.065±0.003	0.065±0.004	0.023±0.001	0.063±0.004	0.065±0.003	0.054±0.004	0.096±0.005
1-Tetradecanol	0.042±0.002	0.055±0.003	0.053±0.003	0.074±0.004	0.092±0.005	0.015±0.001	0.020±0.001
1-Dodecanol	0.088±0.004	0.071±0.004	0.061±0.004	0.038±0.002	0.034±0.002	0.042±0.002	0.058±0.003
2,5,8-Trimethyl-1,2,3,4-tetrahydro--1-naphthalenol	0.076±0.004	0.094±0.005	0.091±0.005	0.094±0.005	0.090±0.005	0.078±0.004	0.176±0.009
1-Tetradecanol	0.371±0.019	0.297±0.015	0.235±0.012	0.281±0.014	0.173±0.009	0.196±0.010	0.336±0.017
Lactones							
δ-Nonalactone	n.d.	0.042±0.003	0.029±0.002	0.050±0.003	0.051±0.003	0.047±0.002	0.037±0.002
Butyrolactone	n.d.	0.013±0.001	0.009±0.001	0.019±0.001	0.018±0.001	0.018±0.001	0.009±0.001
γ-Nonalactone	n.d.	0.511±0.031	0.448±0.032	0.642±0.027	0.653±0.032	0.651±0.028	0.224±0.011
cis-β-Methyl-γ-octalactone	n.d.	0.015±0.002	0.004±0.001	0.028±0.001	0.028±0.001	0.026±0.001	0.024±0.001
trans-β-Methyl-γ-octalactone	n.d.	0.003±0.001	0.002±0.001	0.004±0.001	0.003±0.001	0.004±0.001	0.002±0.000
γ-Undecalactone	n.d.	0.011±0.001	0.011±0.001	0.017±0.001	0.016±0.001	0.016±0.001	0.004±0.001
Pentadecalactone	0.059±0.003	0.066±0.005	0.057±0.003	0.077±0.004	0.074±0.004	0.069±0.004	0.141±0.007
Other compounds							
4-Ethylguaiacol	n.d.	0.009±0.001	0.011±0.001	0.012±0.001	0.013±0.001	0.012±0.001	0.012±0.001
Methylbenzene	0.051±0.003	0.068±0.003	0.068±0.003	0.069±0.003	0.067±0.003	0.063±0.003	0.056±0.003
2-Methylthiophene	0.013±0.001	0.025±0.001	0.021±0.001	0.033±0.002	0.018±0.001	0.040±0.002	0.022±0.001
1,2-Dimethylbenzen	0.116±0.006	0.133±0.007	0.154±0.008	0.130±0.007	0.182±0.009	0.167±0.008	0.131±0.007
Ethenylbenzene (styren)	2.716±0.136	2.357±0.118	2.074±0.104	2.269±0.114	2.001±0.105	1.463±0.073	1.753±0.090
2-Acetylfuran	n.d.	0.011±0.001	0.005±0.001	0.013±0.001	0.016±0.001	0.019±0.001	0.011±0.001
α-Pinene	0.011±0.001	0.008±0.001	0.007±0.001	0.008±0.001	0.007±0.001	0.006±0.001	0.003±0.001
Propylbenzene (isocumene)	0.053±0.003	0.066±0.003	0.077±0.004	0.082±0.004	0.078±0.004	0.084±0.004	0.052±0.003
Phenol	0.003±0.001	0.005±0.001	0.004±0.001	0.007±0.001	0.006±0.001	0.006±0.001	0.002±0.001
1-Ethyl-2-methylbenzene	0.020±0.001	0.018±0.001	0.016±0.001	0.017±0.001	0.017±0.001	0.013±0.001	0.005±0.001
Dihydro-2-methyl-3(2H)-thiophe-none	0.019±0.001	0.009±0.001	0.005±0.001	0.006±0.001	0.006±0.001	0.006±0.001	n.d.
3-Carene	0.038±0.002	0.038±0.002	0.032±0.002	0.029±0.001	0.024±0.001	0.026±0.001	0.039±0.002
1,3,5-Trimethylbenzene	0.037±0.002	0.033±0.002	0.032±0.002	0.028±0.001	0.030±0.001	0.027±0.001	0.024±0.001
p-Cymene	0.065±0.003	0.082±0.004	0.035±0.002	0.037±0.002	0.065±0.001	0.040±0.002	0.034±0.002
ᴅ-Limonene	0.520±0.026	0.496±0.025	0.246±0.012	0.486±0.024	0.471±0.026	0.400±0.020	0.417±0.021
4-Methoxystyrene	9.323±0.466	3.398±0.170	5.329±0.267	3.658±0.183	3.304±0.155	1.730±0.087	2.450±0.123
Tridecane	0.057±0.003	0.052±0.003	0.045±0.002	0.043±0.002	0.047±0.002	0.049±0.002	0.058±0.003
2-Methylnaphtalene	0.042±0.002	0.024±0.001	0.032±0.002	0.028±0.001	0.027±0.001	0.028±0.001	0.019±0.001
1,6,8-Trimethyl-1,2,3,4-tetrahydro-naphthalene	n.d.	0.011±0.001	0.006±0.001	0.012±0.001	0.012±0.001	0.015±0.001	0.010±0.001
1,1,5-Trimethyl-1,2-dihydro-naphthalene (TDN)	1.269±0.064	1.512±0.076	1.244±0.062	1.515±0.076	1.492±0.065	1.653±0.083	1.163±0.058
1,2,3,4-Tetrahydro-1,1,6-trimethyl-naphthalene (α-ionene)	0.092±0.005	0.229±0.011	0.235±0.012	0.245±0.012	0.183±0.012	0.188±0.009	0.257±0.013
β-Sesquiphellandrene	0.093±0.005	0.087±0.004	0.088±0.004	0.079±0.004	0.083±0.004	0.052±0.003	0.073±0.004
cis-α-Bisabolene	0.080±0.004	0.056±0.003	0.056±0.003	0.050±0.003	0.056±0.003	0.046±0.002	0.063±0.003
2,4-Tert-butylphenol	0.112±0.006	0.187±0.009	0.164±0.008	0.227±0.011	0.264±0.013	0.274±0.014	0.250±0.013
α-Muurolene	0.255±0.013	0.225±0.011	0.240±0.012	0.216±0.011	0.265±0.012	0.189±0.010	0.242±0.012
α-Caryophyllene	0.045±0.002	0.034±0.002	0.051±0.003	0.042±0.002	0.036±0.003	0.023±0.001	0.042±0.002
Nerolidol	0.092±0.005	0.083±0.004	0.090±0.005	0.092±0.005	0.073±0.005	0.095±0.005	0.129±0.007

