Research Article

Volatile-Olfactory Profiles of *cv*. Arbequina Olive Oils Extracted without/with Olive Leaves Addition and Their Discrimination Using an Electronic Nose

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Oils from *cv*. Arbequina were industrially extracted together with olive leaves of *cv*. Arbequina or Santulhana (1%, w/w), and their olfactory and volatile profiles were compared to those extracted without leaves addition (control). The leaves incorporation resulted in green fruity oils with fresh herbs and cabbage olfactory notes, while control oils showed a ripe fruity sensation with banana, apple, and dry hay grass notes. In all oils, total volatile contents varied from 57.5 to 65.5 mg/kg (internal standard equivalents), being aldehydes followed by esters, hydrocarbons, and alcohols the most abundant classes. No differences in the number of volatiles were observed. The incorporation of *cv*. Arbequina or Santulhana leaves significantly reduced the total content of alcohols and esters (minus 37–56% and 10–13%, respectively). Contrary, *cv*. Arbequina leaves did not influence the total content of aldehydes or hydrocarbons, while *cv*. Santulhana leaves promoted a significant increase (plus 49 and 10%, respectively). Thus, a leaf-cultivar dependency was observed, tentatively attributed to enzymatic differences related to the lipoxygenase pathway. Olfactory or volatile profiles allowed the successful unsupervised differentiation of the three types of studied *cv*. Arbequina oils. Finally, a lab-made electronic nose was applied to allow the nondestructive discrimination of *cv*. Arbequina oils extracted with or without the incorporation of olive leaves (100% and 99 ± 5% of correct classifications for leave-one-out and repeated *K*-fold cross-validation variants), being a practical tool for ensuring the label correctness if future commercialization is envisaged. Moreover, this finding also strengthened that olive oils extracted with or without olive leaves incorporation possessed quite different olfactory patterns, which also depended on the cultivar of the olive leaves.

1. Introduction

Virgin olive oils (VOO) are worldwide valued by consumers due to the known health benefits [1] as well as due to their delicate sensory attributes [2]. VOOs' physicochemical and sensory (olfactory (direct), gustatory, and retro-olfactory (indirect) sensations) characteristics are influenced by several factors that include olive cultivar, olive maturity index at harvest, agroclimatic conditions, extraction scale, and malaxation conditions [3]. Volatile compounds from different chemical classes have been identified in VOO, playing, both major and minor compounds, a key role in the final VOO quality [4]. The most important volatiles comprise C5 and C6 compounds, namely, aldehydes, alcohols, esters, and hydrocarbons, which are formed following the oxidation of the free polyunsaturated fatty acids, via the lipoxygenase

(LOX) pathway [3]. At the same time, sugar fermentation, metabolism of some amino acids, or oxidation may occur, leading to the appearance of other volatile compounds that, if in excess, can originate off-flavors [3].

Several strategies have been reported to enhance the physicochemical-sensory quality of VOOs, their positive health-related properties, and shelf-life, including the incorporation of olive leaves during the oils' extraction [5–9]. Some of these studies reported the impact of incorporating dry or fresh leaves (1–10%, w/w) at laboratory or pilot scale extractions, on the volatile and sensory profiles of olive oils. The results showed that in general, the volatile fraction and sensory attributes of olive oils extracted with leaves addition, depended on the olive and leaf cultivars as well as on the amount of leaves added in the course of the oils' extraction.

The volatile profiles and olfactory fingerprint of olive oils have been established using either chromatographic based techniques (e.g., gas chromatography coupled to mass spectrometry, GC-MS, and two-dimensional gas chromatography) or by official/trained sensory panels, allowing, among others, the oils classification based on defects evaluation, detection of adulterations, oils' differentiation according to the geographical origin [10–13]. However, chromatographic techniques are laborious and expensive and involve skilled technicians [14]. On the other hand, despite the important role played by the trained panelists regarding the commercial-grade classification of VOO, providing a qualitative and quantitative robust analysis, supported by adequate statistical tools; sensory panels have some limitations. The training process is complex, time-consuming, and expensive, being the evaluations hindered by several factors, namely, the intrinsic subjectivity of the human assessment, the human fatigue, and the low number of samples that can be daily evaluated [2, 15]. Moreover, sensory analysis is usually expensive for small-/medium-sized companies [16]. However, the sensory analysis of VOO is legally required for commercialization namely, to establish the quality grade of the oils [17].

Therefore, it is important to develop feasible techniques for real-time olive oil analysis to ensure the labeling correctness, allowing, for example, identifying oils extracted with or without olive leaves incorporation [18]. Among these alternative analytical tools, electronic noses (E-nose) have emerged as noninvasive, user-friendly, and green devices for possible *in situ* olive oil analysis. An E-nose is a gas sensor device that aims to mimic the human olfactory perception of volatile chemical compounds. It allows establishing an artificial volatile fingerprint of a particular sample, using the electric signals gathered by a set of sensors (multiple sensors) together with qualitative and/or quantitative chemometric tools [19]. Both lab-made and commercial apparatus have been used for oils' adulteration detection [20] to identify specific aroma markers in oils extracted from olives with anthracnose [21], for assessing oils' quality grade and oils' blends discrimination [22, 23], or to establish aroma fingerprints of extra virgin olive oils (EVOO) [24]. More recently, Teixeira et al. [25] and Cano Marchal et al. [26] have used E-noses to classify VOOs according to their fruitiness intensity or to detect sensory defects. Nevertheless, it should

be pointed out that the electrical signals of the gas sensors, comprised in an E-nose device, highly depend on the capacity of controlling the sampling/analysis conditions, namely, temperature, moisture, pressure, gas speed, and vapor phase concentrations.

