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# Chemo-sensory characterization of aroma active compounds of native oak wood in relation to their geographical origins

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ABSTRACT

Oak wood contains aroma-active compounds that contribute significantly to the chemical structure, olfactory and gustatory qualities of alcoholic beverages and vinegars as by-products that have been either fermented and/or aged in oak barrels. The chemical composition of cooperage oak is highly variable, depending on the degree of toasting and natural seasoning. However, it is unclear whether the odor of oak varies according to different geographical regions and pedoclimatic conditions. Especially in view of the actual challenges in forestry in relation to climate change, the present study aimed at elucidating the odorous constituents of nine natural oak samples from Germany, Austria and Hungary with respect to these influencing parameters. The odor profiles of the oaks were compared, the potent odorants were determined, and selected odorants were quantified using stable isotope dilution assays (SIDA). The majority of the identified odorants in all samples were fatty acid degradation products, followed by a series of odorants with terpenoic structure and others resulting from the degradation of lignin. Several different odorants including 2-propenoic acid and cinnamaldehyde are reported here for the first time in oaks from different growth regions. Odor activity values (OAVs), calculated based on odor thresholds (OTs) in water, revealed hexanal, (*E*)-2-nonenal, (*Z*)-3-hexenal, eugenol, vanillin, and whiskey lactone as potent odorants for the oak odor. Principal component analysis of the data obtained from sensory evaluation, comparative aroma extract dilution analysis (cAEDA) and their corresponding quantified odorants showed that the highest separation rate was obtained for Hungarian oak, whereas Austrian and Bavarian oak samples were more similar. Recombination experiments by mixing the dominant odorants in their naturally occurring concentrations revealed a good agreement of the smell properties of the model mixture with the smell of the respective original sample. These findings aim at evaluating and establishing a better understanding of the distinctive smell of oak wood and demonstrated the prospects of new oak sources.

## 1. Introduction

The wood species oak belongs to the Quercus genus of the Fagacea family. The distinctive physical and chemical nature of oak makes it a desirable timber to work with for several reasons. It has medium to high density and can stay moisture-free. This wood is very strong, but it can still be bent. These properties make it perfect timber with great strength and hardness, oxygen exposure control and water penetration (Shanbhag & Sundararaj, 2013). As for all wood species, cellulose (40%), hemicellulose (25%) and lignin (20%) are the main components (Fernández de Simón, Cadahía, Conde, & García-Vallejo, 1996; Nonier, Vivas, Vivas de Gaulejac, Absalon, Soulié, & Fouquet, 2006). In addition

*Abbreviations:* cAEDA, comparative aroma extract dilution analysis; 2D GC–MS/O, two-dimensional gas chromatography–mass spectrometry/olfactometry; FD, flavor dilution; FID, flame ionization detector.

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to these base structural components, non-structural compounds such as extractives, organic and inorganic compounds are also contained within the heartwood of oak species.

Oak is used for a broad range including the production of alcoholic beverages such as wine and vinegars where oak barrels or oak chips and shavings are utilized in the storage process (Culleré, Fernández de Simón, Cadahía, Ferreira, Hernández-Orte, & Cacho, 2013; Pérez-Coello, Sánchez, et al., 2000; Pérez-Coello, González-Viñas, García-Romero, Cabezudo, & Sanz, 2000; Torija et al., 2009). In the course of this process, aged beverages notably mature, i.e. with regard to color, flavor, and mouthfeel. These attributes are strongly related to the compounds released by the wood. The aging of spirit vinegar for example involves changes in color, aroma and polyphenol profile. During the aging process of vinegars in oak barrels, the volatile components gain complexity and enrichment as a result of three processes: the release of odorants from oak matrices into the samples, the volatile compounds are concentrated since water evaporates through the wood barrel pores, and new odorants such as esters are formed (Morales, Benitez, & Troncoso, 2004; Morales, Tesfaye, García-Parrilla, Casas, & Troncoso, 2002; Ríos-Reina, Segura-Borrego, García-González, Morales, & Callejón, 2019; Ubeda et al., 2011).

Regarding the influence of aroma compounds from oak on alcoholic beverages and vinegars during maturation and aging, it is important to consider that oak barrels and chips are commonly not produced in their untreated and natural form, but heat treated as in toasting or modified by seasoning (Anjos, Carmona, Caldeira, & Canas, 2013; Fernández de Simón, Cadahía, & Jalocha, 2003; Prida & Puech, 2006; SAVILL, 1996; Sefton, Francis, & Williams, 1990). Particularly the influence of treatments such as toasting and seasoning during cooperage, as well as factors like single tree variation, tree species, and geographical location can change and modify both the chemical and physical qualities of oak (Doussot, De Jeso, Quideau, & Pardon, 2002; Prida & Puech, 2006). Accordingly, many investigations focused on the odorants of treated oak wood generated during seasoning and toasting (Cutzach, Chatonnet, Henry, & Dubourdieu, 1997; Díaz-Maroto, Guchu, Castro-Vázquez, de Torres, & Pérez-Coello, 2008; Jordão et al., 2006; Li et al., 2015; Nonier, De Gaulejac, Vivas, & Vitry, 2004; Pérez-Coello, Sanz, & Cabezudo, 1998). Several research groups have investigated the botanical species in relation to climate, soil, topography, and geographical origin.

American white oak (*Quercus alba L.*) and French red oak (*Quercus robur L.* and *Quercus petraea Liebl.*) are three of the main commercially used sources of oak in coopering (Glabasnia & Hofmann, 2006). Some previous studies also focused on investigating new regions and new types of oak wood from Spain, Russia and other East European countries (Romania and Hungary) with the prospect to find suitable new alternative species of oak aging sources, adapted to their specific locations of growth to cover potential shortages in wood availability (Cadahía, Varea, Muñoz, Fernández de Simón, & García-Vallejo, 2001; Díaz-Maroto, Guchu, Castro-Vázquez, de Torres, & Pérez-Coello, 2008; Fernández de Simón, Cadahía, & Jalocha, 2003; Marco, Artajona, Larrechi, & Rius, 1994; Mosedale, Feuillat, Baumes, Dupouey, & Puech, 1998). In this respect it is interesting to note that relevant studies revealed that wines aged in Spanish oak barrels showed characteristics similar to the same wines aged in French oak barrels and significant difference compared to wines aged in American oak barrels. Others suggested that Eastern European grown oak contained higher amounts of aromatic compounds, such as volatile phenols and phenolic aldehydes than French oak, even though they belong to the same species (Fernández de Simón, Hernández, Cadahía, Dueñas, & Estrella, 2003; Prida & Puech, 2006).

However, the general chemical odorant composition of the native untreated natural wood itself from different botanical species and geographical origin has not been regarded in this respect as the relevant starting point to date. We made a start with our previous investigation that specifically targeted the structural elucidation of aroma-active compounds in natural Hungarian oak (*Quercus frainetto L.*), grown in the Deszk forest, region in Hungary (Ghadiriasli, Wagenstaller, & Buettner, 2018). There, we demonstrated a method to analyse several important odorants in unmodified oak samples. The present study now focuses on the potential influence of geographical origin and pedoclimatic conditions on quantitative differences in the odorous constituents, and on the related odor profiles of natural oak. To this aim, the goal was to identify the odorants in natural untreated oak samples (*Quercus robur L.*) from Germany, Austria, and Hungary geographical locations and to assess the contribution of single odorants to the overall odor impression of different oak woods. Therefore, quantitative investigation of selected odorants belonging to prominent substance classes was carried out using stable isotope dilution assays (SIDA), followed by a calculation of odor activity values (OAVs, ratio of concentration to odor threshold). Aroma recombination experiments were then performed for sensory confirmation.

## 2. Materials and methods

### 2.1. Oak wood material

Oak samples of *Quercus robur L.* species were supplied by nine different forestry stations, namely Mecsek forest in Baranya region Hungary; Röttenbach, Zeitlofs-Bad Kissingen, Thüngen, and Aschaffenburg, all located in Bavaria, Germany; Weißenbach and Leonberg, both in Baden-Württemberg, Germany; and Horn and Lengenfeld in Niederösterreich, Austria (Table 1), in which none of these sites are protected under the law of their home countries. All samples were delivered as whole tree trunk segments, as commonly used for barrel- and furniture-making. All oak samples obtained in 2017. The cut boards from these untreated samples were then shaved into small pieces of approx. 2 cm × 2 cm each and were directly used for analysis without any further treatment.

### 2.2. Chemicals

A homologous series of n-alkanes from n-hexane to n–tetratriacontane (50 μg/ml) dissolved in pentane (>99% purity) was prepared for the determination of retention indices (all reagents from Sigma-Aldrich, Steinheim, Germany). Dichloromethane and pentane were freshly distilled prior to use, and sodium sulfate was used in its anhydrous state (both obtained from Th. Geyer GmbH & Co. KG, Renningen, Germany). The sources of the chemicals used in this study are listed in supplementary material File 1.

### 2.3. Sensory evaluation

The panel consisted of 10 trained volunteers (4 male, 6 female; age 24–54) from the Department of Sensory Analytics, Fraunhofer Institute for Process Engineering and Packaging IVV (Freising, Germany). The panelists were healthy and their olfactory function was normal during the testing. Their olfactory capabilities were trained and evaluated during weekly training sessions in which the panelists were tested for their ability to recognize, describe, and name selected in-house made

**Table 1**
Description of the oak samples used for analysis.

| Oak Sample | Geographical origin | Code |
|---|---|---|
| Hungarian Oak (Oak H) | Mecsek forest – Baranya region-Hungary | H-B |
| German Oak (Oak G1) | Röttenbach-Bavaria-Germany | G1-B |
| German Oak (Oak G2) | Zeitlofs-Bad Kissingen- Bavaria-Germany | G2-B |
| German Oak (Oak G3) | Thüngen- Bavaria-Germany | G3-B |
| German Oak (Oak G4) | Aschaffenburg-Bavaria-Germany | G4-B |
| German Oak (Oak G5) | Weißenbach-Baden-Württemberg-Germany | G5-BW |
| German Oak (Oak G6) | Leonberg- Baden-Württemberg-Germany | G6-BW |
| Austrian Oak (Oak A1) | Horn- Niederösterreich- Austria | A1-NO |
| Austrian Oak (Oak A2) | Lengenfeld- Niederösterreich-Austria | A2-NO |

odorant pens, corresponding to a total of over 150 different odorants. Panelists were trained for at least six weeks prior to inclusion in the panel. The correctness of the individual answers was evaluated according to DIN EN ISO 8586:2014–05. The samples were rated during two sessions alongside with the corresponding reference substances. In this procedure, the reference substances of the selected attributes, namely 2,4,6-trichloroanisole (cork-like, musty), (*E*)-2-nonenal (fatty cardboard-like), carvacrol (woody), acetic acid (vinegar-like), hexanal (green-grassy), whiskey lactone (coconut-like), octanal (citrus-like), and $\alpha$-pinene (resin-like), were presented to the panelists in the form of sniffing sticks.