A–E=see Table 1; [m(oak chip)/V(plum distillate)]/(g/L) =5 g/L, t(maturation)= 12 months; n.d.=not determined

The detected compounds included many acetate esters, esters of higher acids (C3–C17) and methanol or ethanol, as well as esters of higher carboxylic acids and higher alcohols (C3–C8). The presence of esters of unsaturated carboxylic acid and ethanol, such as ethyl 2-hexenoate, ethyl 2,4-hexadienoate, ethyl 4-hexenoate, ethyl 3-hexenoate, diethyl butanedioate, ethyl 4-octenoate acid, ethyl 9-decenoate, ethyl 3-phenyl-2-propenoate (ethyl cinnamate) and others was also noted (Table 7).

The first and the second component of the PCA of ester concentrations in all matured plum distillate samples (Fig. 4) explain nearly 64 % of the total variance. PC1 accounted for 41.02 % of the total variance (the left region in the PCA plot) from the control sample (the right region in the PCA plot), whereas PC2 accounted for nearly 23 % of variance.

In comparison with the control sample, maturation caused a decrease in the concentration of esters such as ethyl thiolacetate, ethyl pentanoate, 2-methyl-pentyl propanoate, 2-methylpropyl octanoate, 2-methylpropyl decanoate, methyl dodecanoate, 3-methylbutyl decanoate, 1-methylbutyl decanoate, isobutyl dodecanoate and isopropyl tetradecanoate. On the other hand, an increase in the concentration of 2-methylpropyl acetate, ethyl propanoate, ethyl 2-methylbutanoate, ethyl benzoate, diethyl butanedioate, dibutyl decanedioate and ethyl 3-phenyl-2-propenoate was determined. Moreover, differences in the concentration of 2-methylbutyl dodecanoate, 3-methylbutyl dodecanoate and ethyl octanoate were found, which were affected by ageing system applied during maturation. In plum distillate samples treated with ultrasound, amyl alcohol esters of dodecanoic acid were found in higher concentrations, whereas the concentration of ethyl octanoate was lower than in the distillate matured in static system (Fig. 4). The changes in concentrations of the majority of esters were probably the result of transesterification (*46*).