In this sense, it was intended to evaluate, for the first time, the possibility of applying a lab-made E-nose with metal oxide semiconductor (MOS) sensors to identify *cv*. Arbequina olive oils extracted, or not, with the addition of *cv*. Arbequina or Santulhana fresh olive leaves (1%, w/w), at an industrial scale. Indeed, the literature mainly focus on the impact of leaves incorporation during the oil extraction in laboratory or pilot scale facilities, which processes cannot mimic the composition of VOOs extracted at an industrial (commercial) scale [27]. In fact, to the authors' best knowledge, only one study, dated back to 1996, also evaluated oils extracted after leaves incorporation at an industrial scale [8]. So, recently, Marx et al. [9] studied the impact of incorporating 1% (w/w) of fresh olive leaves from *cv*. Arbequina or Santulhana on the physicochemical, phenolic, and gustatory characteristics of *cv*. Arbequina oils extracted industrially. Moreover, in the same study, an electronic tongue (E-tongue) was used as an alternative and fast analytical tool, allowing discriminating the three types of oils (i.e., extracted after the addition or not of olive oils) [9]. Despite the successful qualitative discrimination performance of the taste sensor device, the approach required a preliminary aqueous-methanolic extraction of the oils, and thus, it resulted in a destructive/invasive procedure. Thus, this study aimed to complement the previous work, by evaluating the volatile and olfactory characteristics of the extracted oils. This study also aimed to evaluate the possibility of using an E-nose as a nondestructive/noninvasive tool to discriminate olive oils extracted with or without the incorporation of olive leaves. Finally, *cv*. Arbequina was selected since although possessing known strong cultivation advantages compared to other olive cultivars, which are responsible for its worldwide implementation and commercial relevance [9], the extracted oils are of mild quality compared to those obtained from other cultivars [28]. Therefore, their overall quality could be enhanced by the olive tree leaves incorporation at the extraction process. The incorporation of leaves from *cv*. Santulhana was evaluated taking into account its prevalence in traditional olive groves of Trás-os-Montes' region (northeast Portugal) [9].

2. Materials and Methods

2.1. Olives, Olive Leaves, and Olive Oil Samples. As previously described [9], the *cv*. Arbequina olives were picked in mid-November 2019 from an olive grove located in the Trás-os-Montes region (northeast Portugal). Olives had a maturity index between two and three. Leaves from *cvs*. Arbequina and Santulhana were collected in the olive mill, being firstly separated manually from branches, and then washed with water. In total, 6 kg of cleaned leaves of each cultivar were added to 600 kg of *cv*. Arbequina olives (1%, w/w) before the milling process. The amount of olive leaves incorporated was fixed according to the lower levels reported in the literature

[5–8], aiming to mimic the possible undeliberated incorporation of olive leaves when oil is extracted at the industrial level. The oils were extracted (22°C, 45 min, 12 RPM) using a two-phase commercial industrial unit mill (Alfa Laval, Italy). A two-phase unit mill from Alfa Laval (Italy) was used, which comprised a hammer crusher (5,000 kg of olives per hour capacity); a sieve (11 mm of diameter) coupled with two malaxers (Type Gramula 700), each one with four bodies (capacity of 650 kg of paste); a two-phase horizontal decanter centrifuge (capacity of 5,000 kg of paste per hour); and a vertical centrifuge (UVPX 507 AGT14). The olive oils were extracted within the first 24 h after the harvest of the olives [9].

Three *cv*. Arbequina oils were obtained, namely, without leaves addition (control), with the addition of *cv*. Arbequina leaves (1%, w/w), and with the addition of *cv*. Santulhana leaves (1%, w/w). The high amount of olives required for performing each industrial extraction batch and the cost involved limited the number of independent extractions. Although this number could be increased for laboratory or pilot scale extractions, this option was not considered since it is reported that the composition of olive oils extracted at these scales can hardly mimic that obtained at the industrial level [27]. From each independent industrial extraction, 5 oil bottles (amber glass bottles) were filled, closed, and transported to the laboratory (Bragança, Portugal). Then, the oils' water traces were removed using anhydrous sodium sulfate (1 g for 100 mL of olive oil), being the oils subsequently filtered using cellulose filters. In total, 15 independent samples were obtained and stored in amber glass bottles (~100 mL), in the dark, at 18–22°C, for 6 months to promote the progress of the sensory sensations [29].

2.2. Volatile Characterization by HS-SPME-GC-MS. The characterization of the volatile fraction of the olive oils was performed by headspace solid-phase microextraction (HS-SPME) gas-chromatography-mass spectrometry (GC/MS). A divinylbenzene/carbonex/polydimethylsiloxane (DVB/CAR/PDMS, 50/30 μm) fiber (Supelco, Bellefonte, USA) was used. The assays were performed in a Shimadzu GC-2010 Plus chromatographer equipped with a mass spectrometer Shimadzu GC/MS-QP2010 SE detector, as previously described [5]. For analysis, 3 g of olive oil were placed in vials (50 mL) and spiked with 5 μL of the internal standard (4-methyl-2-pentanol, 98% from Sigma Aldrich) solution (0.127 mg/mL). The volatiles were allowed to adsorb into an SPME fiber coated with divinylbenzene/carbonex/polydimethylsiloxane fiber. The vials were kept at 50°C for 5 min in order to achieve an effective release of the volatile compounds. After, the SPME fiber was exposed for 30 min, at 50°C, allowing the adsorption of the volatile compounds present in the headspace, being the samples kept under agitation (350 RPM).