Wood samples (2 g ± 0.1 g) were presented to the panel in 140 mL covered glass vessels. Sensory evaluation analysis were performed in a room equipped with a table and chairs, in a well-lit and ventilated sensory panel room and free of noise and odor at 21 °C degree room temperature. For the descriptive evaluation during the first session, the panelists were required to list their orthonasal odor impressions (during smelling). They described the odor on their memory protocol of the training sessions. After collecting the main odor attributes (consensus approval from more than half of the group of panelists), in the second session the panelists were asked to evaluate the intensities of these attributes on a scale from 0 (no perception) to 10 (strong perception). In addition, the panelists were asked to rate the overall odor intensity of the samples on a scale from 0 (no perception) to 10 (strong perception). A further hedonic rating of each sample (corresponding to the pleasantness or unpleasantness of the smell) was based on a scale from 0 (very unpleasant) via 5 (neutral) to 10 (very pleasant).

### 2.4. Solvent extraction of volatile compounds

Volatiles were extracted from ~7 g (±0.01 g) of oak shavings by stirring in 150 mL dichloromethane in an iodine-determination flask for ~1.5 h at room temperature. The extract was passed through a folded filter paper (Whatman No. 2; WH1202-240) to remove non-volatile components, followed by solvent assisted flavour evaporation (SAFE) as described by Engel, Bahr and Schieberle (Engel, Bahr, & Schieberle, 1999). The resulting distillate was then dried over anhydrous sodium sulfate and concentrated to a total volume of ~100 μL at 50 °C by Vigreux distillation and microdistillation (Bemelmans, 1979). The final distillate was then stored at –80 °C and analysed within 3 weeks maximum, but commonly as freshly as possible.

### 2.5. Comparative aroma extract dilution analysis

The odor potency of the aroma-active compounds was obtained using a dilution to threshold method, the so-called comparative aroma extract dilution analysis (cAEDA). Based on a series of dilutions, flavor dilution (FD) factors were obtained. The same sample preparation and dilution protocol was used in each case (Buettner & Schieberle, 2001a; Grosch, 2001). The original solvent distillates of the samples (FD1) were diluted stepwise, with dichloromethane by volume (ratio 1:1 v/v), resulting in 10 solutions for each sample corresponding to the FD factors FD2 to FD1024. For each odorant, the FD-factor represented the last solution in which the aroma was still perceivable. A 2 μL aliquot of all dilutions and 2 μL of the original extract (FD-factor = 1) were then analyzed by GC-O using DB-FFAP capillary column. The cAEDA values were calculated based on the average of three independent measurements.

### 2.6. Gas chromatography–olfactometry (GC-O)

GC-O was performed using a helium TRACE GC Ultra (Thermo Fisher Scientific, Waltham, MA, USA) equipped with either a DB-FFAP (30 m × 0.32 mm fused silica capillary, free fatty acid phase (FFAP), film thickness 0.25 μm) or DB-5 column (30 m × 0.32 mm fused silica capillary DB-5, film thickness 0.25 μm), both supplied by J & W Scientific (Agilent Technologies, Santa Clara, CA, USA). 2 μL were injected manually into the GC system using the cold on-column technique at 40 °C. After 2 min, the temperature of the GC containing the DB-FFAP capillary was increased at 8 °C/min to 240 °C, whereas using the DB-5 capillary the oven temperature was increased at the same rate to 250 °C. On both columns, the final temperature was held for 5 min. The flow rate of the helium carrier gas was 2.2 mL/min. At the end of the capillary, the effluent was split into two equal parts and transferred to a sniffing port and a flame ionization detector (FID) in the ratio 1:1 using a Y-glass splitter equipped with two deactivated, uncoated fused silica capillaries (0.5 m × 0.2 mm). The temperature of the FID and the sniffing port were held at 270 °C and 250 °C, respectively. GC-O analysis was performed by up to three trained panellists from both genders. Expert panelists were asked to sniff the oak sample, with sniffing durations for each panelist of 40 min on DB-5 and 37 min on FFAP.

### 2.7. Gas chromatography-mass spectrometry/olfactometry (GC–MS/O)

Samples were analyzed using a TRACE GC Ultra coupled to a TRACE DSQ mass spectrometer (both from Thermo Electron Corporation, Waltham, MA, USA) equipped with the DB-FFAP or DB-5 (30 m × 0.25 mm, film thickness 0.25 μm, Agilent technologies, Santa Clara, USA) columns in the same dimensions as described above. The cold on-column technique using 2 μL sample was applied at 40 °C using an MPS2 multipurpose autosampler (Gerstel GmbH & Co.KG, Mühlheim an der Ruhr, Germany). The flow rate of the helium carrier gas was 3.3 mL/min. At the end of the capillary column, the effluent was split into an odor detection port and a mass spectrometer using deactivated, uncoated fused silica capillaries and a Y-splitter as described above. Mass spectra were recorded in electron impact mode with an ionization energy of 70 eV over the *m/z* range 35–300. Data were collected and analyzed using Xcalibur v1.4 (Thermo Fisher Scientific).

### 2.8. Heart-cut two-dimensional gas chromatography–mass spectrometry/olfactometry (2D-GC–MS/O)

Trace constituents in the samples were identified by heart-cut 2D-GC–MS/O using a system comprising two Varian CP 3800 gas chromatographs combined with a Saturn 2200 Ion Trap mass spectrometer (all equipment supplied by Agilent Technologies, Darmstadt, Germany). The analytes were separated on a DB-FFAP capillary in the first oven, before the transfer of selected compounds to the CTS1 cryo-trap system at –100 °C using the MCS2 multi-column switching system (both devices supplied by Gerstel). Following thermodesorption at 250 °C, the volatiles were transferred to the second oven and were separated on a DB-5 capillary as described above.

Thereby, the cold on-column technique was used to apply 2 μL of each FD1 of the samples to the 2D-HRGC–MS/O system using the MPS 2XL multipurpose sampler (Gerstel) at 40 °C. After 2 min, the temperature of the first oven was increased at 8 °C/min to 230 °C and held for 5 min. The temperature of the second oven was increased at the same rate from 40 °C to 250 °C and held for 1 min. At the end of the first capillary, the effluent was split into a sniffing port (290 °C) and a FID (250 °C). The effluent of the second GC was split again in a 1:1 ratio to transfer the volatiles both to the mass spectrometer and a sniffing port (290 °C). Mass spectra in the electron impact (MS-EI) mode were generated at 70 eV over the *m/z* range 35–300. The cut time intervals were calculated by injecting the reference substances or according to the perception of an odor at the olfactory detection port (ODP).

### 2.9. Identification criteria of odorants

Retention indices were calculated for each aroma-active compound based on a reference series of homologous alkanes ($C_6$–$C_{34}$) (van Den Dool & Dec. Kratz, 1963). Odorants were identified based on three comparisons. These were odor qualities, odorant linear retention indices

compared to those of authentic reference compounds on columns with different polarities (DB-FFAP and DB-5) and on mass spectra compared with the spectra of original reference compounds or to reference spectra sourced from the NIST Mass Spectral Library v2.0d (National Institute of Standards and Technology, USA).

## 3. Methods for the quantification of selected odorants

For quantitative analysis of selected odorants, the instrumental analyses and sample preparation detailed in the following sections were performed. For analytical measurements, the ODP was installed as standard configuration of the instruments, but for quantitative analysis the ODP was not applied for human olfactory evaluation.

### 3.1. Sample preparation for quantification of odorous constituents

The quantification of the selected odorants was carried out by means of SIDA. To this aim, each suspension of 250 mL dichloromethane and 10 g of the oak samples was additionally spiked with known amounts of the ten isotopically labeled standards and stirred for 3 h at room temperature. Therefore, the extraction time for the quantification of the ador-active compounds with DCM was extended to 3 h, to ensure sufficient recovery of the odorants (Guth & Grosch, 1990; Schieberle & Grosch, 1987). The subsequent sample preparation including filtration, distillation, drying, and concentration steps were the same as described before.

### 3.2. Instrumental analyses used for quantification

The resulting concentrated distillates were analysed using 2D-GC–MS and GC–MS instruments. Thereby, hexanal, $\alpha$-pinene, nonanal, (*Z*)-3-hexenal, (*E*)-2-nonenal, eugenol, cis- & *trans*-whiskey lactone, and vanillin, were quantified via their mass spectra generated by means of 2D-GC–MS (same parameters as detailed before) in chemical ionization (CI) mode (*m/z* range 35–249) with methanol as reagent gas. Quantification of decanoic acid and acetic acid were accomplished via one-dimensional GC–MS in single ion monitoring (SIM) EI mode. For both systems, analyte/standard mixtures in known concentration (5:1, 3:1, 1:1, 1:3, 1:5, v/v) were analyzed, and calibration functions were calculated based on the relative intensities of the selected *m/z* ions. Peak areas corresponding to stable isotope-labeled standard and analyte were obtained from the extracted ion chromatograms as detailed in Table 4. The concentration of each target odorant in the oak wood samples was then calculated from the area counts of the respective labeled standard peak and the area counts of analyte peak, the amount of sample used, and the amount of standard added, by employing a calibration line equation (Czerny & Grosch, 2000; Lopez Pinar, Rauhut, Ruehl, & Buettner, 2017). Quantification of all mentioned odorants was performed in duplicates.

### 3.3. Calculation of OAVs

To obtain an estimation of the potential contribution of the respective odor-active compounds to the overall odor of the oak wood, OAVs were calculated for each quantified odorant. Generally, an odor-active compound with an OAV $\geq 1$ is expected to potentially contribute to the overall odor impression of a sample (Grosch, 2001). However, this generalized concept does not take into consideration potential additive, synergistic or suppressive effects; yet, it has shown to be, over decades, a valid and straightforward approach to focus on those compounds that are amongst the main contributors to specific smells. The OAVs are thereby determined by dividing the concentration of the odorants by their odor threshold in a specific medium. Ideally, that medium should be as close as possible to the respective matrix. However, comprehensive threshold data for the diversity of matrix materials, and especially complex systems such as wood, are not at hand, so that, for an approximation, water or oil is commonly used. A series of odor threshold values in water have been reported in literature, partially with large variation, depending on how the experimental determination has been achieved, and on the subjects and panel numbers involved (Buettner & Schieberle, 2001b, 2001c; Buttery, Ling, & Stern, 1997; Milo & Grosch, 1993; Pino & Mesa, 2006; Schuh & Schieberle, 2006). Here are values reported that have been determined complying comparative protocols with the same general procedure, namely in case of hexanal (10 μg/L), $\alpha$-pinene (41 μg/L), nonanal (8 μg/L), (*Z*)-3-hexenal (0.21 μg/L), (*E*)-2-nonenal (0.69 μg/L), eugenol (2.5 μg/L), acetic acid (180,000 μg/L), vanillin (210 μg/L) and decanoic acid (2200 μg/L) (Czerny et al., 2008). For whiskey lactone (23 μg/L), the odor threshold was newly determined within this study, also on basis of a using triangle test. Based on these values, OAVs $\geq 1$ were obtained for all the quantified odorants except decanoic acid and acetic acid. Thereby, acetic acid gave an OAV $\geq 1$ value in samples G2-B, A2-NO, G1-B, G3-B, H-B and A1-NO, whereas the rest of the samples was determined with an OAV for acetic acid below 1.

### 3.4. Aroma recombination experiments

Recombination experiments were performed to validate the obtained quantitative data of the oak samples. For this purpose, a stock solution of all selected odorants was prepared in ethanol for the aroma model. For the creation of an appropriate matrix, a defined amount of oak was shaved and then extracted several times with solvents of different polarities (methanol, acetone, dichloromethane, hexane, and pentane) until the residue was odorless. The deodorized oak matrix was then spiked with the determined amounts of all quantified aroma compounds that had given an odor activity value greater or equal to one (OAV $\geq 1$). The final aroma model was compared with the original oak sample by the panelists by performing an odor profile analysis (Christlbauer & Schieberle, 2011).