The PCA of the results of acetal concentrations in maturated plum distillate samples is shown in Fig. 5. The PC1 and PC2 explain nearly 84 % of the total variance. The analysis was able to separate tested samples based on the maturation system applied in our experiments. The cluster corresponding to the samples obtained by maturation at 35 and 45 °C was located in the positive region of the first component (the right side of the PCA plot). PC2
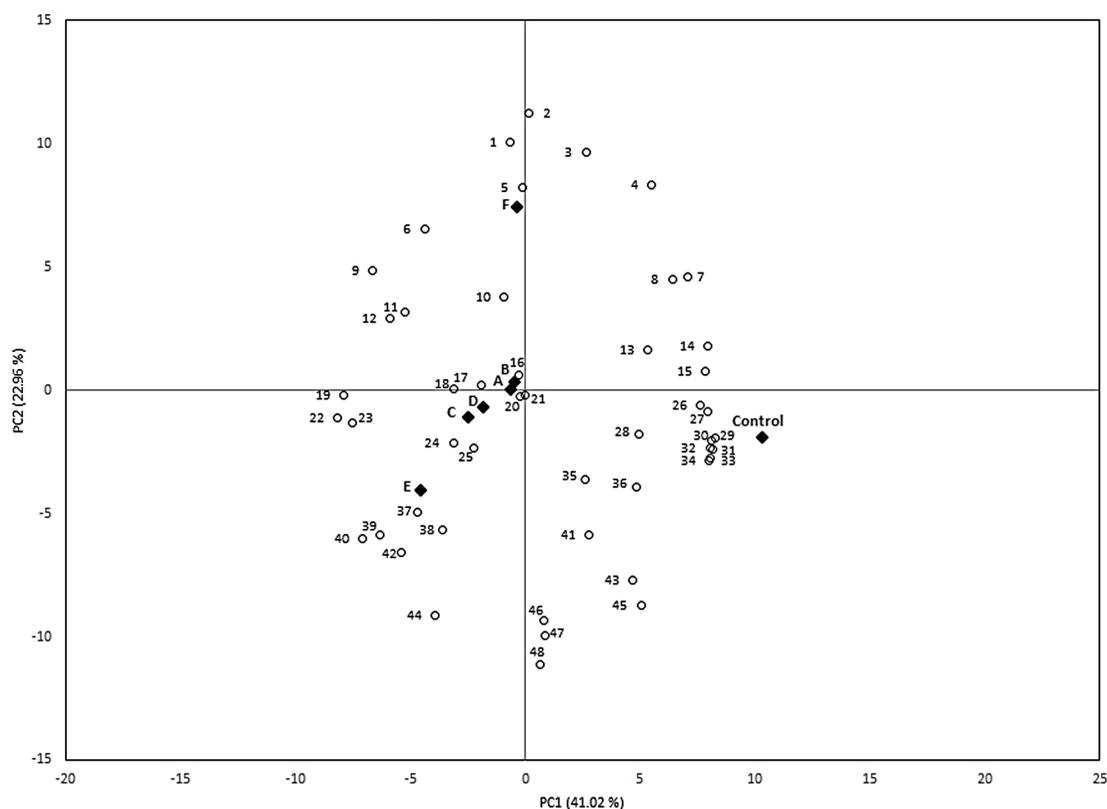
Fig. 4. Principal component analysis (PCA) biplot of esters in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=3-methylbutyl dodecanoate, 2=2-methylbutyl dodecanoate, 3=ethyl hexadecanoate, 4=decyl acetate, 5=pentyl nonanoate, 6=phenyl octadecanoate, 7=1-methylbutyl decanoate, 8=3-methylbutyl decanoate, 9=2-methylpropyl acetate, 10=ethyl heptadecanoate, 11=ethyl propanoate, 12=3-methyl-1-butyl acetate, 13=3-methyl-1--butyl propanoate, 14=2-methylpropyl decanoate, 15=2-methylpropyl octanoate, 16=4-octenoic acid ethyl ether, 17=ethyl 2,4-hexadienoate, 18=ethyl 2-hexenoate, 19=ethyl 3-methylbutanoate, 20=2-methylbutyl 2-furancarboxylate, 21=methyl decanoate, 22=ethyl 2-methylbutanoate, 23=diethyl butanedioate, 24=methyl octanoate, 25=ethyl 9-octadecenoate, 26=ethyl thiolacetate, 27=ethyl tetradecanoate, 28=3-methyl-1-butyl isobutyrate, 29=ethyl 3-hexenoate, 30=ethyl pentanoate, 31=methyl dodecanoate, 32=ethyl 4-hexenoate, 33=isopropyl tetradecanoate, 34=2-methyl pentyl propanoate, 35=3-methyl-1-butyl butyrate, 36=isobutyl dodecanoate, 37=ethyl heptanoate, 38=2,3-dihydrofarnesyl acetate, 39=ethyl benzoate, 40=dibutyl decanedioate, 41=propyl acetate, 42=ethyl 3-phenyl-2--propenoate, 43=ethyl pentadecanoate, 44=ethyl 2-butenoate, 45=ethyl 2-methyl propanoate, 46=nerolidyl acetate, 47=pentyl acetate, 48=ethyl octanoate
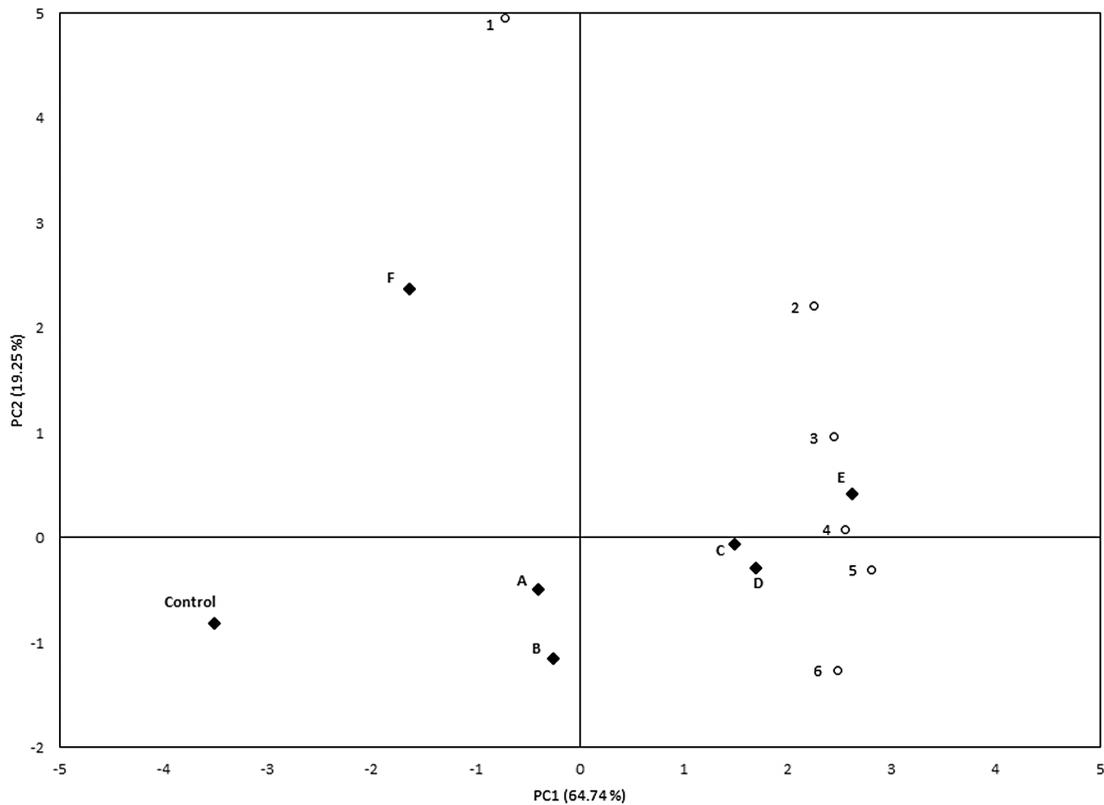
Fig. 5. Principal component analysis (PCA) biplot of acetals in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=octanal diethyl acetal, 2=acetaldehyde ethyl amyl acetal, 3=iso-butyraldehyde diethyl acetal, 4=formaldehyde diethyl acetal, 5=acetaldehyde ethyl propyl acetal, 6=valeraldehyde diethyl acetal