In total 30 chromatographic assays were made (3 *cv*. Arbequina olive oil types × 5 bottles × 2 extractions × 1 injection). Separation was accomplished on a TRB-5MS (30 m × 0.25 mm × 0.25 μm) column (Teknokroma, Spain). The injector temperature was 220°C, and the injections were made in splitless mode. Helium (Praxair, Portugal) was used as the mobile phase (linear speed of 30 cm/s and total flow of 24.4 mL/min). A temperature gradient of the oven was applied (40°C/1 min; 2°C/min until 220°C; and then 30 min at 220°C). The ionization source was kept at 250°C with ionization energy of 70 electronvolts and an ionization current of 0.1 kilovolts. The mass spectra were acquired by electron ionization, being the spectra fragments identified by comparison with the database of the NIST 11 Library (National Institute of Standards and Technology, Gaithersburg, MD, USA) and with the spectra of commercial standards. Reconstructed peaks were obtained from the full scan chromatogram using the ion base (m/z intensity, 100%) for each compound, being then the peaks' areas determined. The identified volatiles were semiquantified as the ratio of each base ion peak area to the area of the internal standard base ion peak area, without considering the response factors, and converted to mass equivalents based on the internal standard mass used.

2.3. Olive Oil Olfactory Analysis. A trained sensory panel, from the School of Agriculture of the Polytechnic Institute of Bragança, Portugal [30], composed of 8 tasters (3 men and 5 women, aged between 25 and 54 years), plus the panel leader, established the olfactory profile of the olive oils, following the methodologies described by the European Union standard methods (Annexes II and IX in the Commission Regulation (EEC) N° 2568/91 from 11 July and amendments) [17]. The intensities of the perceived attributes were scored using an unstructured continuous intensity scale, ranging from 0 (no sensory sensation perceived) to 10 (maximum perceived intensity). The descriptive profile was assessed using a test sheet, according to the International Olive Council (IOC) [31] and as previously described [30].

2.4. E-Nose Analysis

2.4.1. Lab-Made Device. The E-nose used in this study was designed and was built in the laboratory as previously described by Teixeira et al. [25]. The device comprised a heated sampling unit (~28°C) and a heated multisensor detection array (~35°C). The sample's headspace vapor phase was carried to the detection chamber by means of a diaphragm vacuum air pump (model SC3502PM, from SKOOCOM, China). For cleaning the system and sensors, nitrogen (UN 1066, Linde 089 cyl 02/15) was used as a constant flow until a stabilized baseline was attained. The E-nose device had nine commercial MOS sensors, which electrical signals changed due to the adsorption of the volatile compounds on the sensors' surface. According to the literature, each MOS sensor reacted with different target gases (Table 1). Furthermore, it was previously shown [25] that these nine MOS sensors can recognize and distinguish standard solutions of acetic acid, cis-3-hexenyl, cis-3-hexen-1-ol, hexanal, 1-hexenol, and nonanal, which can be related with both positive (e.g., fatty, floral, fruit, grass, green, and green leaves attributes) and negative (e.g., sour and vinegary defects) attributes that can be commonly detected in olive oils. The

TABLE 1: Metal oxide semiconductor (MS) sensors of the E-nose device.

Commercial designation	E-nose sensor code	Target gases
TGS 2600 B00	S1	General air contaminants
TGS 2602	S2	General air contaminants
TGS 2610 C00	S3	Butane, liquid petroleum gases
TGS 2611 C00	S4	Methane, natural gas
TGS 2610 D00	S5	Butane, liquid petroleum gases
TGS 2611 E00	S6	Methane, natural gas
TGS 2612	S7	Methane, propane, isobutane
TGS 826 A00	S8	Ammonia
TGS 823 C12N	S9	Organic solvent gases

electrical resistance (in ohms, Ω) was recorded using an Agilent data acquisition unit (model 34970A), controlled by the Agilent BenchLink Data Logger software.

2.4.2. Olive Oil Samples Conditioning and Analysis. As previously described [25], for each assay, 0.5 mL of each olive oil sample was pipetted into a glass vial (25 mL) and placed in the sampling chamber (28°C, as recommended by the IOC for olive oils sensory analysis), for 13 min. Simultaneously, the E-nose system was cleaned using nitrogen until a stable signal baseline was obtained. The gas headspace was then delivered to the detection chamber where it interacted with the MOS sensors for 2.5 min, being the resistance signals recorded at 4 sec intervals.

2.4.3. Data Acquisition, Feature Extraction, and Signal Treatment. The signals of each of the 9 MOS, acquired by the data logger, during each sample's analysis, resulted in 37–38 resistance values, which were recorded. Six feature extraction methods were applied to obtain a representative E-nose fingerprint of each olive oil sample's volatile fraction, as proposed by Gila et al. [32] and previously described [25]: last response point (LP), the integral of the response curve (INT), the maximum response point (MAX), the minimum response point (MIN), the sum of the response curve (SUM), and the mean of the response curve (MEAN).

