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ORIGINAL PAPER

Classification of olive fruits and oils based on their fatty acid ethyl esters content using electronic nose technology

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Abstract

Among several parameters defined for the commercial classes of virgins olive oils (VOOs), there is one, the fatty acid ethyl ester (FAEE), that is only define for the best quality (EVOO). Fruit condition mainly determine these compounds, although, extraction process or deplorable storage condition could rise them up. The FAEE oxidation compound are originated by adding an alcohol chain into the oil molecule. Therefore, the hypothesis of this study is that the inherent constitution of FAEE entails a modification of the volatile profile of oils and olives and this is significant enough to be detected using an electronic nose. With this aim, different samples of olives and oils were analyzed in an accredited laboratory. On the other hand, volatiles from the same samples were captured by an electronic nose. The classification problem was analyzed from two points of view or models. The first was to classify fruits and oils based on whether they are within or outside the legal limits. And the second problem was oriented to classify fruits and oils based on their high or low FAEE content but being within the legal limits. To solve this problem, three classification algorithms were evaluated: Naïve Bayes (NB), Multilayer Perceptron (MLP) and Sequential Minimal Optimization (SMO). For the first model, a well-classified sample rate of 80.3% was obtained for NB and 100% for SMO and MLP, for measurements on oils. The same model evaluated with measurements on olives yielded a success rate of 87.5% with NB, 87.7% with MLP and 82.1% with SMO. For the second model, the success rates remained within the same orders of magnitude. For measurements on oils, the results were 89.7% for NB, 92.5% for MLP and 100% for SMO. And for measurements on olives the results were 77.9% for NB, 88.6% for MLP and 90.9% for SMO. In all cases, the characteristics that worked best were those obtained from the first derivative of the electronic nose response. Based on these results, the e-nose demonstrate to be a non-invasive technology suitable for the classification of olive fruits and oils based on their FAEE content.

Keywords Electronic nose · Ethyl esters · Extra virgin oil · Olive fruit

Introduction

The extra virgin olive oil international demand, increases every year far away from the traditional and ancient center of the Mediterranean diet [1, 2]. New consumers overseas are willing to pay for the nutraceutical values that this high quality oil provides [3–6]. Long shipments require to enforce the oil quality to avoid oxidative reactions that could download the quality, being the most common cause of degradation in oils as others similar produces [7]. Alternatively, to grant the safeness and properties of the oil, the industry, is searching access to quality control able to be perform in real-time at each step of the extra virgin olive oil production [8], which is beginning to be an option [9]. The diversity of sensors able to extract on-time information, entails futures options to automatize part of the production line, or guiding the process to improve the oil quality or to increase the oil extraction yield [10], and also to storage by qualities at the cellar [11, 12]. The correct information of standardize and official methods analysis are needed to calibrate sensors [13]. From all the different sensors, there

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is one, the electronic nose, the one that seems more suitable for the quality control of beverages [14–16]. First essays with e-nose showed its suitability to distinguish varieties and geographical origins in extra virgin oils, honeys, tea and even fruits [16–21]. This device is integrated by different sensors connected to generate a response to different volatiles compounds [22–24]. This sensor arrays originates a signal pattern that is recorded, for a previous established time [25, 26]. Furthermore than the recognition abilities, the e-nose device counts with the advantages of delivering on-time information, and to be a non-invasive technology [27]. On the contrary, the methodology for the determination of chemical parameters described in the European Union normative [28–30], precise the use of a large amount of chemical solvents, toxic with the environment and usually requires expertise and prolonged time to obtain the results [31], in this case the e-nose represents an environmental friendly options with instant results [27].

Moreover, recently researches are proposing different methodologies with the E-Nose applied directly over the fruits, with substantial corroborations, as fruit ripeness, healthiness [32–34], and even to identify the commercial class of the oils [35]. Each of the commercial olive oil classes are define by the regulated limits of different oxidative parameters. These parameters are official regulated by the international of Olive Oil Council (COI) [36, 37]. This legislations have also the purpose of fraud controlling, as the mix of different qualities is usual pursued by economics interest to obtain a low cost produces [38–40]. The fruit quality and the extraction conditions affect directly these oxidative parameters. Any mishandling of the fruit and the oil along its extraction, or storage will be express not only in this chemical parameters but also as volatiles compounds that represents healthy fruits and tide process, or in the contrary, volatiles knowns as off-flavors, related to oxidative and fermentative process [41].