divided these three samples into a different group lying on the positive side of PC2 (upper part of PCA plot – sample E) and on the negative side of PC2 (lower part of PCA plot – samples C and D). This result can be related to higher (than 10-fold) concentration of formaldehyde diethyl acetal and acetaldehyde ethyl amyl acetal in samples aged at higher temperature than in those matured at room temperature. It was probably related to the liberation and oxidation/reduction of methylated oak wood components to formaldehyde, which subsequently reacted with ethanol to form the acetaldehyde. PCA allowed discrimination of plum distillate samples matured by using static system from those obtained by the dynamic maturation.

Among minor compounds determined in the plum distillate before and after maturation were aldehydes such as hexanal, benzaldehyde, benzeneacetaldehyde, nonanal, cinnamaldehyde, decanal, *trans*-2-dodecenal, dodecanal and tetradecanal. Moreover, samples after maturation contained 5-hydroxymethylfurfural, which was absent from the fresh distillate. However, the content of 5-hydroxymethylfurfural (Table 7) was approx. 5 times lower than that of furfural (Table 3). Similar results were found by Fernández de Simón *et al.* (*58*), who performed a quantitative evaluation of volatile compounds in extracts from various *Quercus pyrenaica* oak wood products.

PCA results for aldehydes were presented in Fig. 6. PC1 and PC2 accounted for 45.44 and 22.46 % of the total variance, respectively. The first component separated the samples matured with lightly toasted French oak chips (located on the positive side of PCA plot) from those mat-

urated with oak chips after ageing cognac (located on the negative side of PCA plot). This difference was explained by 5-hydroxymethylfurfural and cinnamaldehyde; the distillates obtained after maturation with lightly toasted French oak chips were richer in the above-mentioned aldehydes. The formation of 5-hydroxymethylfurfural as carbohydrate derivative can be intensified during toasting, since a variety of reactions take place (*58*). Regarding changes in cinnamaldehyde content, it can be supposed that it may be produced during condensation of benzaldehyde and acetaldehyde (*59*). The second component (PC2) clearly separated tested plum distillate samples according to the applied ageing system. Distillate sample after maturation in static system (variant A) was located in upper (positive) region of PCA plot, while distillate obtained after maturation with ultrasound (variant F) was located in the lower (negative) side of the PCA plot. The obtained results also indicate difference in the concentration of *p*-methylbenzaldehyde and dodecanal (Fig. 6).