### 3.5. Statistical analysis

First, all data were standardized using the z-score. Prior to statistics analysis, the raw data of the sensory results were tested for outliers using the Grubbs test. Outliers were removed and then the data were tested for normality using the Jarque-Bera tests, in which they were found to be normally distributed. The sensory analysis data were tested for outliers (Gruber test) and normal distribution (Jarque-Bera test) using XLSTAT 2019® (Addinsoft, Paris, France). They were then averaged and plotted in spider-web diagrams using Excel 2016®. Product characterisation test was then applied to identify the discrimination between sensory attributes. Two-way analysis of variance (ANOVA) followed Tukey test (a $p$-value $< 0.05$ was considered significant) was carried out. Prior to Pearson correlation and PCA ploting, the data (intensity ratings of sensory attributes and FD factors of odorants) were standardized using the z-score, as the data were expressed in different unites. Pearson correlation test was then conducted between the significantly sensory attributes and the OD values of odorants. To investigate the effect of geographical origin of the investigated trees on their odor profile, the sensory data, cAEDA and its significantly correlated odorant quantities were selected for running the principal component analysis (PCA) using XLSTAT 2019® (Addinsoft, Paris, France).

## 4. Results

### 4.1. Sensory evaluations

The overall odor intensity (ODI) of G6-BW was rated as the most intense of all samples (ODI = 8.0) followed by sample G2-B with the rate of ODI = 7.5 With respect to the hedonic evaluation, G6-BW also was rated as the most pleasant with a value of 6.5. The Hungarian oak (H-B), however, was rated as less pleasant with a value of 3.5 (Fig. 1, File 2 in

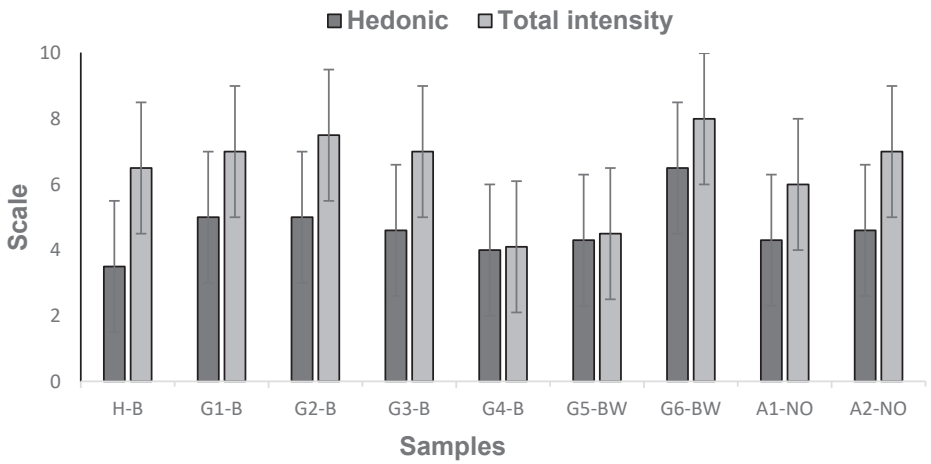


**Fig. 1.** Hedonic scaling and overall odor intensity of all oak samples in direct comparison. Data are displayed as mean values of *ortho*-nasal sensory evaluation. The abbreviations are referred to in Table 1.

supplementary material and the oak samples were specified in material and method section Table 1.).

ANOVA test results with their corresponding standard deviation are shown in File 2 and in detail in File 3 in supplementary material. Considering of X as samples and Y as descriptors, as for the source of variation, indicated that the interaction between variables is 22.46, sample type 4.38%, and descriptors is 37.4%.

The sensory analysis revealed a consensus on nine odor attributes for all samples; these were resin-like; cork-like, musty; fatty cardboard-like; rancid; woody; vinegar-like; green-grassy; coconut-like; and citrus-like. Fig. 2 displays the odor profiles of the nine samples by showing the respective perceived intensities as mean values across the panel for each attribute. The odor attribute yielding the highest intensities in the samples G2-B, G1-B, G3-B, A2-NO, H-B, and A1-NO was vinegar-like, which was scored with rank intensities between 6 and 7.5, being one of the leading contributors to the overall smell profiles of these samples. Oak sample G6-BW, on the other hand, was dominated by a coconut-like odor with an intensity rating of 8.0, and this attribute was also important in samples G1-B, G4-B and G5-BW. Apart from that, a clear resin-like odor impression was perceived with a mean value of 6.0 in the Hungarian oak. The attribute green-grassy was scored also higher in this oak with a value of 5.0 than in the remaining samples. Only one sample (G2-B) revealed a relatively high rating of 5.0 for the attribute cork-like, musty. Four samples (G4-B, G1-B, G2-B, and G3-B) exhibited a medium fatty, cardboard-like smell with values of 4.0–5.5.

To identify the discrimination power of the sensory attributes of oak samples, the product characterization test was additionally applied. The results showed that the descriptors coconut-like, green-grassy, vinegar-

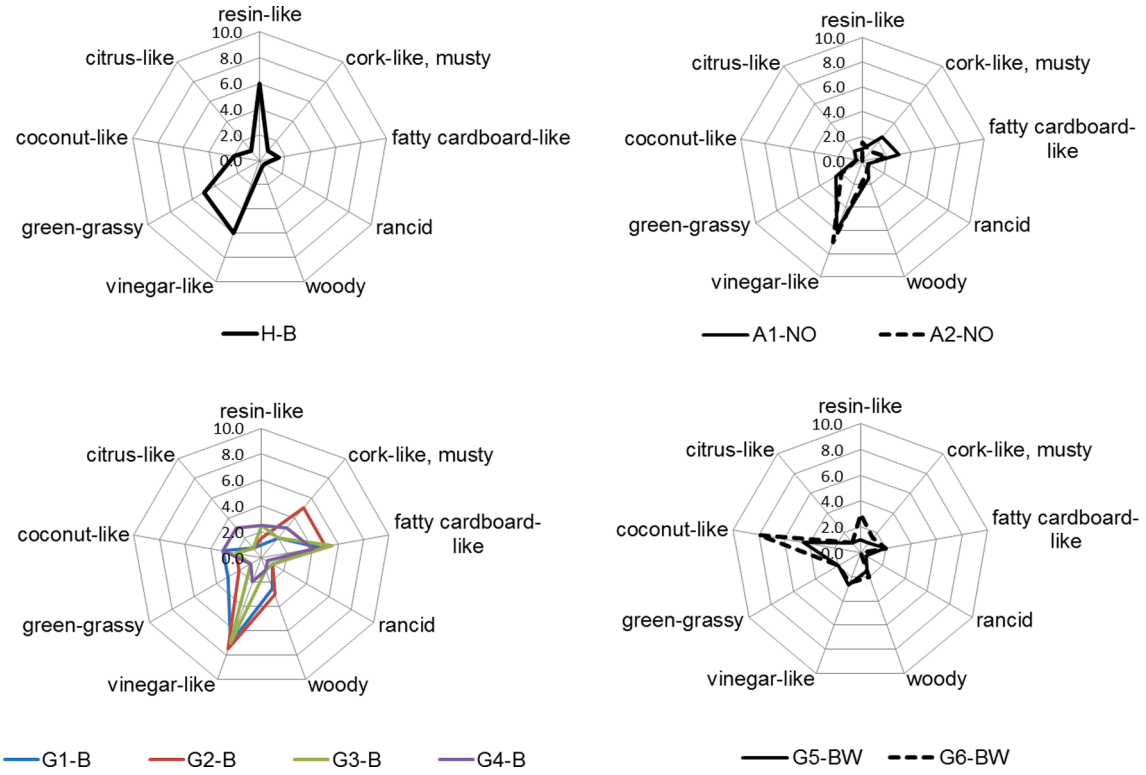


**Fig. 2.** Odor profiles of oak samples: the intensities of the respective attributes were rated on a scale from 0 (no perception) to 10 (strong perception) for each sample.

like, and resin-like were the most important attributes to discriminate between samples (Table 2).

### 4.2. cAEDA and identification of odorants in oak

For further elaboration of the most potent odorants, the oak samples were subjected to cAEDA with analysis of dilution steps corresponding to flavor dilution (FD) factors in the range from FD 2 to FD 1024. Thereby, cAEDA analysis revealed 16 compounds that were detectable in the highly diluted extracts of all oak samples in the range from FD128 to FD1024. Among these potent odorants were several resin-like, green-grassy, citrus-like, fatty cardboard-like, vinegar-like, coconut-like, cinnamon-like, clove-like, and woody smelling substances. The most odorous constituents with FD $\geq$ 128 in all samples were identified by means of GC–MS and 2D-GC–MS/O based on their odor description, retention indices, and mass spectra. Using these techniques we successfully identified $\alpha$-pinene (resin-like), hexanal (green-grassy), (*Z*)-3-hexenal (green-grassy), $\beta$-myrcene (earthy, green-grassy), ocimene (citrus-like, green-grassy, fruity), 1-octen-3-one (mushroom-like), nonanal (soapy, citrus-like), acetic acid (vinegar-like), (*E*)-2-nonenal (fatty cardboard-like), whiskey-lactone (coconut-like), cinnamaldehyde (cinnamon-like), eugenol (clove-like), decanoic acid (plastic-like, waxy, soapy), phenylacetic acid (honey-like, flowery), vanillin (vanilla-like), and thymoquinone (pencil-like). These substances were identified as potent odor-active volatiles in all oak samples. Furthermore, syringaldehyde (green, woody), mustakone (woody), and p-cymene (oregano-like, woody, herbal, green-grassy) were found to be potent odorants in samples G1-B and G2-B. Pentanoic and butanoic acid with cheesy smell were found to be potent in samples G1-B, G3-B, G5-BW, and G6-BW, whereas 2,4,6-trichloroanisole (musty, cork-like) and fenchol (earthy, musty) were amongst the most potent odorants in G2-B, G3-B, and G4-B, and camphene (green, camphor-like) in samples H-B and G4-B. In sample H-B, the compounds linalool (flowery, citrus-like, fresh) and $\alpha$-humulene (resin-like, woody) were detected with FD factors $\geq$ 256. Smoky, vanilla-like smelling guaiacol and flowery, fresh smelling geraniol were shown to be detectable with high intensity in samples G2-B, G4-B, and G6-BW. With regard to the coconut-like character, $\gamma$-octalactone and $\delta$-decalactone were perceived with FD factors $\geq$ 128 in samples G1-B, G2-B, G3-B, G4-B, G5-BW, and G6-BW, and whiskey lactone was also present with high FD factors in some samples, especially in G6-BW with FD 1024. Moreover, $\alpha$-terpineol (eucalyptus-like, mint-like) was detected with high FD-factors in samples G1-B and G4-B, $\alpha$-bisabolol (clove-like, flowery) in samples A1-NO and G4-B, and the metallic smelling tr-4,5-epoxy-(*E*)-2-decenal in samples A1-NO and G5-BW. An overview of the obtained results of the identification and AEDA experiments are given in Table 3.