The volatile emission is also evaluated under a stablished methodology which consist in a panel test, that have to point positives and negative attributes, already define in a standardize methodology [36, 42]. Most of the studies related to e-nose applications in olive oil classification have been in concern to emulate the panel test evaluations, classifying by quality classes [22, 43] or tasting intensities [44] such as fruitiness intensity [45]. Notwithstanding, there is an intrinsic relationship between the chemical and the panel test evaluation, the high levels of oxidation compounds developed by mishandlings are also express in the oil with negative flavors that also pulldown the oil quality class [41]. It is considered that if healthy fruits, with favorable transport and storage conditions, under low temperature extraction process, the best quality is obtained, the Extra virgin oil, this oil can be consumed immediately after its extraction [10]. The following class, the virgin olive oil, is also able to be directly consume. This second class results from fruit with some affections or mishandlings along the transport or the process, with the consequent upraising of the oxidative parameters and also generating off-flavors [44]. The off-flavors presence it is forbidden in EVOO, and if they appear the commercial class fall into the VOO, the second category [36]. Finally, if fruit condition or the malpractices before and along the process are deplorable, the third class is obtain, the Lampante olive oil (LOO), which is the lowest quality oil and the one that requires an extended industrial chemical procedure before its safe for consumption [46]. Only mechanical procedures are perform along the extraction process independently of the final class obtained [47].

Experiments have demonstrated that mixing healthy with spoiled fruits downgrade the oil quality by increasing oxidation parameters, and affecting the sensory features [48]. The e-nose has the possibility to be integrated in the reception yard, for a quick fruit classification according to its healthiness or ripeness [32, 34, 43].

From all the oxidative compounds legally restricted there is one that mainly represents a marker of the phytosanitary state of the fruit, and this is are the fatty acid ethyl esters, because its concentration is soon determined after the olives harvest, with only small variations along the extraction process [48, 49]. The stablished limits of FAEE where settle in 2016 by the Commission Delegated Regulation (EU) 2016/2095 [46]. The limits are only stablished for EVOO, and not for VOO neither for LOO [31]. This parameter regulation also intends to avoid frauds by illegal mixtures of high quality, EVOO, with mild deodorized olive oils, a tentative illegal practice [39, 50]. According to literature [49, 51], the development of FAEE is strictly related to the concentration of free fatty acids and short-chain alcohols, mainly ethanol, usually generated in fermentative processes triggers by inappropriate fruit condition [52]. The constitution of FAEE is a combination of a free fatty acid with an alcohol chain, and so it means with the possibility to be express in as volatiles pattern able to be recognize by the e-nose. In concordance to this, recent studies expose that high FAEE content in oils, induce higher variations between the participants of the panel test [36], which indicates the volatile expression of the FAEE compounds.

Furthermore, a recent research confirms that other chemical parameters associated to oxidation reactions are usefull to discriminate by classes with the e-nose [53]. In view of this knowledge we consider that the E-nose could be a suitable sensor to classify olive oil according to a pre-established categories of FAEE content. From the e-nose signal main features will be extract to evaluated the machine learning performance of three different algorithms The Naïve Bayes (NB), the Multilayer Perceptron (MLP), and the Support Vector Machine, based in Sequential Minimal Optimization (SMO). In consideration that FAEE are mainly settle in olives batches, with only few variations along the extraction process [48, 54, 55], the olives fruit batches will be evaluated by the e-nose before being submitted to a standardize laboratory oil extraction methodology. Similarly to the methodology implemented by [53], the chemical results of FAEE oil content will guide the clusters definition for the classification model. Firstly, two cluster will be defined by the regulated limits of FAEE. This means that first class is limited to a maximum of 35 mg/kg, and the second class includes those above that limits, as it is indicated to differentiate EVOO from VOO. The recognition of a *supreme* EVOO with very low levels of FAEE could be useful in deciding those EVOO suitable for long term storage or overseas shipments, granting that this compounds will not rise over the legal limit declared in the labelling [11]. This need has encouraged the generation of a second model, in which only the one classes samples will be defined by an even more restricted limits of FAEE, a *supreme* EVOO. This e-nose application could work as a first scan, useful to narrow the number of laboratory analysis, saving time and money. Besides it could support panel taste, by indicating those EVOO with higher content of FAEE that might confuse the tasting [36]. The approach to this technology entails benefits to oil producers, retailers, and also safety for consumers.