Regarding PCA results for ketones (Fig. 7), the first two components (PC1 and PC2) explained over 73 % of the total variance. The first principal component explained 45.13 % of the total variance that separated the matured samples of plum distillate from the control (before maturation). Analysis of the correlation coefficient values of PC1 with the concentration of ketones showed that the differentiation was related mainly with the concentration of 2-butanone, 2-nonanone and 2-tridecanone. The second component (over 28 % of the total variance) was correlated mainly with the sample after maturation
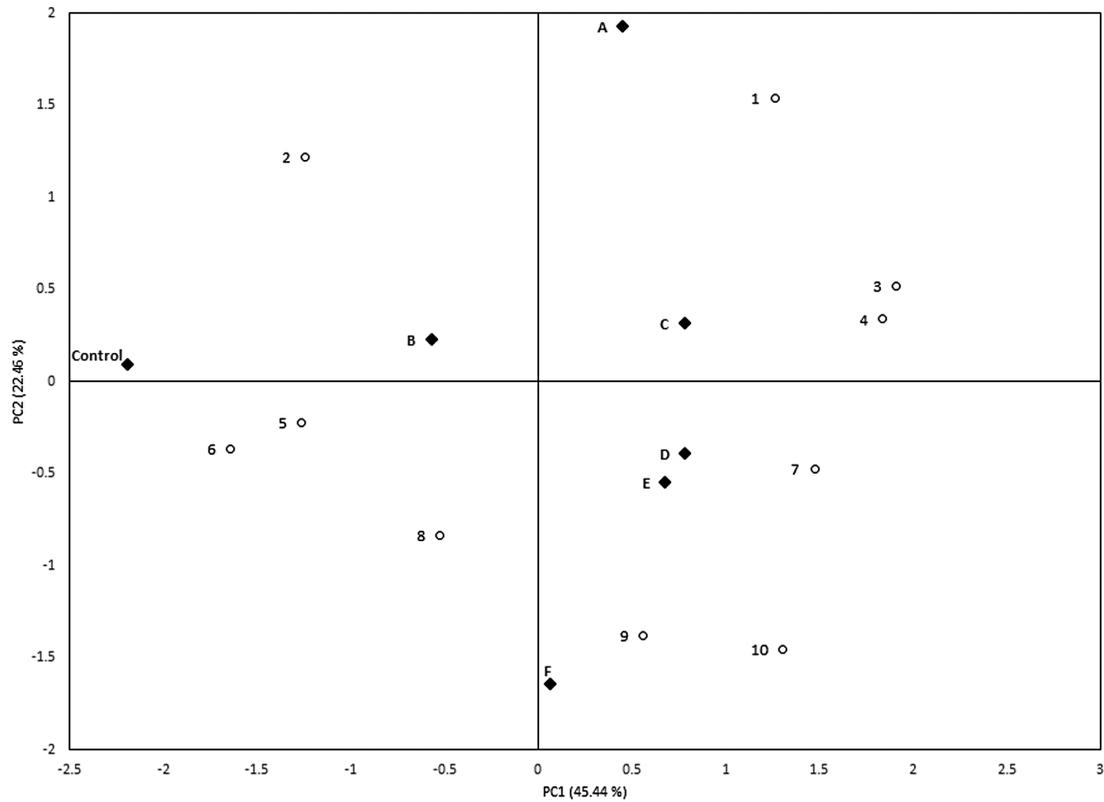
Fig. 6. Principal component analysis (PCA) biplot of aldehydes in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=*p*-methylbenzaldehyde, 2=benzaldehyde, 3=cinnamaldehyde, 4=5-hydroxymethylfurfural, 5=*trans*-2-dodecenal, 6=nonanal, 7=hexanal, 8=tetradecanal, 9=decanal, 10=dodecanal
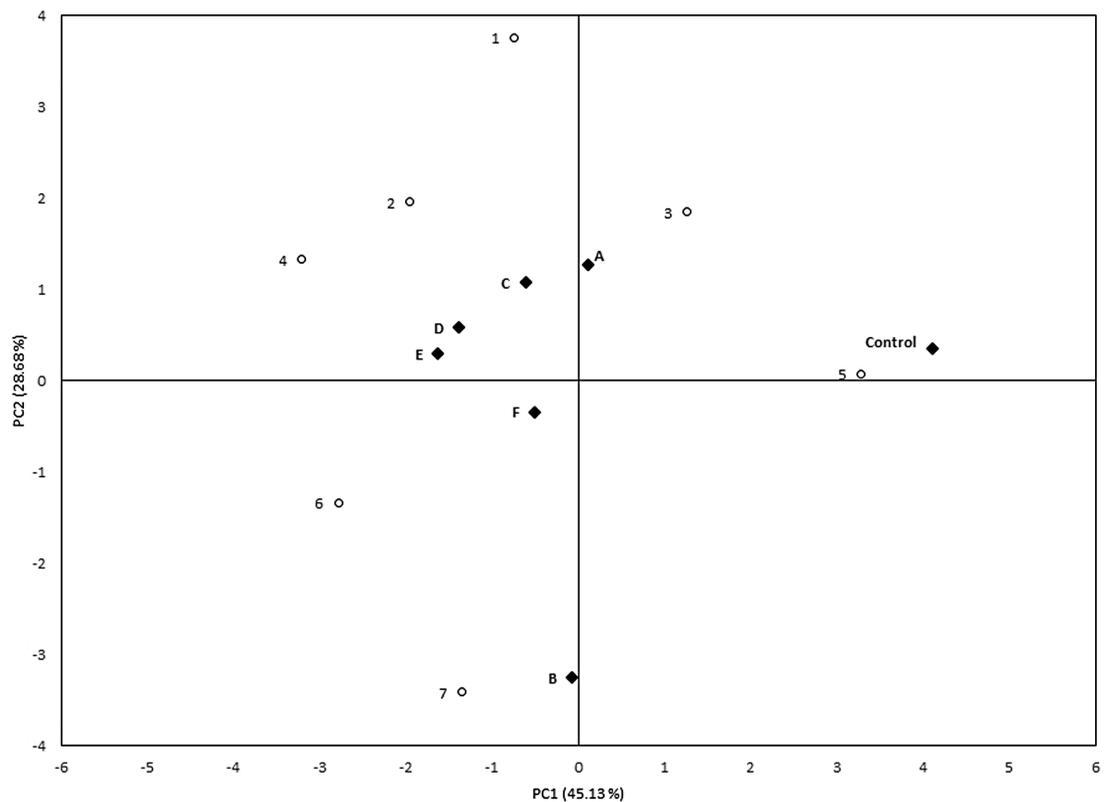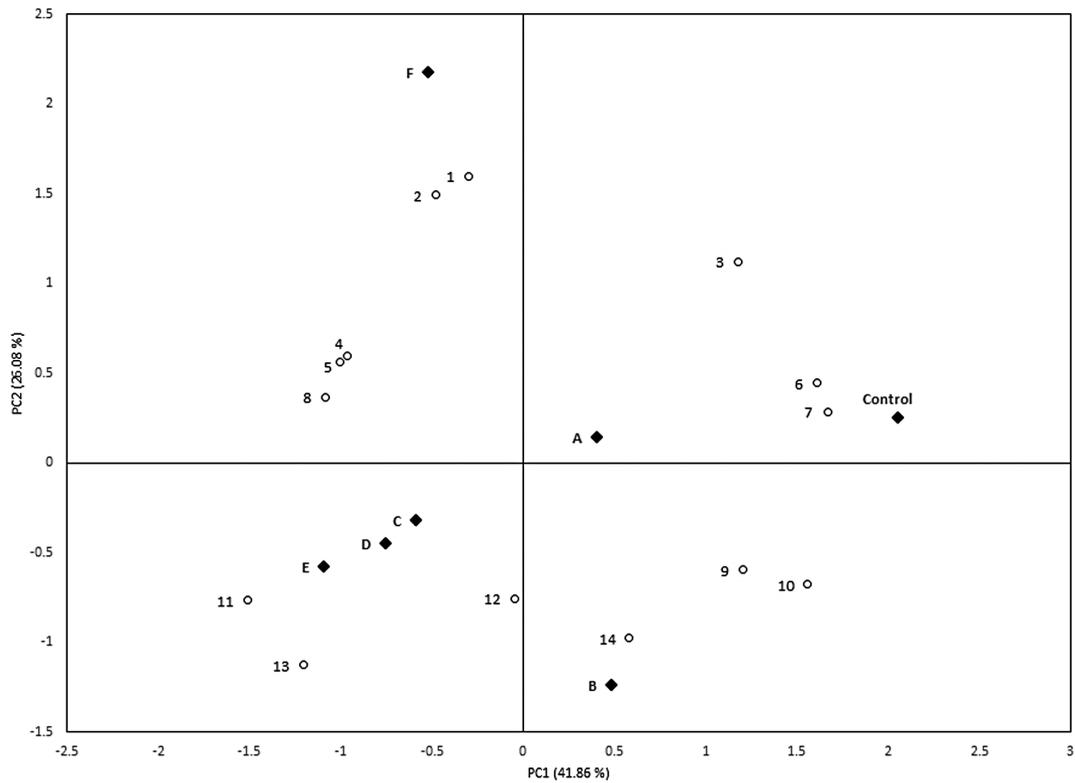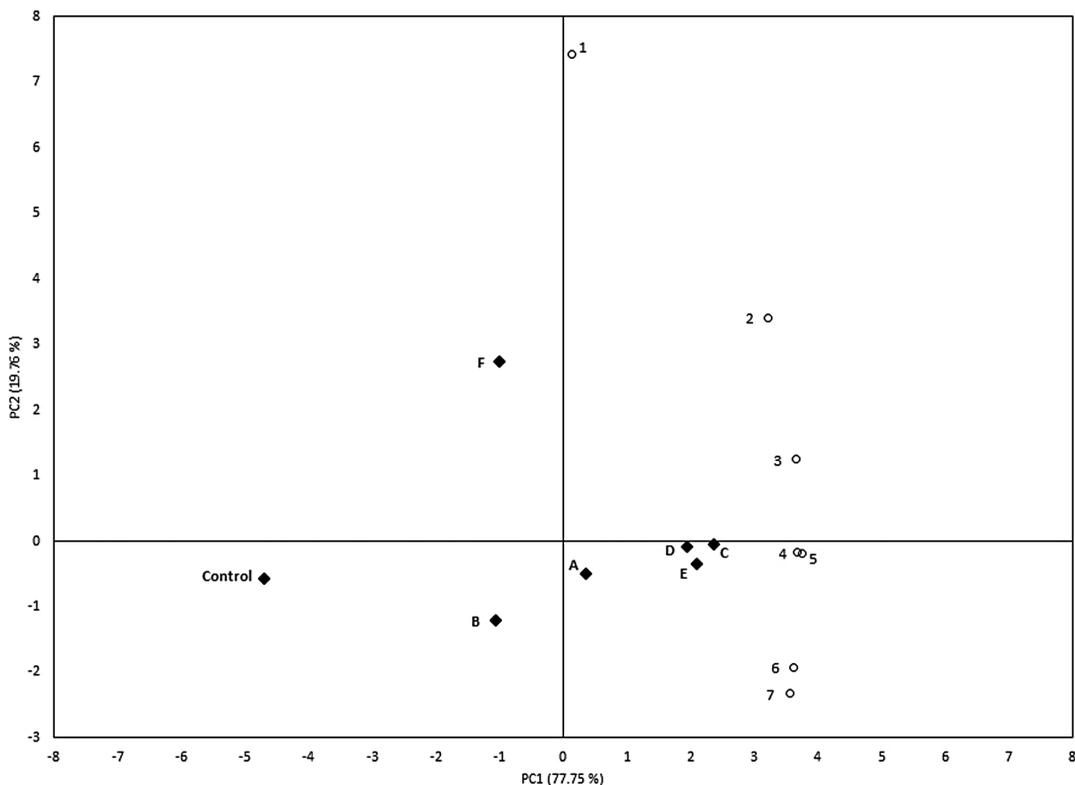