The heat map data visualisation on the basis of FD-factors of AEDA analysis (Fig. 4) shows that the fatty acids represented the largest group of odor-active constituents in oak wood.

**Table 2**
The product characterization test results are displayed with descriptors and their corresponding test value.

| Descriptors | Test values |
|---|---|
| coconut-like | 8.8 |
| green-grassy | 6.9 |
| vinegar-like | 5.9 |
| resin-like | 5.8 |
| fatty cardboard-like | 4.6 |
| citrus-like | 4.5 |
| cork-like, musty | 4.0 |
| rancid | 3.8 |
| woody | 1.8 |

### 4.3. Quantification of selected odorants

The quantification of odorants in the oak samples was carried out by means of SIDA. Ten target odorants were selected based on high FD-factors of these substances (FD-factors $\geq$ 128) in all samples (Tables 4 and 5). The following substances were selected for quantification: hexanal, $\alpha$-pinene, nonanal, (*Z*)-3-hexenal, (*E*)-2-nonenal, eugenol, acetic acid, decanoic acid, cis- and *trans*-whiskey lactone, and vanillin. The main consideration for the selection of odorants for quantification experiments was that these compounds were identified as potent odorants in all samples as well as being one of those constituents representing one of the three main substances classes of woods. Thereby, the aim was not to choose all of the most odor-active constituents but to cover most of the prevalent substance classes. Quantitation experiments revealed that the selected odorants were detectable within a wide range of concentration. Regarding the analyzed acids (acetic acid and decanoic acid), a concentration range from 110.5 to 299 mg/kg was obtained for acetic acid, and of 122 to 335 μg/kg for decanoic acid. The highest concentrations of acetic acid were obtained for oak sample G2-B (299 mg/kg). In the case of decanoic acid the highest concentration was determined in sample G3-B (335 μg/kg), and the lowest in A2-NO (122 μg/kg).With regard to the group of aldehydes, hexanal was detected at much higher levels in H-B and A1-No (1255 μg/kg and 924 μg/kg, respectively) than in the other oak samples, whereas (*Z*)-3-hexenal yielded the highest concentration in sample H-B, and nonanal in sample G4-B. The phenolic compound vanillin was determined within a concentration range from 4026 μg/kg in G4-B sample to 6714 μg/kg in G6-BW oak sample, whereas eugenol was present in elevated concentrations in samples G5-BW, and G6-BW with 1144 μg/kg and 1210 μg/kg, respectively. Moreover, high concentrations of *trans*-whiskey lactone were found in samples G5-BW and G6-BW, whereas *cis*-whiskey lactone was determined with higher levels in samples G4-B and G6-BW. The determined concentrations of the odorants are compiled in Table 5.

### 4.4. Odor recombination experiments

To confirm the screening by AEDA and OAV calculation for the main odorants of the oak samples, additional experiments were carried out reconstituting the determined odorant profiles. Thereby, recombination experiment for G3-B exemplarily was prepared since this sample elicited the high overall odor intensity. The developed odorless oak matrix was therefore spiked with an ethanolic solution comprising the odor-active compounds in their quantitated concentrations. The reconstituted oak smell sample together with the originally analyzed sample were then presented to a trained panel. The two samples were compared by rating the intensities of the predefined odor attributes, as done in the initial odor profile analyses. The smell of the model containing all odorants quantified in oak thereby revealed a very good similarity to the aroma of the oak sample itself. The odor profile analysis showed that both G3-B and the recombinate elicited the same intensities for the odor qualities resin-like, citrus-like, rancid, and coconut-like, and nearly identical intensities for the odor qualities fatty cardboard-like as well as vinegar-like. Only the odor attribute green-grassy was rated slightly higher in the aroma model compared to the odor profile of the original sample. The overall odor of the model was, accordingly, rated to be in good similarity to that of the oak sample. The overall intensity of the recombinate was rated with a median value of 6.5, whereas the overall intensity in the oak sample was slightly higher with a value of 7.0. The results of the sensory evaluation of the oak sample compared to its recombinate are displayed in Fig. 3.

### 4.5. Geometric projection of data using PCA

Principle component analysis was performed to understand the variation of smell character among the oak samples from different locations in Germany, Austria, and Hungary. For PCA analysis, we only

**Table 3**
Identified aroma-active substances, their odor qualities, retention indices, and flavor dilution (FD) factors in oak wood samples.

| No. | Odorant | Odor quality[a] | RI value[b] on | | FD-factor[c] on DB-FFAP | | | | | | | | | Identification |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | DB-FFAP | DB-5 | H-B | A1-NO | A2-NO | G1-B | G2-B | G3-B | G4-B | G5-BW | G6-BW | |
| 1 | 3-Methyl butanal | malty | 978 | 654 | 64 | 16 | 64 | 4 | 4 | 4 | 4 | 8 | 128 | e |
| 2 | 2,3-Butanedione (Diacetyl) | butter-like | 983 | 605 | 16 | 256 | 64 | 128 | 128 | ≤ 1 | 128 | 128 | ≤ 1 | e |
| 3 | Pentanal (Valeraldehyde) | malty, yeast dough-like | 1005 | 718 | 256 | 512 | 256 | 16 | 128 | 32 | 512 | 64 | 128 | f |
| 4 | α-Pinene | resin-like | 1011 | 940 | 1024 | 128 | 128 | 128 | 256 | 512 | 512 | 128 | 512 | g |
| 5 | Camphene | green, camphor-like | 1049 | 954 | 128 | 64 | 16 | 32 | 64 | 64 | 512 | 64 | 64 | g |
| 6 | n-Butyl acetate | fruity, solvent-like | 1080 | 825 | 32 | 512 | 64 | 32 | 128 | 256 | 64 | 256 | 512 | f |
| 7 | Hexanal | green-grassy | 1083 | 801 | 1024 | 512 | 256 | 512 | 256 | 128 | 256 | 256 | 256 | f |
| 8 | 3-Methylbutyl acetate | fruity, banana-like | 1110 | 875 | 256 | 64 | 256 | 256 | 512 | 512 | 64 | 512 | 128 | f |
| 9 | (*Z*)-3-Hexenal | green-grassy | 1140 | 790 | 512 | 128 | 256 | 128 | 128 | 128 | 128 | 256 | 128 | f |
| 10 | 1,2-Dimethyl benzene (o-xylene) | geranium-like, fatty | 1158 | 897 | 32 | 16 | 256 | 64 | 16 | 128 | 256 | 256 | 512 | f |
| 11 | β-Myrcene | earthy, green-grassy, geranium-like | 1160 | 990 | 256 | 512 | 256 | 128 | 512 | 512 | 256 | 256 | 128 | g |
| 12 | (*R*)-Limonene | orange peel-like | 1177 | 1025 | 64 | 16 | 64 | 256 | 64 | 256 | 64 | 16 | 64 | g |
| 13 | Heptanal | green-grassy, fatty, soapy | 1180 | 901 | 128 | 32 | 64 | 16 | 16 | 128 | 512 | 16 | 128 | g |
| 14 | 1,8-Cineole (Eucalyptol) | eucalyptus-like, menthol-like | 1190 | 1030 | 128 | 8 | 16 | 16 | 64 | 64 | 64 | 16 | 128 | g |
| 15 | Ocimene | citrus-like, green-grassy, fruity | 1235 | 1068 | 256 | 512 | 512 | 128 | 128 | 128 | 512 | 256 | 128 | g |
| 16 | p-Cymene | oregano-like, woody, herbal, green-grassy | 1270 | 1029 | 64 | 64 | 16 | 512 | 16 | 128 | 16 | 32 | 64 | g |
| 17 | Octanal | fatty, soapy citrus-like | 1280 | 998 | 256 | 256 | 64 | 64 | 128 | 64 | 512 | 256 | 128 | f |
| 18 | 1-Octen-3-one | mushroom-like | 1291 | 979 | 128 | 256 | 256 | 256 | 256 | 256 | 256 | 256 | 128 | f |
| 19 | 2-Acetyl-1-pyrroline | roasty, popcorn-like | 1330 | 926 | 64 | 32 | 32 | 16 | 128 | 16 | 64 | 32 | 64 | g |
| 20 | Nonanal | soapy, citrus-like | 1382 | 1104 | 256 | 512 | 256 | 256 | 256 | 256 | 512 | 256 | 256 | g |
| 21 | α-Thujone | minty, menthol-like | 1398 | 1120 | 8 | 64 | 32 | 32 | 128 | 128 | 16 | 32 | 128 | g |
| 22 | (*E*)-2-Octenal | fatty, woody, grassy | 1416 | 1058 | 64 | 32 | 128 | 256 | 256 | 32 | 256 | 16 | 64 | f |
| 23 | δ-3-Carene | citrus-like, eucalyptus-like | 1429 | 1230 | 32 | 64 | 32 | 32 | 16 | 16 | 64 | 32 | 8 | f |
| 24 | Acetic acid | vinegar-like | 1445 | 619 | 512 | 512 | 512 | 512 | 1024 | 512 | 128 | 256 | 256 | g |
| 25 | 2-Furaldehyde (Furfural) | almond-like, woody, roasty | 1457 | 836 | 64 | 128 | 32 | 64 | 16 | 64 | 256 | 64 | 64 | e |
| 26 | Unknown | cucumber-like | 1480 | – | 64 | 256 | 512 | 64 | 256 | 256 | 64 | 32 | 128 | – |
| 27 | Benzaldehyde | bitter almond-like, marzipan-like | 1490 | 964 | 64 | 64 | 32 | 32 | 64 | 32 | 64 | 32 | 64 | e |
| 28 | (*E*)-2-Nonenal | fatty, cardboard-like | 1522 | 1160 | 128 | 128 | 128 | 256 | 512 | 512 | 256 | 128 | 128 | g |
| 29 | Isopulegol | peppermint, fresh | 1536 | 1145 | 32 | 64 | 64 | 32 | 256 | 64 | 16 | 16 | 64 | g |
| 30 | Linalool | flowery, citrus-like, fresh | 1550 | 1105 | 512 | 64 | 32 | 32 | 16 | 16 | 8 | 32 | 64 | g |
| 31 | (*E,Z*)-2,6-Nonadienal | cucumber-like | 1578 | 1159 | 256 | 64 | 256 | 32 | 512 | 512 | 64 | 64 | 512 | g |
| 32 | Fenchol | earthy, musty | 1580 | 1141 | 32 | 64 | 64 | 16 | 256 | 128 | 256 | 64 | 64 | f |
| 33 | β-Caryophyllene | earthy, green, clove-like | 1588 | 1433 | 64 | 64 | 256 | 64 | 256 | 64 | 16 | 64 | 128 | g |
| 34 | Butanoic acid | cheesy | 1611 | 805 | 32 | 128 | 32 | 256 | 64 | 512 | 64 | 256 | 512 | f |
| 35 | 2-Propenoic acid | geranium-like, vinegar-like | 1621 | – | 64 | 256 | 256 | 64 | 64 | 128 | 64 | 512 | 512 | f |
| 36 | (*E*)-2-Decenal | fatty, green, coriander-like | 1627 | 1262 | 16 | 32 | 512 | 256 | 32 | 64 | 16 | 16 | 512 | g |
| 37 | Phenylacetaldehyde | honey-like, flowery | 1640 | 1044 | 32 | 128 | 64 | 64 | 16 | 16 | 16 | 16 | 64 | g |
| 38 | Verbenone | champhoreous, mentholic | 1646 | 1200 | 256 | 16 | 16 | 256 | 128 | 512 | 16 | 16 | 16 | g |
| 39 | α-Humulene | resin-like, woody | 1653 | 1467 | 256 | 32 | 64 | 16 | 64 | 16 | 32 | 16 | 64 | g |
| 40 | 3-Methylbutanoic acid | cheesy | 1662 | 869 | 128 | 128 | 32 | 64 | 64 | 64 | 64 | 512 | 256 | f |
| 41 | Estragole | anise-like | 1675 | 1198 | 64 | 64 | 16 | 16 | 64 | 16 | 16 | 64 | 64 | g |
| 42 | (-)-Borneol | earthy, moldy | 1680 | 1188 | 64 | 64 | 16 | 16 | 64 | 64 | 16 | 64 | 64 | g |
| 43 | α-Terpineol | eucalyptus-like, mint-like | 1685 | 1196 | 64 | 32 | 32 | 512 | 32 | 32 | 128 | 64 | 64 | g |
| 44 | Germacrene D | fruity, green, woody | 1690 | 1490 | 32 | 16 | 32 | 256 | 32 | 32 | 16 | 64 | 256 | g |
| 45 | (*L*)-Carvone | minty, spicy, fresh | 1710 | 1255 | 32 | 32 | 16 | 16 | 256 | 32 | 16 | 16 | 16 | g |
| 46 | Pentanoic acid | cheesy | 1731 | 911 | 32 | 16 | 16 | 256 | 16 | 512 | 64 | 128 | 256 | f |
| 47 | (*E*)-β-Farnesene | fruity, cucumber-like, woody | 1748 | 1458 | 8 | 128 | 64 | 32 | 16 | 128 | 16 | 16 | 16 | g |
| 48 | Propanoic acid | cheesy, vinegar-like | 1750 | 875 | 64 | 32 | 16 | 32 | 16 | 32 | 16 | 512 | 16 | f |
| 49 | Cuminaldehyde | woody, cumin-like | 1766 | 1394 | 8 | 16 | 16 | 512 | 128 | 16 | 16 | 64 | 128 | f |
| 50 | (*E*)-β-Damascenone | grape juice-like | 1775 | 1386 | 128 | 16 | 16 | 64 | 256 | 32 | 64 | 16 | 64 | g |
| 51 | (*E*)-2-Butenoic acid | cheesy | 1786 | – | 64 | 256 | 128 | 64 | 32 | 512 | 32 | 64 | 32 | f |
| 52 | Nerol | flowery | 1795 | 1233 | 64 | 128 | 64 | 64 | 256 | 128 | 64 | 16 | 64 | g |
| 53 | β-Citronellol | lemongras-like, flowery | 1802 | 1253 | 128 | 64 | 256 | 64 | 64 | 64 | 32 | 64 | 16 | g |
| 54 | 2,4,6-Trichloroanisole | musty, cork-like | 1807 | 1355 | 32 | 64 | 32 | 64 | 512 | 128 | 256 | 16 | 64 | g |
| 55 | Geraniol | flowery, fresh | 1841 | 1244 | 32 | 64 | 8 | 64 | 512 | 32 | 256 | 16 | 128 | g |
| 56 | Guaiacol | smoky, vanilla-like | 1860 | 1092 | 8 | 16 | 128 | 32 | 256 | 16 | 256 | 16 | 512 | g |
| 57 | 2-Methylhexanoic acid | musty, cheesy | 1863 | 1133 | 32 | 16 | 16 | 64 | 64 | 16 | 8 | 16 | 16 | f |
| 58 | 5-Methylpentanoic acid | cheesy | 1885 | – | 256 | 16 | 256 | 64 | 64 | 64 | 64 | 64 | 64 | f |
| 59 | | metallic | 1888 | 1277 | 128 | 128 | 256 | 64 | 256 | 512 | 512 | 256 | 512 | g |