2.5. Statistical Analysis. One-way ANOVA and Tukey's post hoc multicomparison test were used to analyze the volatile components as well the olfactory sensory attributes of the three different *cv*. Arbequina olive oils (i.e., industrially extracted without or with the addition of *cvs*. Arbequina or Santulhana leaves). When only two types of olive oils were compared, the Student's t-test was applied. Multivariate statistical tools were applied, namely, unsupervised (principal component analysis, PCA) and supervised (linear discriminant analysis, LDA) techniques, to assess the classification power of the lab-made E-nose MOS device. In the present work, the signals resulting from the six feature extraction methods applied were simultaneously used, totalizing 54 signals for each sample (9 MOS × 6 different feature extractions) [33]. LDA together with the simulated annealing (SA) algorithm was further used to select the best subsets of nonredundant signals (among the 54) that enabled the best classification performance according to the

leave-one-out cross-validation (LOO-CV) and the repeated *K*-fold CV procedures. The classification performances were discussed in terms of the sensitivity (i.e., the percentage of corrected classified samples) as well as through 2D and 3D plots. The Sub-select [34] and MASS [35] packages of the open-source statistical program *R* (RStudio version 1.2.5033) were used for the statistical analysis, at a 5% significance level.

3. Results and Discussion

3.1. Impact of Leaves Incorporation during the Extraction of Oils on the Olfactory and Volatile Profiles. The panelists did not perceive any sensory defect in the studied *cv*. Arbequina oils, but identified different positive olfactory sensations, namely, ripe, or green fruity, apple, tomato, dry fruits, banana, dry hay grass, fresh herbs, and cabbage, depending on if the oils were extracted without or with olive leaves incorporation (Table 2). The olfactory profiles established for the *cv*. Arbequina oils extracted without leaves addition (control oils) are in line with the literature data, although with a slightly greater ripe fruity intensity compared to the scores reported for *cv*. Arbequina oils by Rodrigues et al. [36] and Sánchez-Rodríguez et al. [37]. Overall, the results also showed that the addition of olive leaves had a significant effect on the number and intensity of the perceived olfactory sensations (*P* value <0.05, one-way ANOVA). The incorporation of fresh-green olive leaves of *cv*. Arbequina or Santulhana resulted in oils with a green fruity olfactory sensation, being significantly higher for the latter oils (green fruity intensities of 2.1 and 5.3 for oils extracted with leaves from *cv*. Arbequina or Santulhana, respectively). The observed shift from ripe to green fruity sensation due to the incorporation of olive leaves was also reported by Sonda et al. [7]. Finally, it should be noticed that olive leaves incorporation during oils extraction promoted the appearance of fresh herbs and cabbage olfactory notes and the disappearance of banana and dry hay grass notes, including the fact that the leaves increment did not present any sensory defect.

In which concerns the volatiles profile, 21 compounds from 7 chemical classes (acids, alcohols, aldehydes, esters, ethers, hydrocarbons, and terpenes) were detected in all studied *cv*. Arbequina oils (Table 3). For identification purposes, a minimum similarity percentage of 90% was used, meaning that only peaks with a similarity of at least 90% with those of the NIST 11 Library database were identified. The

Table 2: Olfactory attributes perceived and respective intensities (mean ± standard deviation; $n = 5$ olive oil bottles × 2 samples × 8 panelists) regarding the studied *cv*. Arbequina olive oils.

Olfactory attributes perceived by the sensory panel	Intensities perceived in industrially extracted *cv*. Arbequina olive oils[#]			P value[‡]
	Without leaves addition (control)	With *cv*. Arbequina leaves	With *cv*. Santulhana leaves	
Ripe fruity	6.1 ± 0.8	0.0 ± 0.0	0.0 ± 0.0	—
Green fruity	0.0 ± 0.0	2.1 ± 0.3	5.3 ± 0.5	<0.0001
Apple	5.0 ± 0.5[A]	4.3 ± 0.4[B]	3.9 ± 0.3[B]	<0.0001
Tomato	4.1 ± 0.6[B]	3.5 ± 0.3[C]	5.3 ± 0.2[A]	<0.0001
Dry fruits	2.6 ± 0.4[A]	1.7 ± 0.2[B]	2.5 ± 0.1[A]	<0.0001
Banana	1.3 ± 0.23	0.0 ± 0.0	0.0 ± 0.0	—
Dry hay grass	3.4 ± 0.7	0.0 ± 0.0	0.0 ± 0.0	—
Fresh herbs	0.0 ± 0.0	2.0 ± 0.2	2.7 ± 0.4	0.0006
Cabbage	0.0 ± 0.0	3.5 ± 0.4	3.8 ± 0.3	0.1816
Harmony	8.4 ± 0.2[A]	8.3 ± 0.4[A]	6.4 ± 0.2[B]	<0.0001

#Intensity means ($n = 30$) in the same line with the same uppercase letter are not significantly different from a statistical point of view according to the test of Tukey, at a significance level of 0.05, following the IOC regulations [31]. Intensity scale: from 0 (absence of attribute: not perceived by the panelists) to 10 (maximum attribute intensity). ‡P values for the one-way ANOVA (comparison 3 groups) or Student's t-test (comparison among the mean values of only 2 groups).

relative area of the identified peaks was 75–79% of the total area of detected peaks. Although the olive leaves incorporation had a significant effect on the volatiles' contents (P value <0.05, one-way ANOVA), being the volatiles individual contents leaf-cultivar dependent, the most abundant chemical classes were aldehydes followed by esters, alcohols, and hydrocarbons, which is in agreement with the literature data [39, 40]. For the control oils, the established volatile profile was similar to that reported in the literature for *cv*. Arbequina oils [37, 38]. On the other hand, it should be noticed that a wide range of contents has been reported for the same volatiles or chemical class, which could be attributed to the agroclimatic conditions, olive ripening index at harvest, and extraction methods used [3].