Fig. 7. Principal component analysis (PCA) biplot of ketones in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=2-methyl-3-hexanone, 2=3-octanone, 3=4-undecanone, 4=2-butanone, 5=2-tridecanone, 6=2-nonanone, 7=2-pentanone

Fig. 8. Principal component analysis (PCA) biplot of alcohols in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=1-decanol, 2=2,5,8-trimethyl-1,2,3,4-tetrahydro-1-naphthalenol, 3=1-tetradecanol, 4=1-octanol, 5=3-octanol, 6=1-dodecanol, 7=4-methyl-1-pentanol, 8=heptanol, 9=2-methyl-3-furanthiol, 10=benzyl alcohol, 11=3-ethyl-2-methyl-1-penten-3-ol, 12=1-tetradecanol, 13=citronellol, 14=3-methyl-1-pentanol

Fig. 9. Principal component analysis (PCA) biplot of lactones in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation). For designation of maturation variants A–F see Fig. 1. 1=pentadecalactone, 2=*cis*-β-methyl-γ-octalactone, 3=δ-nonalactone, 4=*trans*-β-methyl-γ-octalactone, 5=butyrolactone, 6=γ-nonalactone, 7=γ-undecalactone

with oak chips after cognac ageing (at 20 °C). This result was related to higher concentration of 2-pentanone than in the distillate sample aged with lightly toasted French oak chips.

The principal component analysis of alcohols was also done (Fig. 8). The first component (PC1) explained 41.86 % of the total variance and was characteristic mainly for the control sample, while the second component (PC2) accounted for about 26 % of variance and was correlated mainly with distillate samples obtained with ultrasound treatment during maturation (variant F; upper part of PCA plot). In contrast, distillates aged at higher temperature were scattered in the negative region of PC2. In these samples, concentrations of 3-ethyl-2-methyl-1--penten-3-ol, citronellol and 1-tetradecanol were the highest in comparison with the other samples.

The plum distillate samples after maturation contained compounds originating from the oak wood, including oak lactones and volatile phenols (Table 7). Lactones are usually described as having aromas that are fruity or coconut-like (γ-octalactone), creamy, lactonic (γ--nonalactone, γ-hexalactone), peach-like, milky (γ-decalactone) or fruity and sweet floral (γ-dodecalactone). These compounds are formed by cyclisation of the corresponding γ-hydroxycarboxylic acids (*60,61*). Data related to lactone concentration were also submitted to PCA analysis.

The first (PC1) and second (PC2) principal components explained 97.51 % of the total variance. PC1 accounted for 77.75 % of total variance, characterized mainly by control sample and distillate obtained by maturation at the higher temperature (variants C, D and E) (Fig. 9). It was related to the presence of the highest concentration of lactones, such as: δ-nonalactone, butyrolactone, γ-nonalactone, *cis*--β-methyl-γ-octalactone, *trans*-β-methyl-γ-octalactone (known as whisky lactone), and γ-undecalactone, in comparison with other matured plum distillate samples. In turn, the presence of the above-mentioned compounds was not observed in the control sample. PC2 explained 19.76 % of the total variance and was characterized by distillate treated with ultrasound during maturation. In this sample, the concentration of lactones was relatively low, while the concentration of pentadecalactone was the highest, in comparison with other samples. Moreover, PC2 explained differences between distillates matured with lightly toasted French oak chips (the right side of the PCA plot) and those aged with French oak chips after cognac ageing (the left side of the PCA plot). The results of lactone concentration indicated their lower level in samples matured with cognac barrel oak chips (variant B). Also, their concentrations did not increase when circulation was applied.