(*continued on next page*)

**Table 3** (*continued*)

| No. | Odorant | Odor quality[a] | RI value[b] on | | FD-factor[c] on DB-FFAP | | | | | | | | | Identification |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | DB-FFAP | DB-5 | H-B | A1-NO | A2-NO | G1-B | G2-B | G3-B | G4-B | G5-BW | G6-BW | |
| | tr-4,5-Epoxy-(*E*)-2-nonenal | | | | | | | | | | | | | |
| 60 | 2-Phenylethanol | rose-like | 1900 | 1135 | 16 | 128 | 128 | 128 | 64 | 128 | 256 | 256 | 128 | f |
| 61 | γ-Octalactone | coconut-like | 1913 | 1263 | 64 | 64 | 64 | 512 | 128 | 128 | 256 | 128 | 512 | f |
| 62 | Heptanoic acid | cheesy | 1930 | 1087 | 32 | 64 | 64 | 16 | 64 | 64 | 512 | 16 | 16 | f |
| 63 | Benzothiazole | car tire-like, rubber-like | 1939 | 1238 | 16 | 8 | 16 | 8 | 16 | 4 | 8 | 8 | 8 | g |
| 64 | Whiskey lactone | coconut-like | 1950 | 1309 | 128 | 128 | 128 | 128 | 128 | 128 | 256 | 512 | 1024 | g |
| 65 | Maltol | caramel-like | 1968 | 1120 | 32 | 32 | 32 | 16 | 16 | 32 | 256 | 16 | 256 | g |
| 66 | γ-Nonalactone | coconut-like | 1980 | 1360 | 256 | 128 | 128 | 32 | 64 | 256 | 256 | 64 | 256 | g |
| 67 | tr-4,5-Epoxy-(*E*)-2-decenal | metallic | 1997 | 1372 | 32 | 1024 | 32 | 64 | 64 | 64 | 128 | 512 | 128 | g |
| 68 | Phenol | phenolic, ink-like | 2009 | 911 | 16 | 128 | 16 | 16 | 16 | 16 | 16 | 16 | 16 | g |
| 69 | p-Anisaldehyde | woodruff-like | 2045 | 1283 | 8 | 16 | 16 | 16 | 64 | 8 | 16 | 16 | 8 | g |
| 70 | δ-Nonalactone | coconut-like | 2052 | 1345 | 256 | 16 | 16 | 16 | 128 | 32 | 256 | 64 | 32 | f |
| 71 | Octanoic acid | cheesy | 2058 | 1249 | 32 | 128 | 32 | 32 | 128 | 256 | 64 | 256 | 32 | f |
| 72 | 4-Methylphenol (p-Cresol) | horse stable-like | 2078 | 1084 | 128 | 64 | 16 | 32 | 32 | 16 | 32 | 256 | 128 | g |
| 73 | Cinnamaldehyde | cinnamon-like | 2080 | 1280 | 512 | 128 | 128 | 512 | 128 | 128 | 256 | 256 | 128 | g |
| 74 | Eugenol | clove-like | 2120 | 1361 | 256 | 128 | 256 | 256 | 128 | 512 | 512 | 512 | 512 | g |
| 75 | γ-Decalactone | peach-like, fruity | 2128 | 1469 | 16 | 64 | 16 | 32 | 64 | 64 | 512 | 64 | 32 | g |
| 76 | 3-Ethylphenol | animalic, leather-like | 2180 | 1171 | 8 | 256 | 256 | 128 | 128 | 64 | 64 | 64 | 64 | g |
| 77 | δ-Decalactone | coconut-like | 2190 | 1501 | 16 | 32 | 64 | 512 | 512 | 128 | 512 | 512 | 256 | g |
| 78 | Thymol | pencil-like, thyme-like | 2200 | 1296 | 32 | 16 | 128 | 64 | 16 | 256 | 256 | 128 | 128 | g |
| 79 | Acetyleugenol | pepper-like | 2233 | – | 32 | 32 | 16 | 64 | 128 | 32 | 256 | 128 | 256 | g |
| 80 | Rotundone | pepper-like | 2250 | 1716 | 32 | 256 | 64 | 64 | 512 | 256 | 512 | 64 | 256 | g |
| 80 | Mustakone | woody | 2256 | 1685 | 64 | 32 | 64 | 256 | 256 | 32 | 64 | 32 | 32 | g |
| 81 | α-Bisabolol | clove-like, flowery | 2265 | 1446 | 64 | 256 | 32 | 64 | 16 | 64 | 128 | 64 | 64 | f |
| 82 | Decanoic acid | plastic-like, waxy, soapy | 2278 | 1371 | 128 | 128 | 128 | 512 | 512 | 512 | 256 | 256 | 128 | f |
| 83 | Benzoic acid | balsamic | 2280 | 1198 | 32 | 32 | 16 | 16 | 16 | 8 | 16 | 8 | 8 | f |
| 84 | (*E,E*)-2,6-Farnesol | fatty, oily, fruity | 2338 | 1722 | 8 | 128 | 64 | 64 | 32 | 32 | 256 | 256 | 32 | g |
| 85 | 2-Methylundecanoic acid | soapy, fatty | 2398 | – | 32 | 128 | 32 | 512 | 128 | 256 | 128 | 256 | 128 | f |
| 86 | Dodecanoic acid | plastic-like, fatty | 2450 | 1550 | 64 | 128 | 256 | 128 | 128 | 64 | 256 | 256 | 256 | f |
| 87 | Ethylvanillin | vanilla-like | 2535 | 1486 | 64 | 256 | 256 | 256 | 256 | 256 | 256 | 256 | 256 | d |
| 88 | Phenylacetic acid | honey-like, flowery | 2555 | 1260 | 256 | 256 | 128 | 256 | 256 | 256 | 256 | 256 | 256 | g |
| 89 | Vanillin | vanilla-like | 2572 | 1400 | 512 | 1024 | 512 | 256 | 256 | 256 | 256 | 256 | 1024 | g |
| 90 | 3-Phenylpropanoic acid | cheesy, balsamic, flowery | 2611 | 1364 | 64 | 512 | 128 | 64 | 64 | 128 | 32 | 256 | 64 | g |
| 91 | Unknown | sweaty, androstenone-like | 2780 | 2060 | 512 | 128 | 64 | 64 | 64 | 32 | 32 | 32 | 256 | – |
| 92 | Unknown | sweaty, androstenone-like | 2887 | 2250 | 128 | 128 | 64 | 512 | 512 | 256 | 64 | 512 | 256 | – |
| 93 | Syringaldehyde | green, woody | 2924 | 1656 | 64 | 32 | 64 | 256 | 256 | 128 | 128 | 32 | 64 | g |
| 94 | Unknown | sweaty, androstenone-like | 2990 | 2278 | 128 | 256 | 64 | 64 | 64 | 128 | 256 | 256 | 256 | – |
| 95 | Thymoquinone | pencil-like | 3100 | 1250 | 256 | 128 | 128 | 128 | 128 | 128 | 256 | 512 | 256 | g |

a. Odor quality perceived at the sniffing port during GC-O analysis.
b. Retention index on capillaries DB-FFAP and DB-5 according to van Den Dool and Kratz (van Den Dool & Dec. Kratz, 1963).
c. FD: determined flavor-dilution factor according to Grosch (Grosch, 2001a).
d. The odorant was tentatively identified by comparison of odor quality and retention indices on capillaries DB-FFAP and DB-5.
e. Proposed structure by comparison of odor quality and retention index on capillaries DB-FFAP or DB-5.
f. The odorant was identified by comparison of odor quality, retention index on capillary DB-FFAP and mass spectrum (MS-EI), obtained by GC–MS/O analysis, with the properties of the reference compound.
g. The odorant was identified by comparison of odor quality, retention index on both capillaries and mass spectrum (MS-EI), obtained by 2D-GC–MS/O analysis, with the properties of the reference compound.
.