Following Sect Materials and Methods, will present, the procurement of samples and its chemical analysis, also the features extraction from the e-nose response and the classification algorithms to be evaluated. Results of the volatile emission after the classification algorithm will be discuss in Sect Results. And finally, in Sect Conclusions, the conclusion of this research will be presented.

Materials and methods

Olive samples

The olive fruit batches, belonging to a typical Spanish cultivar *Picual*, were harvested along the 2017/2018 season from different producers of Jaen province (Spain) that belong to the local Olive Oil Cooperative PICUALIA (Bailen—Jaen).

From November 2017 to January 2018, 84 batches of olives, 3 kg each, were weekly randomly collected at the Cooperative, immediately after the cleaning step and right away shipped to the Group of Robotics, Automation and Computer Vision (GRAV) laboratory of the University of Jaen in order to be processed within 12 h.

Olive oil extraction

The extraction runs were carried out following the protocol performed by [54] and using a laboratory scale oil mill Abencor ® System, able to mill 8 kg of olives per hour and equipped with a hammer mill, a thermo-mixer and a centrifugal machine.

For each oil extraction experimental runs, oil was immediately stored in two dark glass 125 mL bottles, in the dark at 20 ± 1 °C, in order to delivered one to the accredited CM Europe laboratory for the chemical analysis. And the other one was destined to the electronic nose evaluation. The e-nose analysis was performed in less than 24 h after the oil extraction, to measure the best volatile expression of the oil, avoiding oxidative degradation related to storage.

Chemical analysis of oil and its statistics treatment

Each olive oil sample were analysed by the official methods in triplicates. Acidity index, peroxide value, K_{232}, K_{270}, and FAEE according to the European Regulation [28, 30, 46], in the accredited CM Europe laboratory.

Statistical analysis

Chemical results were analysis by one-way ANOVA using the Tukey's test, significant differences estimated at $P < 0.05$. Tukey's test is a statistical test used generally and in conjunction with ANOVA. The Tukey test is used in experiments that they involve a large number of comparisons. The computational complexity is low since a single comparator is defined, resulting from the product of the standard error of the mean by the tabulated value in Tukey's table using as numerator the number of treatments and as denominator the degrees of freedom of the error.

All statistical analyses were performed with the InfoStat software 2018 version. Significant differences will be presented with different letters in the tables shown in the results section.

Electronic nose characterization

The analysis of the headspaces of each batch of olives fruits and their corresponding olive oil was performed using an electronic nose device, the PEN3 (Airsense Analytics GmbH, Schwerin, Germany), consisting of a gas sampling unit (maximum flow rate of 600 mL/min), a software (Win Muster v. 1.6.2), and an integrated sensor array composed of 10 different thermos-regulated (200–500 °C), metal oxide thick film sensors (MOS), sensitive to different classes of chemical compounds: W1C (aromatic organic compounds), W5S (very sensitive, broad range sensitivity, reacts to nitrogen oxides, very sensitive with negative signals), W3C (ammonia, also used as sensor for aromatic compounds), W6S (detects mainly hydrogen gas), W5C (alkanes, aromatic compounds, and non-polar organic compounds), W1S (sensitive to methane and a broad range of organic compounds), W1W (detects inorganic sulphur compounds, and also sensitive to many terpenes and sulphur containing organic compounds), W2S (detects alcohol, partially sensitive to aromatic compounds, broad range), W2W (aromatic compounds, inorganic sulphur and organic compounds), W3S (reacts to high concentrations of methane and aliphatic organic compounds).

Data acquisition by the electronic nose

For each olives' batch, two samples of 250 g each, were randomly selected and put into a 1000 mL glass beaker tightly sealed with parafilm (Fig. 1). Olives were then left to rest 8 min at room temperature (23 ± 1 °C), before the measurement process started, in order to increase the volatile concentration in the headspace. For each oil samples the procedure was similar, in this case 5 g of oil were put into a glass vial (13.5 mL), hermetically sealed with parafilm and conditioned for 5 min at 30 ± 1℃, before the measurement process started, in order to accumulate the volatiles in the headspace (Fig. 2).