The C6 aldehydes (e.g., hexanal, (Z)-3-hexenal, and (E)-2-hexenal) and alcohols (hexanol, (Z)-3-hexenol, and (E)-2-hexenol), and respective acetyl esters (hexylacetate and (Z)-3-hexen-1-yl acetate) are the most significant aroma compounds of EVOO [41], accounting, usually, for 60 to 80% of the total volatile fraction [42]. These compounds are responsible for the green notes usually perceived in the VOOs and are formed due to a cascade of biochemical reactions (LOX pathway), due to the action of enzymes that transform polyunsaturated fatty acids into aldehydes, which are then reduced to alcohols and esterified to produce esters [43]. This latter group is also quite relevant for the sensory quality of EVOO [43].

In this study, the results showed that the oils (extracted without or with leaves incorporation) had a high content of C6 aliphatic compounds (namely, (E)-2-hexenal, 1-hexanol, (Z)-3-hexen-1-ol, (E)-2-hexen-1-ol, (Z)-2-hexen-1-yl acetate, and (Z)-3-hexen-1-yl acetate), representing more than 80% of the total volatile contents, which are formed from linoleic and linolenic acids via the LOX pathway [3, 44]. Among them, (E)-2-hexenal is the most abundant aldehyde in all oils, which is in line with the literature for varietal oils from different geographical origins (Portugal, Tunisia, and Morocco) [39, 45, 46]. Besides, its content was leaf-cultivar-dependent, since compared to the control oils, the

incorporation of *cv*. Arbequina leaves promoted a slight decrease (minus 8%) in agreement with Sanmartin et al. [6] study, while *cv*. Santulhana leaves endorsed an increase (plus 10%). This increasing trend due to the olive leaves incorporation was previously reported in the literature [5, 7, 8], depending on the increased level and final content on olive/leaf cultivar as well as on the level of incorporated leaves. Regarding (Z)-3-hexen-1-yl acetate content, the most abundant detected ester, its content slightly decreased with the incorporation of leaves, especially of *cv*. Santulhana leaves, in agreement with the findings of Malheiro et al. [5]. Finally, the contents of the oils' volatile alcohols were significantly influenced by the leaves' incorporation. A significant decrease in some C6 alcohols, like (Z)-3-hexen-1-ol, (Z)-2-hexen-1-ol. and 1-hexanol, was observed (Table 3). Also, it was found that total amounts of C6 aldehydes are greater than that of C6 alcohols in *cv*. Arbequina oils (Table 3), which can be attributed to the enzymatic action through the LOX pathway, which is in line with the findings of Malheiro et al. [5] but contrary to those reported by Sonda et al. [7].

Volatile compounds are related to the oils' different sensory sensations perceived, being, for example, C5 and C6 compounds called "green volatiles" [3]. Specifically, C6-aldehydes are among the main contributors to the green, fruity, and sweet sensations of VOOs [47]. These relationships may be used to tentatively justify the higher green fruity intensity of oils extracted with *cv*. Santulhana leaves, which also presented a greater content of (E)-2-hexenal (C6-aldehyde) compared to the control oils. In fact, for the studied oils, a linear correlation could be established between the C6- alcohols volatile compounds (namely, 1-hexanol, (Z)-3-hexen-1-ol, and (Z)-2-hexen-1-ol) and the literature [40, 48] aroma sensory descriptors, like apple and banana sensations ($0.818 \leq R$-Pearson ≤ 0.986). Regarding the hydrocarbons, three pentene dimer isomers (I, II, and III; Table 3) were identified, which are related to the LOX pathway, through the dimerization of 1,3-pentadiene [3]. Taking into account the significant increase of these three isomers in the oils extracted with *cv*. Santulhana

TABLE 3: Identified volatile compounds and respective contents (mean ± standard deviation; mg of compound/kg of olive oil (as internal standard equivalents); for each oil, $n = 5$ olive oil bottles × 2 analytical extractions × 1 GC-MS analysis) of the studied *cv.* Arbequina olive oils.