applied the chemical variables between oak samples, the sensory attributes that causes significant difference and their correlated quantified odorant (cf. section material and method, statistical analysis). Accordingly, these variable compounds comprised 11 quantified odorants, with their representative variable sensory attributes, namely: resin-like; citrus-like; vinegar-like; cork-like, musty; fatty cardboard-like; green-grassy and coconut-like, all are illustrated in a PCA-bi Plot in Fig. 5. Two principle components explained 60.56% of the variation. The first principle component (PC1) successfully differentiated between Hungarian oak and the other samples. PC1 is essentially positively defined by green-grassy, resin-like and vinegar-like sensory attributes correlated with sample H-B and moderately with samples A1-NO, A2-NO, G1-B, G2-B and G3-B as opposed to samples G5-BW, G6-BW and G4-B. The second principal component (PC2) distinguished between Bavarian and Austrian samples from one side and Hungarian and Baden-Württemberg oak samples from the other, except for G4-B oak from Bavaria. This is reflected by the positioning of the samples on the chart, with being positioned in separated quadrants (G6-BW, G5-BW, and G4-B oak samples in quadrant –PC1/+PC2, A1-NO, A2-NO, G1-B, G3-B in quadrant +PC1/-PC2 and G2-B with the little different from G3-B sample in quadrant -PC1/-PC2). Interestingly the first and second principal components differentiated between Hungarian oak and the other samples by the positioning the sample in separated quadrats than the others in +PC1/+PC2. The sensory evaluation of oak G4-B showed that this sample is dominated in citrus-like odor and cork-like, musty smell was more potent in sample G2-B which are also matched with PCA analysis.

**Table 4**
Selected mass traces and calibration functions of the odorants used for quantification by SIDA.

| Odorant | Isotope label | Ion (*m/z*) | | Calibration line equation | $R^2$ |
|---|---|---|---|---|---|
| | | Analyte | Standard | | |
| Hexanal | [$^2H_2$]-Hexanal | 153 | 155 | y = 0.8046x + 0.0049 | 0.9998 |
| *α*-Pinene | [$^2H_3$]-*α*-Pinene | 137 | 140 | y = 0.7675x + 0.0055 | 0.9994 |
| Nonanal | [$^2H_4$]-Nonanal | 143 | 147 | y = 0.6266x + 0.0037 | 0.9973 |
| (*Z*)-3-Hexenal | [$^2H_2$]-(*Z*)-3-Hexenal | 99 | 101 | y = 0.7107x + 0.0193 | 0.9924 |
| (*E*)-2-Nonenal | [$^2H_2$]-(*E*)-2-Nonenal | 123 | 125 | y = 0.9004x + 0.0416 | 0.9972 |
| Eugenol | [$^2H_3$]-Eugenol | 165 | 168 | y = 0.6609x + 0.0063 | 0.9973 |
| Acetic acid | [$^{13}C_2$]- Acetic acid | 60 | 62 | y = 1.1137x + 0.0042 | 0.9220 |
| Vanillin | [$^{13}C_6$]-Vanillin | 153 | 156 | y = 1.0023x + 0.0758 | 0.9992 |
| Decanoic acid | [$^2H_3$]-Decanoic acid | 172 | 175 | y = 1.0229x + 0.0029 | 0.9660 |
| *trans*-Whiskey lactone | [$^2H_6$]-*trans*-Whiskey lactone | 157 | 163 | y = 1.1255x + 0.0145 | 0.9913 |
| *cis*-Whiskey lactone | [$^2H_6$]-*cis*-Whiskey lactone | 157 | 163 | y = 1.2047x + 0.0011 | 0.9983 |

Based on the results showed in Fig. 1 (on terms of total intensity and hedonic values) and PCA analysis, no significant differences were found between Austrian and Bavarian oak samples. The position of the samples on the PCA is in some way comparable with the position of the oak wood samples on the location map. This implies that the environmental conditions, e.g. the distance between sample origins, might significantly impact on the cAEDA, odor profile, and the concentration of odorants in the samples. The near borders might be the cause of the similarities (Fig. 5).

## 5. Discussion

### 5.1. Odorant composition and potential odorant sources

In total 95 odorants were detected in all nine samples applying GC-O analysis as well as one-dimensional and two-dimensional GC–MS/O analyses, 91 of which successfully identified using one- and two-dimensional GC–MS analyses. Thereby, the vast majority of the identified compounds was found in all the samples with different flavor dilution (FD) factors. Most of the odorants that were identified in all of the samples had also been described in our previous study as constituents of Hungarian oak (*Quercus frainetto*) (Ghadiriasli, Wagenstaller, & Buettner, 2018). 2-Propenoic acid **35** (geranium-like, vinegar-like) and cinnamaldehyde **73** (cinnamon-like) are reported here for the first time as odorants in oak.

The identified odorants belong to diverse substance classes and exhibit a great variety of odor characteristics. Many these aroma-active

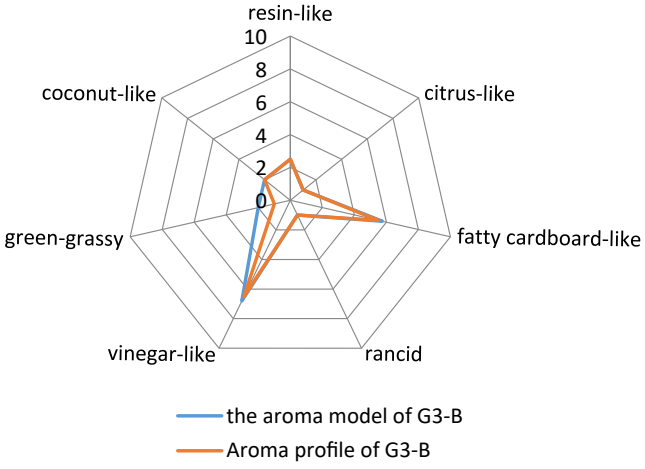


**Fig. 3.** Results of the comparative odor profile analysis for the G3-B oak sample and its corresponding recombinate. Abbreviations are referred to in Table 1.

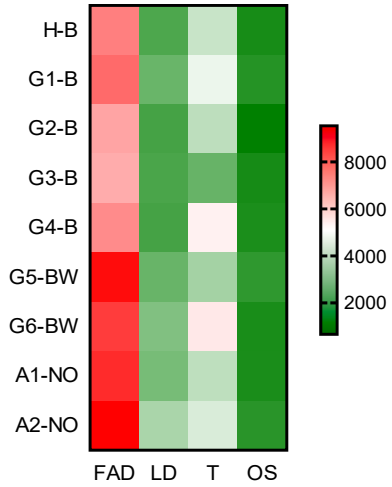


**Fig. 4.** Heat map representation of the absolute amount of fatty acid-derived odorants (FAD) in comparison to lignin degradation products (LD), terpenes (T), and other substance of deviating origins (OS).

**Table 5**
Concentration of the ten aroma-active compounds in oak samples. The values represent the mean values from determinations in duplicate.

| Odorants | Concentration in µg/kg[a] | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | H-B | A1-NO | A2-NO | G1-B | G2-B | G3-B | G4-B | G5-BW | G6-BW |
| Hexanal | 1255 | 924 | 340 | 936 | 359 | 251 | 255 | 375 | 272 |
| *α*-Pinene | 142 | 59 | 60 | 59 | 69 | 110 | 119 | 59 | 123 |
| Nonanal | 95 | 100 | 89 | 82 | 87 | 83 | 215 | 90 | 99 |
| (*Z*)-3-Hexenal | 701 | 40 | 86 | 36 | 56 | 67 | 50 | 70 | 58 |
| (*E*)-2-Nonenal | 553 | 304 | 503 | 1517 | 1843 | 2112 | 878 | 479 | 162 |
| Eugenol | 212 | 144 | 150 | 183 | 173 | 778 | 951 | 1144 | 1210 |
| Acetic acid | 255,800 | 227,000 | 260,000 | 258,500 | 299,000 | 258,000 | 110,500 | 162,000 | 152,000 |
| Vanillin | 5179 | 5408 | 5088 | 4462 | 4420 | 4406 | 4026 | 5972 | 6714 |
| Decanoic acid | 172 | 229 | 122 | 286 | 292 | 335 | 274 | 278 | 236 |
| *trans*-Whiskey lactone | 119 | 143 | 146 | 569 | 132 | 182 | 492 | 960 | 2100 |
| *cis*-Whiskey lactone | 51 | 61 | 72 | 326 | 366 | 184 | 1041 | 265 | 1589 |

[a] Mean values of duplicate, with relative standard deviation < 10%.

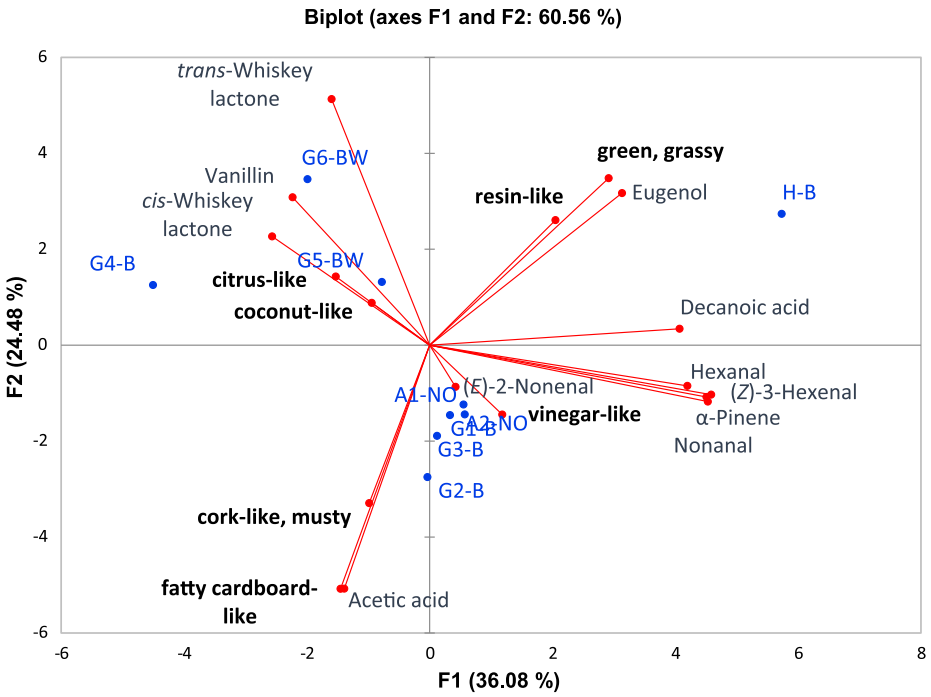


**Fig. 5.** PCA bi-plot of odorants showing two principal components that explain 60.56% of the variation. The light blue color represents oak samples, the black color shows sensory attributes and the dark blue color represents the quantified chemical compounds variables. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

compounds stem from fatty-acid degradation which is promoted by natural exposure of oak to air, relative humidity and sun, resulting in the formation of alkylic acids, alkenals, ketones, and lactones. The odor impressions of these fatty acid degradation products range from cheesy (pentanoic acid, propanoic acid, butanoic acid), green, grassy (pentanal, hexanal, (*Z*)-3-hexenal), citrus-like (octanal, nonanal, linalool) to fatty cardboard-like ((*E*)-2-nonenal), and coconut-like ($\gamma$-octalactone, $\gamma$-nonalactone, whiskey lactone). The pronounced vinegar-like smell of the oak samples is, accordingly, likely of being related to the fatty acid fraction, being dominated by large quantities of acetic acid. The results of the heat map data visualization are in-line with the quantification of compounds and sensory evaluation data (Fig. 4). This further explains the dominancy of fatty and cardboard-like smell impressions that are likely to relate to characteristic fatty acid degradation products such as (*E*)-2-nonenal, octanal and (*E*)-2-decenal with high FD factor. One might assume that the overall low hedonic rating in comparison to cembran pinewood is caused by the dominance of these substances, so that the overall smell of this type of wood is prated as less pleasant (Ghadiriasli, Mahmoud, Wagenstaller, van de Kuilen, & Buettner, 2020). Thereby, the oaks from Baden-Württemberg showed significantly higher hedonic values, and accordingly higher smell appreciation, compared to the Hungarian, and also the Bavarian and Austrian oak samples investigated in this study. Thus, there might be a relation between hedonic rates and geographical origin, or rather exposure to oxidation-promoting conditions during wood production (Fig. 1 and File 2 in supplementary material).