Toasting *Quercus* sp. oak wood is one of the key stages in the manufacture of both barrels and oak wood chips
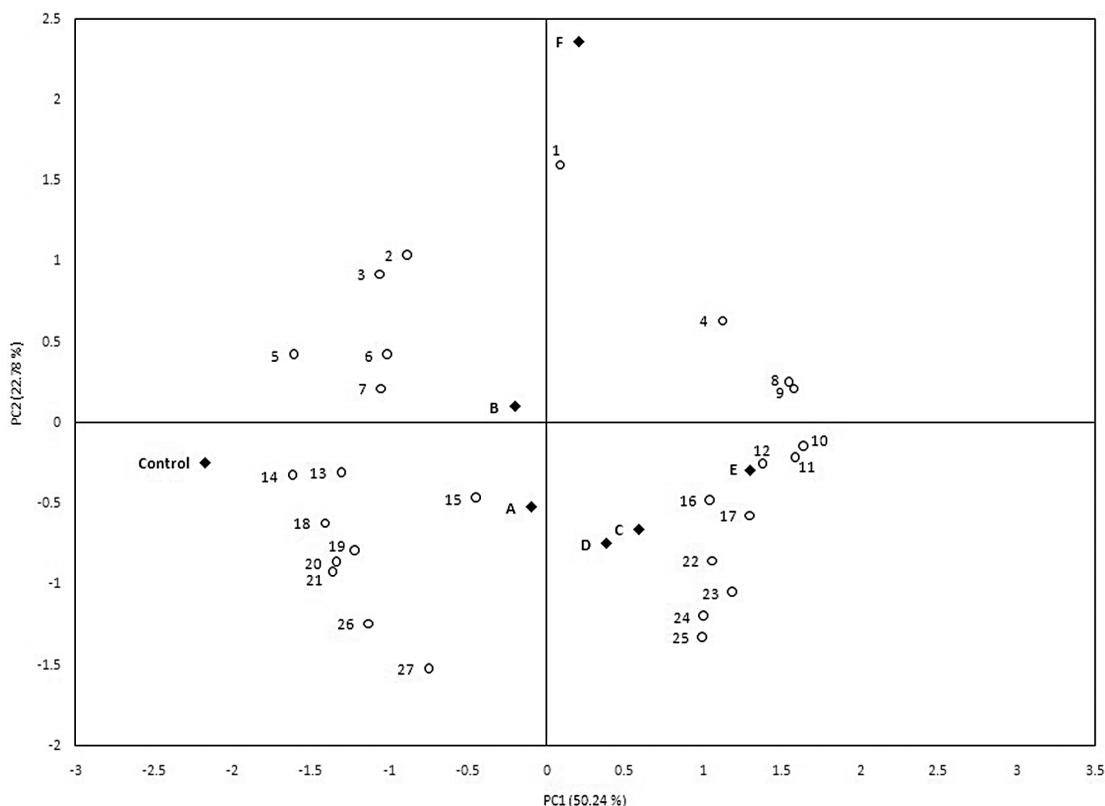

Fig. 10. Principal component analysis (PCA) biplot of other minor compounds (terpenic compounds, benzene, phenol and naphthalene derivatives) in matured plum distillate samples (5 g/L of oak chips, 12 months of maturation).
For designation of maturation variants A–F see Fig. 1. 1=nerolidol, 2=tridecane, 3=3-carene, 4=α-ionene, 5=*cis-α*-bisabolene, 6=α--caryophyllene, 7=α-muurolene, 8=2,4-*tert*-butylphenol, 9=4-ethylguaiacol, 10=1,6,8-trimethyl-1,2,3,4-tetrahydronaphthalene, 11=2-acetylfuran, 12=2-methylthiophene, 13=β-sesquiphellandrene, 14=4-methoxystyrene, 15=ᴅ-limonene, 16=1,2-dimethylbenzen, 17=*p*-cymene, 18=ethenylbenzene, 19=2-methylnaphtalene, 20=dihydro-2-methyl-3(2H)-thiophenone, 21=1,2,5-trimethylbenzene, 22=methylbenzene, 23=propylbenzene, 24=1,1,5-trimethyl-1,2-dihydronaphthalene, 25=phenol, 26=α-pinene, 27=1-ethyl-2-methylbenzene

intended for ageing wines and spirits. During this operation, the increased temperature causes various modifications in the physical structure and, more importantly, in the chemical composition of the wood. Aromatic hydrocarbon molecules formed during toasting are subsequently extracted by the wine or spirits (*61*). These aromatic hydrocarbon molecules include volatile phenols, which impart smoky and spicy characteristics to matured spirits (*62*).

The PCA biplot of terpenic compounds, benzene, phenol and naphthalene derivatives is presented in Fig. 10. PC1 component explained 50.24 % of the total variance, characterized mainly by control sample, as well as by samples C and E, whereas the PC2 explained over 22 % of the total variance, characterized by distillates obtained with using ultrasound treatment (variant F).

Control sample was located on the left side of the PCA plot, while distillate samples matured at higher temperature (variants C, D and E) were distributed on the right side. The differences between these samples were mainly in the concentrations of 4-ethylguaiacol, 2-acetylfuran, 1,6,8-trimethyl-1,2,3,4-tetrahydronaphthalene and 2,4-*tert*-butylphenol. The first two compounds were not detected before maturation, while 2,4-*tert*-butylphenol had the lowest concentration. According to the literature (*63*), the concentration of 2-acetylfuran varies depending on the type of cask and the length of ageing.

Conclusions

The microscale experiments conducted to investigate the accelerated ageing with added oak wood chips resulted in changes of the composition of plum distillate. Significant decreases in the concentrations of compounds characteristic of immature distillate (aldehydes, higher alcohols and lower esters such as ethyl acetate) were observed, with a simultaneous increase in the concentrations of many esters of higher acids and alcohols, acetals and others. Contact with oak wood chips also improved the colour of the plum distillate and enhanced the concentrations of compounds important for aroma and taste, such as phenolics (syringaldehyde, coniferaldehyde and vanillin), lactones and phenols. These changes depended on the type and dose of the used oak chips, and on the conditions of maturation. To accelerate changes in the composition of matured distillate, periodical heating (preferred temperature 35 °C) could be applied.

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