| Volatile compounds[#] | Industrially extracted *cv.* Arbequina oils | | | Aroma sensory descriptor[‡] | P value[§] |
	Without leaves addition (control)	With *cv.* Arbequina leaves	With *cv.* Santulhana leaves		
Acids					
Acetic acid	1.0 ± 0.12^{B}	1.0 ± 0.07^{B}	1.9 ± 0.32^{A}	Sour, vinegary	<0.0001
Alcohols					
(Z)-3-hexen-1-ol	0.9 ± 0.2^{A}	0.7 ± 0.2^{A}	0.3 ± 0.1^{B}	Apple, banana	<0.0001
(Z)-2-hexen-1-ol	0.6 ± 0.1^{A}	0.3 ± 0.04^{B}	0.2 ± 0.03^{C}	Green, apple, flowers, fruity, grass	<0.0001
1-hexanol	3.7 ± 0.3^{A}	2.3 ± 0.1^{B}	1.8 ± 0.1^{C}	Fruit, banana, soft, tomato, cut grass	<0.0001
1-octanol	0.08 ± 0.015^{A}	0.07 ± 0.008^{A}	0.02 ± 0.005^{B}	Rancid, musty	<0.0001
Phenylethyl alcohol	0.08 ± 0.019^{A}	0.08 ± 0.013^{A}	0.07 ± 0.013^{A}	Floral, sweet	0.2428
1-nonanol	0.07 ± 0.014^{A}	0.06 ± 0.006^{B}	0.01 ± 0.003^{C}	Rancid	<0.0001
Aldehydes					
(E)-2-hexenal	35.3 ± 2.4^{B}	32.7 ± 1.2^{B}	39.2 ± 3.3^{A}	Green, apple-like, bitter, almonds, cut grass, bitter almond like	<0.0001
2,4-hexadienal	0.5 ± 0.1^{B}	0.3 ± 0.1^{B}	0.7 ± 0.1^{A}	Ripe fruity	<0.0001
Nonanal	0.9 ± 0.15^{A}	0.9 ± 0.11^{A}	0.2 ± 0.03^{B}	Fatty, waxy, pungent	<0.0001
Esters					
(Z)-3-hexen-1-yl, acetate	14.9 ± 1.67^{A}	13.5 ± 0.85^{AB}	13.0 ± 2.07^{B}	Green-banana, fruity, Green, green leaves	0.0398
(Z)-2-hexen-1-yl, acetate	0.3 ± 0.04^{A}	0.2 ± 0.03^{B}	0.2 ± 0.03^{C}	Apple, banana, grape	<0.0001
Ethers					
(Z)-2-Pentene, 1-ethoxy-4-methyl	0.1 ± 0.02^{B}	0.1 ± 0.02^{B}	0.2 ± 0.03^{A}	—	<0.0001
Hydrocarbons					
Pentene dimer isomer I	1.3 ± 0.08^{B}	1.4 ± 0.08^{B}	2.0 ± 0.18^{A}	—	<0.0001
Pentene dimer isomer II	1.0 ± 0.08^{B}	1.0 ± 0.06^{B}	1.6 ± 0.16^{A}	—	<0.0001
Pentene dimer isomer III	2.5 ± 0.28^{B}	2.5 ± 0.16^{B}	3.7 ± 0.58^{A}	—	<0.0001
Dodecane	0.04 ± 0.010^{A}	0.01 ± 0.011^{B}	0.04 ± 0.036^{AB}	Undesirable	0.0345
Terpenes					
d-limonene	0.3 ± 0.24^{A}	0.3 ± 0.30^{A}	0.3 ± 0.46^{A}	Greenery, fruity	0.9767
α-copaene	0.05 ± 0.008^{A}	0.03 ± 0.006^{B}	0.02 ± 0.004^{C}	Wood, spicy	<0.0001
β-curcumene	0.02 ± 0.005^{A}	0.01 ± 0.002^{A}	0.02 ± 0.004^{A}	—	0.0585
α-farnesene	0.02 ± 0.007^{A}	0.02 ± 0.005^{A}	0.01 ± 0.003^{B}	Soft cooking of vegetable	0.0021
Σ_{Acids}	1.0 ± 0.1^{B}	1.0 ± 0.1^{B}	1.9 ± 0.3^{A}	—	<0.0001
$\Sigma_{Alcohols}$	5.5 ± 0.5^{A}	3.4 ± 0.3^{B}	2.4 ± 0.2^{C}	—	<0.0001
$\Sigma_{Aldehydes}$	36.6 ± 2.5^{B}	33.9 ± 1.3^{B}	40.1 ± 3.4^{A}	—	<0.0001
Σ_{Ethers}	0.12 ± 0.02^{B}	0.09 ± 0.01^{C}	0.17 ± 0.03^{A}	—	<0.0001
Σ_{Esters}	15.2 ± 1.7^{A}	13.7 ± 0.9^{AB}	13.2 ± 2.0^{B}	—	0.0317
$\Sigma_{Hydrocarbons}$	4.9 ± 0.4^{B}	5.0 ± 0.3^{B}	7.3 ± 0.9^{A}	—	<0.0001
$\Sigma_{Terpenes}$	0.4 ± 0.2^{A}	0.4 ± 0.3^{A}	0.4 ± 0.5^{A}	—	0.9973
Σ, Total volatile compounds	63.7 ± 4.8^{A}	57.5 ± 2.5^{B}	65.5 ± 7.2^{A}	—	0.0050

[#]Peaks identification based on mass spectra comparison with the spectrometer database of the NIST 11 Library, being set a minimum similarity of 90%. [‡]Aroma sensory descriptor found in the literature [2, 3, 38]. [§]P values for the one-way ANOVA. Different letters in the same row show statistical differences from the given mean ($P < 0.05$).

leaves, as well as the significant increase in green fruity sensation perceived in the same oils, it can be hypothesized that there is a relationship between these pentene dimer compounds and the fruity sensation. In fact, a linear correlation could be established between them (R-Pearson = 0.933).

It should be referred that the olive oil sensory characteristics are not only related to their volatile profiles.