Apart from the fatty acid degradation products, a series of odorous compounds with terpenoic structure was identified in the oak samples. Representative of this group of terpenes are the monoterpenes $\alpha$-pinene (resin-like), $\beta$-myrcene (earthy, green-grassy), $\delta$-3-carene (citrus-like, eucalyptus-like), and linalool (flowery, citrus-like), as well as the oxygenated monoterpenes $\alpha$-thujone (minty-menthol-like), isopulegol (peppermint, fresh), estragole (anise-like), and geraniol (flowery, fresh). Several sesquiterpenes such as (*E*)-$\beta$-farnesene (fruity, cucumber-like, and woody), $\alpha$-humulene (resin-like, woody), rotundone (pepper-like), and mustakone (woody) were also found to be present. Together with several phenyl compounds, stemming from degradation of lignin in the cell walls, they form a noticeable group of odorants with wood-related smell impressions. Odor-active phenyl derivatives identified in the oak samples were guaiacol (smoky, vanilla-like), cinnamaldehyde (cinnamon-like), phenylacetic acid (honey-like, flowery), vanillin (vanilla-like), and syringaldehyde (green, woody). The pencil-like smelling thymoquinone and pencil-like, thyme-like smelling thymol are newly reported here as smell constituents in oak wood. Thymoquinone has previously been reported by our group as odorant in cedar wood and cembran pinewood (Ghadiriasli, Mahmoud, Wagenstaller, van de Kuilen, & Buettner, 2020; Schreiner, Loos, & Buettner, 2017).

Three substances had a sweaty, androstenone-like smell, but their chemical structures could not be resolved. Generally, sterols are known compounds in the extractable fractions of all wood types, therefore the formation of androstenone-like compounds with a steroid-related structure might be possible. Apart from that, a cucumber-like smelling substance with retention index 1480 on DB-FFAP also remained unknown as the obtained chromatographic and mass spectrometric data did not provide sufficient information for unequivocal identification of this trace compound.

### 5.2. *Comparison of the results obtained by sensory evaluations, cAEDA and SIDA*

Overall, the odor attributes reported during sensory evaluation matched very well with the main odorants, their respective odor qualities, and also the corresponding concentrations of the potent aroma-active compounds as determined by SIDA. Based on the results from the sensory evaluation, the odor impression vinegar-like, which is characteristic for acetic acid, was rated as the most intense in the G2-B (7.5), G1-B (7.0), G3-B (7.0) and A2-NO (7.0), which is in line with the elevated concentrations of acetic acid in these samples (299 mg/kg (G2-B), 258.5 mg/kg (G1-B), 258 mg/kg (G3-B) and 260 mg/kg (A2-NO), respectively). In agreement with that, the G4-B oak sample with the least intense vinegar-like smell was found to contain acetic acid with a concentration of 110.5 mg/kg only. According to cAEDA, $\alpha$-pinene (resin-

like smell) was further found to be a potent odorant in the H-B oak sample, reaching the highest FD factor there, followed by samples G6-BW, G3-B, and G4-B with lower FD factors. Again, this corresponded well with elevated levels of $\alpha$-pinene in these samples. Generally, variations in odorant concentration in the oak samples were quite pronounced in several cases; $\alpha$-pinene, for example, showed variations comprising a factor of three, ranging from 59 μg/kg in G1-B, G5-BW and A1-NO to 142 μg/kg value in H-B oak sample. Hexanal showed concentration differences of a factor of 5 between 251 μg/kg and 1255 μg/kg, and (*Z*)-3-hexenal even of a factor of about 20 (36–701 μg/kg). Such variations clearly serve as explanation for the observed sensory differences between the wood samples. Likewise, the fatty cardboard-like odor impression characteristic for (*E*)-2-nonenal, was perceived with a high intensity in samples G1-B (4.5), G2-B (5.0) and G3-B (5.5), corresponding with high concentrations of this characteristic odorant in these samples. The rancid odor impression, on the other hand, relates to the fatty acids as discussed above; cAEDA revealed, amongst others, high FD factors $\geq$ 128 for decanoic acid in all oak wood samples, but especially high values for G3-B, G2-B and G1-B corresponding to a higher rancid odor impression as well as higher concentrations of decanoic acid in these samples. The highest concentration for whiskey lactone was obtained for G6-BW (2100 μg/kg and 1589 μg/kg for trans and *cis*-whiskey lactone, respectively), being in line with the highest perceived intensity for the attribute coconut-like (8.0) during sensory evaluation.

We performed our analysis on natural oak wood. This wood is subjected to heat treatments like toasting or drying processes before being used for the aging and storage of wine and vinegar. Some of the attributes might be enhanced or suppressed by these treatments. In order to understand the chemical transformation of compounds during roasting, we need to analyze the composition of the natural wood first, and that was the aim of our study. However, potential further variations due to for instance, the forest stand, within tree variability, soil, climatic differences, different processing regimes and possible interrelationships between these would also need to be investigated in more detail. This knowledge may serve as a basis for generating optimized oak quality for different fields of application, be it as material for barrel making in view of usage for alcoholic beverage productions or for its use in aging vinegars.

According to hedonic scaling and PCA analysis the G6-BW sample was the most preferred oak wood from a sensory point of view among samples by the panel and this high hedonic might be attributed to the vanillin and whiskey lactone odorants and have good correlation with the concentration of these odorants in this oak sample.

With our work we aim at demonstrating that it is high time to take into consideration the prospects of new oak sources for aging and storage purposes in the field of food industry – and that we urgently not only need to think about sustainable alternatives but also about their potential impact on quality in production processes.

## 6. Conclusions

The qualitative and quantitative odorant composition in oak (*Quercus robur L.)* from different origins has been investigated and it has been revealed relevant differences between samples from different geographical origin on the overall odor profile and the corresponding concentrations of characterising odorants. In total 95 aroma-active compounds were detected in all samples. By applying GC-O analysis as well as one-dimensional and two-dimensional GC–MS/O analyses, 91 of these odorants could be identified. The chemical structure as well as the odor of the compounds were found to be very different, indicating diverse formation pathways and sources of these compounds. Thereby, most of the aroma-active compounds have already been reported in our former study on Hungarian oak (*Quercus frainetto*), whereas 2-propenoic acid (geranium-like, vinegar-like) and cinnamaldehyde (cinnamon-like) are reported here for the first time as smell constituents of natural oak samples. We could further show that the smell of oak is generally dominated by a series of fatty acid degradation products, being accompanied by several mono- and sesquiterpene constituents, some of them as oxygenated derivatives, and a number of lignin degradation products. All in all, the most potent aroma-active compounds in all the samples were green-grassy and citrus-like alkenals and the mushroom-like 1-octen-3-one, as well as vinegar-like and rancid smelling alkylic acids, resin-like terpenes, and clove-like, cinnamon-like, vanilla-like phenolic compounds. The vinegar-like smell was one of the lead impressions of the general smell profile of these samples, whereas resin-like, green-grassy, fatty cardboard-like, cork-like, musty, and coconut-like odor impressions differentiated the individual smell character between samples. Our study shows that, generally, most of the odorants were found in all samples, even from different geographical origin, but that quantitative differences were pronounced. With regard to this study and in relation to our previous findings on Cembran pinewood (Ghadiriasli, Mahmoud, Wagenstaller, van de Kuilen, & Buettner, 2020), it became evident that geographical vicinity had significant influence on the quantity and quality of wood volatile compounds. We propose that such quantitative deviations may be used in the future to relate samples to the respective growth area. Moreover, future studies now need to reveal how such variations potentially translate into treated wood quality and their products.

## CRediT authorship contribution statement

**Rahil Ghadiriasli:** Conceptualization, Methodology, Investigation, Visualization, Formal analysis, Validation, Writing – original draft. **Mohamed A.A. Mahmoud:** Formal analysis, Methodology, Investigation, Visualization, Validation, Writing – review & editing. **Maria Wagenstaller:** Validation, Writing – review & editing. **Jan-Willem van de Kuilen:** Resources, Writing – review & editing. **Andrea Buettner:** Supervision, Conceptualization, Writing – review & editing.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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*Ethical statement*

The study was conducted in agreement with the Declaration of Helsinki and in agreement with work safety protocols. The study was approved by the Ethical Committee of the Medical Faculty, Friedrich-Alexander Universität Erlangen-Nürnberg. Informed consent was obtained from all subjects participating in the study.

## Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodres.2021.110776.

## References


Anjos, O., Carmona, C., Caldeira, I., & Canas, S. (2013). Variation of Extractable Compounds and Lignin Contents in Wood Fragments Used in the Aging of Wine Brandies. *Bioresources, 8*(3), 4484–4496.

Bemelmans, J. M. H. (1979). Review of Isolation and Concentration Techniques. In *Progress in flavour research* (pp. 79–98). Applied Science.

Buettner, A., & Schieberle, P. (2001a). Application of a comparative aroma extract dilution analysis to monitor changes in orange juice aroma compounds during processing. *American Chemical SocietyPublications, 782*, 33–45.

Buettner, A., & Schieberle, P. (2001b). Evaluation of Aroma Differences between Hand-Squeezed Juices from Valencia Late and Navel Oranges by Quantitation of Key Odorants and Flavor Reconstitution Experiments. *Journal of Agricultural and Food Chemistry, 49*(5), 2387–2394.

Buettner, A., & Schieberle, P. (2001c). Evaluation of Key Aroma Compounds in Hand-Squeezed Grapefruit Juice (Citrus paradisi Macfayden) by Quantitation and Flavor Reconstitution Experiments. *Journal of Agricultural and Food Chemistry, 49*(3), 1358–1363.

Buttery, R. G., Ling, L. C., & Stern, D. J. (1997). Studies on Popcorn Aroma and Flavor Volatiles. *Journal of Agricultural and Food Chemistry, 45*(3), 837–843.

Cadahía, E., Varea, S., Muñoz, L., Fernández de Simón, B., & García-Vallejo, M. C. (2001). Evolution of Ellagitannins in Spanish, French, and American Oak Woods during Natural Seasoning and Toasting. *Journal of Agricultural and Food Chemistry, 49*(8), 3677–3684.

Christlbauer, M., & Schieberle, P. (2011). Evaluation of the key aroma compounds in beef and pork vegetable gravies a la chef by stable isotope dilution assays and aroma recombination experiments. *Journal of Agricultural and Food Chemistry, 59*(24), 13122–13130.

Culleré, L., Fernández de Simón, B., Cadahía, E., Ferreira, V., Hernández-Orte, P., & Cacho, J. (2013). Characterization by gas chromatography–olfactometry of the most odor-active compounds in extracts prepared from acacia, chestnut, cherry, ash and oak woods. *LWT - Food Science and Technology, 53*(1), 240–248.

Cutzach, I., Chatonnet, P., Henry, R., & Dubourdieu, D. (1997). Identification of Volatile Compounds with a "Toasty" Aroma in Heated Oak Used in Barrelmaking. *Journal of Agricultural and Food Chemistry, 45*(6), 2217–2224.

Czerny, M., Christlbauer, M., Christlbauer, M., Fischer, A., Granvogl, M., Hammer, M., … Schieberle, P. (2008). Re-investigation on odour thresholds of key food aroma compounds and development of an aroma language based on odour qualities of defined aqueous odorant solutions. *European Food Research and Technology, 228*(2), 265–273.

Czerny, M., & Grosch, W. (2000). Potent Odorants of Raw Arabica Coffee. Their Changes during Roasting. *Journal of Agricultural and Food Chemistry, 48*(3), 868–872.

Díaz-Maroto, M. C., Guchu, E., Castro-Vázquez, L., de Torres, C., & Pérez-Coello, M. S. (2008). Aroma-active compounds of American, French, Hungarian and Russian oak woods, studied by GC–MS and GC–O. *Flavour and Fragrance Journal, 23*(2), 93–98.

Doussot, F., De Jéso, B., Quideau, S., & Pardon, P. (2002). Extractives content in cooperage oak wood during natural seasoning and toasting; influence of tree species, geographic location, and single-tree effects. *Journal of Agricultural and Food Chemistry, 50*(21), 5955–5961.

Engel, W., Bahr, W., & Schieberle, P. (1999). Solvent assisted flavour evaporation–a new and versatile technique for the careful and direct isolation of aroma compounds from complex food matrices. *European Food Research Technology, 209*(3-4), 237–241.

Fernández de Simón, B., Cadahía, E., Conde, E., & García-Vallejo, M. C. (1996). Low Molecular Weight Phenolic Compounds in Spanish Oak Woods. *Journal of Agricultural and Food Chemistry, 44*(6), 1507–1511.

Fernández de Simón, B., Cadahía, E., & Jalocha, J. (2003). Volatile Compounds in a Spanish Red Wine Aged in Barrels Made of Spanish, French, and American Oak Wood. *Journal of Agricultural and Food Chemistry, 51*(26), 7671–7678.

Fernández de Simón, B., Hernández, T., Cadahía, E., Dueñas, M., & Estrella, I. (2003). Phenolic compounds in a Spanish red wine aged in barrels made of Spanish, French and American oak wood. *European Food Research and Technology, 216*(2), 150–156.

Ghadiriasli, R., Mahmoud, M. A. A., Wagenstaller, M., van de Kuilen, J.-W., & Buettner, A. (2020). Molecular and sensory characterization of odorants in Cembran pine (Pinus cembra L.) from different geographic regions. *Talanta, 220*, Article 121380.

Ghadiriasli, R., Wagenstaller, M., & Buettner, A. (2018). Identification of odorous compounds in oak wood using odor extract dilution analysis and two-dimensional gas chromatography-mass spectrometry/olfactometry. *Analytical and Bioanalytical Chemistry, 410*(25), 6595–6607.

Glabasnia, A., & Hofmann, T. (2006). Sensory-directed identification of taste-active ellagitannins in American (Quercus alba L.) and European oak wood (Quercus robur L.) and quantitative analysis in bourbon whiskey and oak-matured red wines. *Journal of Agricultural and Food Chemistry, 54*(9), 3380–3390.

Grosch, W. (2001). Evaluation of the key odorants of foods by dilution experiments, aroma models and omission. *Chemical Senses, 26*(5), 533–545.

Guth, H., & Grosch, W. (1990). Deterioration of soya-bean oil: Quantification of primary flavour compounds using a stable isotope dilution assay. *Lebensmittel-Wissenschaft & Technologie, 23*(6), 513–522.

Jordão, A. M., Ricardo-da-Silva, J. M., Laureano, O., Adams, A.n., Demyttenaere, J., Verhé, R., & De Kimpe, N. (2006). Volatile composition analysis by solid-phase microextraction applied to oak wood used in cooperage (Quercus pyrenaica and Quercus petraea): Effect of botanical species and toasting process. *Journal of Wood Science, 52*(6), 514–521.

Li, S., Crump, A. M., Grbin, P. R., Cozzolino, D., Warren, P., Hayasaka, Y., & Wilkinson, K. L. (2015). Aroma Potential of Oak Battens Prepared from Decommissioned Oak Barrels. *Journal of Agricultural and Food Chemistry, 63*(13), 3419–3425.

Lopez Pinar, A., Rauhut, D., Ruehl, E., & Buettner, A. (2017). Quantification of the Changes in Potent Wine Odorants as Induced by Bunch Rot (Botrytis cinerea) and Powdery Mildew (Erysiphe necator). *Frontiers in Chemistry, 5*, 57.

Pérez-Coello, M. S., Sánchez, M. A., García, E., González-Viñas, M. A., Sanz, J., & Cabezudo, M. D. (2000). Fermentation of White Wines in the Presence of Wood Chips of American and French Oak. *Journal of Agricultural and Food Chemistry, 48*(3), 885–889.

Marco, J., Artajona, J., Larrechi, M. S., & Rius, F. X. (1994). Relationship Between Geographical Origin and Chemical Composition of Wood for Oak Barrels. *American Journal of Enology and Viticulture, 45*(2), 192.

Milo, C., & Grosch, W. (1993). Changes in the odorants of boiled trout (Salmo fario) as affected by the storage of the raw material. *Journal of Agricultural and Food Chemistry, 41*(11), 2076–2081.

Morales, M. L., Benitez, B., & Troncoso, A. M. (2004). Accelerated aging of wine vinegars with oak chips: Evaluation of wood flavour compounds. *Food Chemistry, 88*(2), 305–315.

Morales, M. L., Tesfaye, W., García-Parrilla, M. C., Casas, J. A., & Troncoso, A. M. (2002). Evolution of the aroma profile of sherry wine vinegars during an experimental aging in wood. *Journal of Agricultural and Food Chemistry, 50*(11), 3173–3178.

Mosedale, J. R., Feuillat, F., Baumes, R., Dupouey, J.-L., & Puech, J.-L. (1998). Variability of wood extractives among *Quercus robur* and *Quercus petraea* trees from mixed stands and their relation to wood anatomy and leaf morphology. *Canadian Journal of Forest Research, 28*(7), 994–1006.

Nonier, M.-F., De Gaulejac, N. V., Vivas, N., & Vitry, C. (2004). Characterization of carotenoids and their degradation products in oak wood. Incidence on the flavour of wood. *Comptes Rendus Chimie, 7*(6-7), 689–698.

Nonier, M. F., Vivas, N., Vivas de Gaulejac, N., Absalon, C., Soulié, P.h., & Fouquet, E. (2006). Pyrolysis–gas chromatography/mass spectrometry of Quercus sp. wood: Application to structural elucidation of macromolecules and aromatic profiles of different species. *Journal of Analytical and Applied Pyrolysis, 75*(2), 181–193.

Pérez-Coello, M. S., González-Viñas, M. A., García-Romero, E., Cabezudo, M. D., & Sanz, J. (2000). Chemical and sensory changes in white wines fermented in the presence of oak chips. *International Journal of Food Science & Technology, 35*(1), 23–32.

Pérez-Coello, M. S., Sanz, J., & Cabezudo, M. D. (1998). Gas chromatographic-mass spectrometric analysis of volatile compounds in oak wood used for ageing of wines and spirits. *Chromatographia, 47*(7-8), 427–432.

Pino, J. A., & Mesa, J. (2006). Contribution of volatile compounds to mango (Mangifera indica L.) aroma. *Flavour and Fragrance Journal, 21*(2), 207–213.

Prida, A., & Puech, J.-L. (2006). Influence of Geographical Origin and Botanical Species on the Content of Extractives in American, French, and East European Oak Woods. *Journal of Agricultural and Food Chemistry, 54*(21), 8115–8126.

Ríos-Reina, R., Segura-Borrego, M. P., García-González, D. L., Morales, M. L., & Callejón, R. M. (2019). A comparative study of the volatile profile of wine vinegars with protected designation of origin by headspace stir bar sorptive extraction. *Food Research International, 123*, 298–310.

Savill, J. R. M. P. S. (1996). Variation of heartwood phenolics and oak lactones between the species and phenological types of Quercus petraea and Q. robur. *Forestry, 69*(1), 47–55.

Schieberle, P., & Grosch, W. (1987). Quantitative analysis of aroma compounds in wheat and rye bread crusts using a stable isotope dilution assay. *Journal of Agricultural and Food Chemistry, 35*(2), 252–257.

Schreiner, L., Loos, H. M., & Buettner, A. (2017). Identification of odorants in wood of Calocedrus decurrens (Torr.) Florin by aroma extract dilution analysis and two-dimensional gas chromatography-mass spectrometry/olfactometry. *Analytical and Bioanalytical Chemistry, 409*(15), 3719–3729.

Schuh, C., & Schieberle, P. (2006). Characterization of the Key Aroma Compounds in the Beverage Prepared from Darjeeling Black Tea: Quantitative Differences between Tea Leaves and Infusion. *Journal of Agricultural and Food Chemistry, 54*(3), 916–924.

Sefton, M. A., Francis, I. L., & Williams, P. J. (1990). Volatile norisoprenoid compounds as constituents of oak woods used in wine and spirit maturation. *Journal of Agricultural and Food Chemistry, 38*(11), 2045–2049.

Shanbhag, R. R., & Sundararaj, R. (2013). Physical and chemical properties of some imported woods and their degradation by termites. *Journal of insect science (Online), 13*(63), 1–8.

Torija, M. J., Mateo, E., Vegas, C. A., Jara, C., González, A., Poblet, M., …, Mas, A. (2009). Effect of wood type and thickness on acetification kinetics in traditional vinegar production. *International Journal of Wine Research., 1*, 155–160.

Ubeda, C., Callejón, R. M., Hidalgo, C., Torija, M. J., Mas, A., Troncoso, A. M., & Morales, M. L. (2011). Determination of major volatile compounds during the production of fruit vinegars by static headspace gas chromatography–mass spectrometry method. *Food Research International, 44*(1), 259–268.

van Den Dool, H., & Dec. Kratz, P. (1963). A generalization of the retention index system including linear temperature programmed gas—liquid partition chromatography. *Journal of Chromatography A, 11*, 463–471.