Nonvolatile compounds, like the phenolic compounds, also contribute to sensory attributes of VOOs, namely, of the olive oil aroma descriptors [49]. Therefore, the different perceived intensities of some olfactory attributes may not imply a change in the contents of the volatile fraction of the oils under study. Indeed, Marx et al. [9] established positive correlations between the concentrations of oleuropein
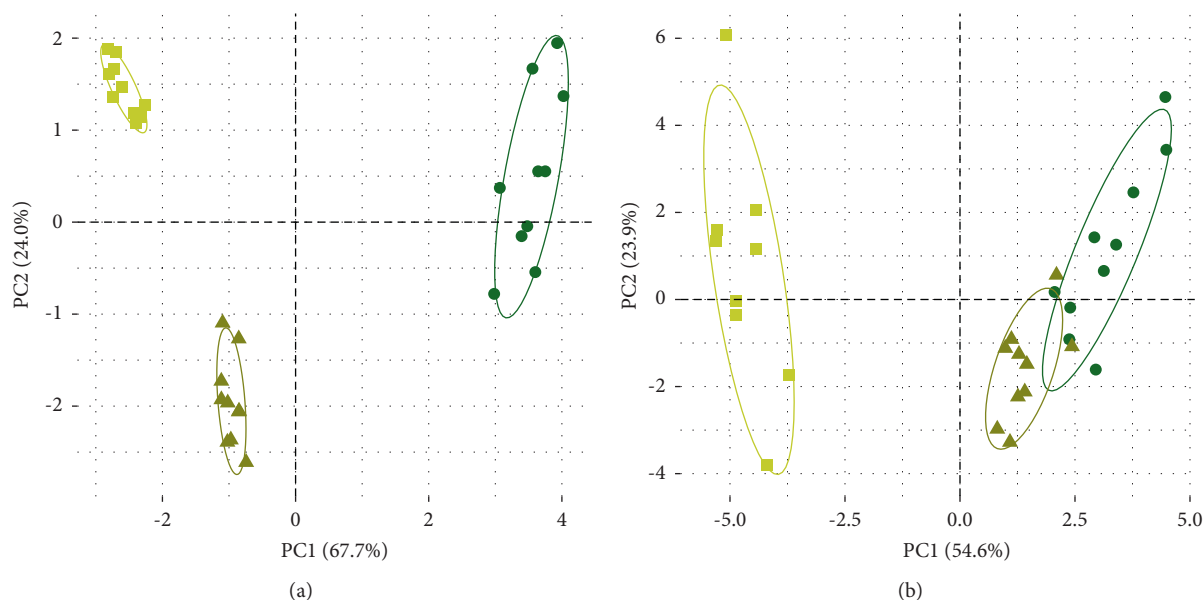
(a)

(b)

FIGURE 1: PCA (biplot) distinction of *cv.* Arbequina olive oils extracted without (control, •) or with the addition of olive leaves (1%, w/w) from *cv.* Arbequina (▲) or *cv.* Santulhana (■): (a) differentiation of *cv.* Arbequina oils based on olfactory sensations and (b) differentiation of *cv.* Arbequina oils based on volatile profile.

derivatives, mainly represented by oleacein, and the intensity of bitter sensation perceived in *cv.* Arbequina oils also extracted with and without the incorporation of leaves from *cv.* Arbequina or Santulhana (*R*-Pearson = 0.999).

3.2. Unsupervised Differentiation of cv. Arbequina Oils According to the Incorporation or Not of Olive Leaves during Extraction. PCA was applied to verify if the olfactory or volatile profiles previously established could allow differentiating *cv.* Arbequina oils without (control) or with leaves (*cv.* Arbequina or Santulhana) incorporation during extraction. As can be observed from the 2D-PCA plots, the olfactory profiles allowed clear differentiation of the three types of studied oils (Figure 1(a)). The volatile profiles were not so successful in differentiating the same olive oils being observed an overlap of the control oils with those extracted after the incorporation of *cv.* Arbequina oils (Figure 1(b)). Although satisfactory overall differentiation results were reported, namely, based on sensory analysis, this procedure is not feasible as a routine monitoring tool, since it is time-consuming, expensive, and would only allow evaluating a low number of samples each day. Therefore, the development of faster and cost-effective complementary analytical strategies, which could allow a noninvasive and direct analysis is envisaged, ensuring the olive oil labeling correctness regarding the deliberated extraction of oils from olives and olive leaves.

3.3. Discrimination of cv. Arbequina Oils Extracted without or with Olive Leaves Incorporation Using a Lab-Made E-Nose. E-noses have been extensively used for olive oil analysis, including its quality grade assessment, the establishment of aroma fingerprints, adulteration detection, and identification of aroma markers related to olive diseases [20–26]. The

present study aimed to study, for the first time, the application of a lab-made E-nose, comprising 9 MOS sensors, to discriminate *cv.* Arbequina oils with similar sensory profiles (i.e., oils extracted with or without the incorporation of 1% (w/w) of olive leaves). The study allowed obtaining a database with the raw resistance signals recorded by the 9 nine MOS sensors, which data matrix was then treated according to 6 different feature extraction techniques, as previously mentioned (LP, INT, MAX, MIN, SUM, and MEAN, totalizing 54 treated signals for each oil sample: 9 MOS × 6 feature extraction techniques).

First, an E-nose-MOS-PCA model was established using the full database (9 MOS × 6 feature extraction techniques for 3 *cv.* Arbequina oils × 5 independent bottles × 2 replicas). The first three PCs (explaining 95.5% of the data variance) allowed the unsupervised differentiation of the studied olive oils, although some oil samples could be misclassified, as can be inferred by the 3D-PCA plot (Figure 2). Thus, to deeper evaluate the feasibility of the lab-made E-nose-MOS to be used as a noninvasive analysis device, an LDA-SA approach was performed. An E-nose-MOS-LDA-SA model, based on the four treated signals from two MOS sensors (S1_INT, S1_MAX, S2_SUM, and S2_MEAN), selected by the SA algorithm from the 54 treated signals, was established as the best model. The model allowed the correct discrimination of all studied olive oils' samples (100% sensitivity and specificity) for both original grouped data (Figure 3) and LOO-CV variant, with a mean sensitivity of 99 ± 5% (ranging from 75 to 100%, for the random 40 *K*-folds used for validation), for the repeated K-fold-CV variant. It should be further noticed that the two MOS sensors selected by the SA algorithm (S1: corresponding to the TGS 2600 commercial sensor, and S2: corresponding to the TGS 2602 commercial sensor) have also been used by other researchers to
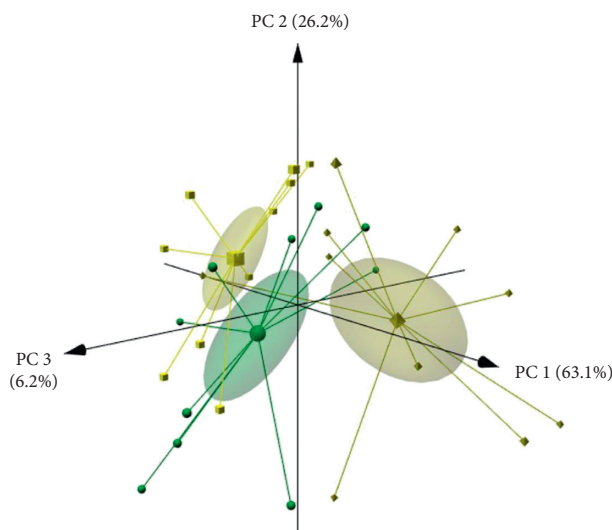
FIGURE 2: E-nose-MOS-PCA model: unsupervised classification of *cv*. Arbequina olive oils extracted without (control, •) or with the addition of olive leaves (1%, w/w) from *cv*. Arbequina (▲) or *cv*. Santulhana (■) differentiation based on the response of resistance signals acquired 9 MOS sensors (9 MOS × 6 different feature extraction parameters = 54 signals: LP, INT, MAX, MIN, SUM, and MEAN).
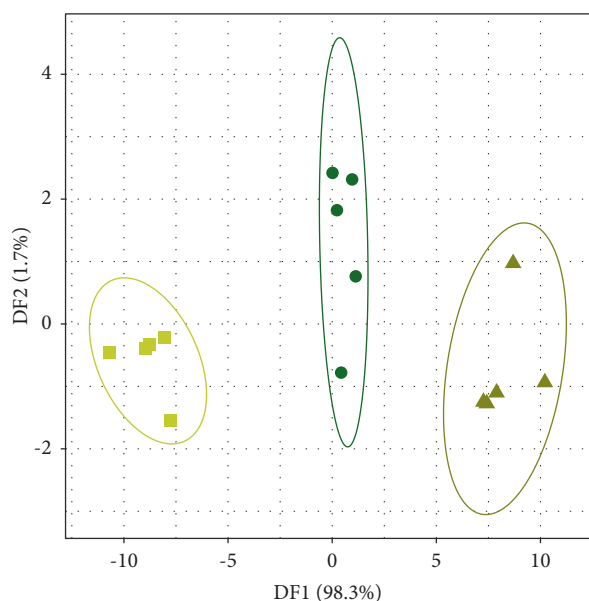

FIGURE 3: E-nose-MOS-LDA-SA model: supervised discrimination (original grouped data) of *cv*. Arbequina olive oils extracted without (control, •) or with the addition of olive leaves (1%, w/w) from *cv*. Arbequina (▲) or *cv*. Santulhana (■) based on four signals recorded by two sensors (S1_INT, S1_MAX, S2_SUM, and S2_MEAN).

satisfactorily discriminate the quality grade or geographical origin of EVOOs [50], seeming that these two TGS sensors may show a specific reactivity towards volatile organic compounds usually found in olive oils.

The satisfactory predictive performance demonstrated the feasibility of using the E-nose as a noninvasive tool to discriminate *cv*. Arbequina oils (control oils) from those extracted with the incorporation of *cvs*. Arbequina or Santulhana leaves, which is of great relevance considering that only the oils extracted without the leaves' addition could be commercialized as EVOO. So the lab-made E-nose-MOS device could be used to detect the (un)deliberated incorporation (of at least 1% w/w) of olive leaves during the oil extraction, being a helpful practical complementary tool for olive oil analysis. Finally, the discrimination capability observed based on the E-nose data also pointed out that the incorporation of olive leaves as well as the leaves cultivar highly influenced the olfactory profiles of the extracted *cv*. Arbequina oils.

4. Conclusions

The incorporation of olive leaves from *cv*. Arbequina or Santulhana (1%, w/w) when *cv*. Arbequina oil is industrially extracted had a significant impact on the olfactory and volatile profiles of the extracted oils. A leaf-cultivar dependency was found to be the observed effects greatly dependent on the cultivar incorporated. Overall, it could be concluded that oils extracted after leaves incorporation possessed a green fruity note while those extracted without leaves showed a ripe fruity note. Regarding the volatile profiles, it was found that in general, the leaves incorporation did not influence the number and type of volatiles detected but influenced their amounts. The contents of alcohols and esters classes significantly decreased with the leaves' incorporation, while the contents of aldehydes and hydrocarbons classes only increased for oils extracted with *cv*. Santulhana leaves, demonstrating the leaf-cultivar dependency. Although the olfactory and volatile profiles could be used to differentiate the extracted oils, the study showed, for the first time, that a lab-made E-nose with MOS sensors could be successfully used to discriminate the studied oils, which is required for labeling purposes, since only oils extracted without leaves deliberated incorporation can be commercialized as extra virgin olive oils. In conclusion, the E-nose could be foreseen as a noninvasive, green, and user-friendly classification device providing an analysis within a short time period (~15 min) and requiring the direct analysis of a low oil volume (~0.5 mL). However, it should be remarked that the study only focused on three different types of oils, all of them extracted from olives of the same cultivar. Thus, considering the wide number of different olive oils commercially available, the satisfactory results achieved with the proposed approach must be validated for a larger database.

Data Availability

The data used to support the findings of this study are available from the authors upon request.

Ethical Approval

This article does not contain any studies with human participants or animals performed by any of the authors.

Consent

Not applicable.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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