As previously described by [25] each measurement process starts with a sensor array cleaning stage (60 s), when air, after passing through an activated carbon filter, reaches the sensor array. In the second step, air crosses again through an activated carbon filter and then through the sample; after passing through a moisture and particle filter, air finally reaches the array of sensors (60 s). When the sample volatile compounds react with the sensing film of the sensor, an oxygen exchange occurs resulting in a decrease of electrical conductivity, detectable by a transducer element (electrode) attached to each sensor. The data acquisition frequency of our experiments was 1 measure per second, for each sensor.

Features extraction

The response signal of the electronic nose for each sample was a transient time curve related to the evolution of the electrical resistance value for every MOS sensors included in the sensor chamber. Each transient responses were normalized on the basis of the final resistance value obtained after the cleaning process of each sensor chamber. For each sensor the normalized transient response was then formalized according to Eq. 1.

$$x'[n] = \frac{x[n]}{x[1]} \tag{1}$$

where x[n] is the resistance value electronically read from the sensor, x[1] is the electronic resistance of the sensor at the beginning of the measurement process (when all sensors are clean) and n is the number of acquisition and goes from 1 to 60. Also the first and the second derivative of $x'[n]$ were considered and then $\dot{x}'[n]$ and $\ddot{x}'[n]$ were computed according to Eqs. 2 and 3 respectively.

$$\dot{x}'[n] = x'[n] - x'[n-1] \tag{2}$$

$$\ddot{x}'[n] = \dot{x}'[n] - \dot{x}'[n-1] \tag{3}$$

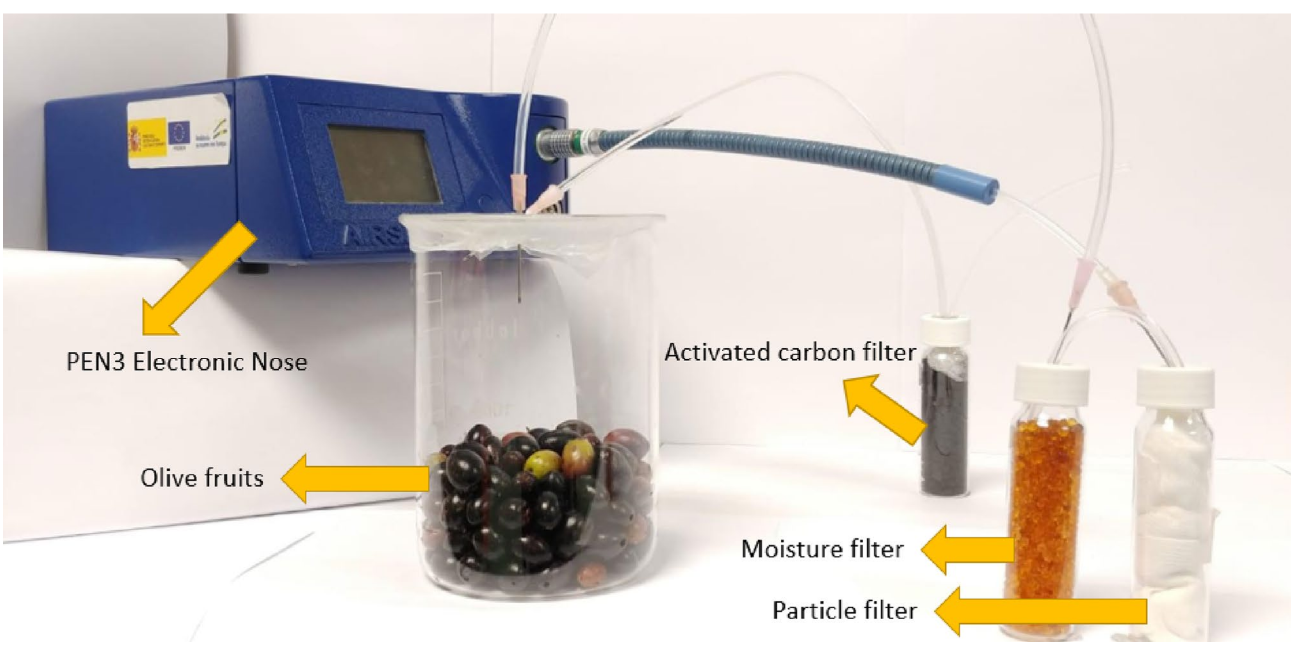

Fig. 1 Setup configured for the olive sample measurement process. In the same image it can see the electronic nose device, the container of olives and the air filters

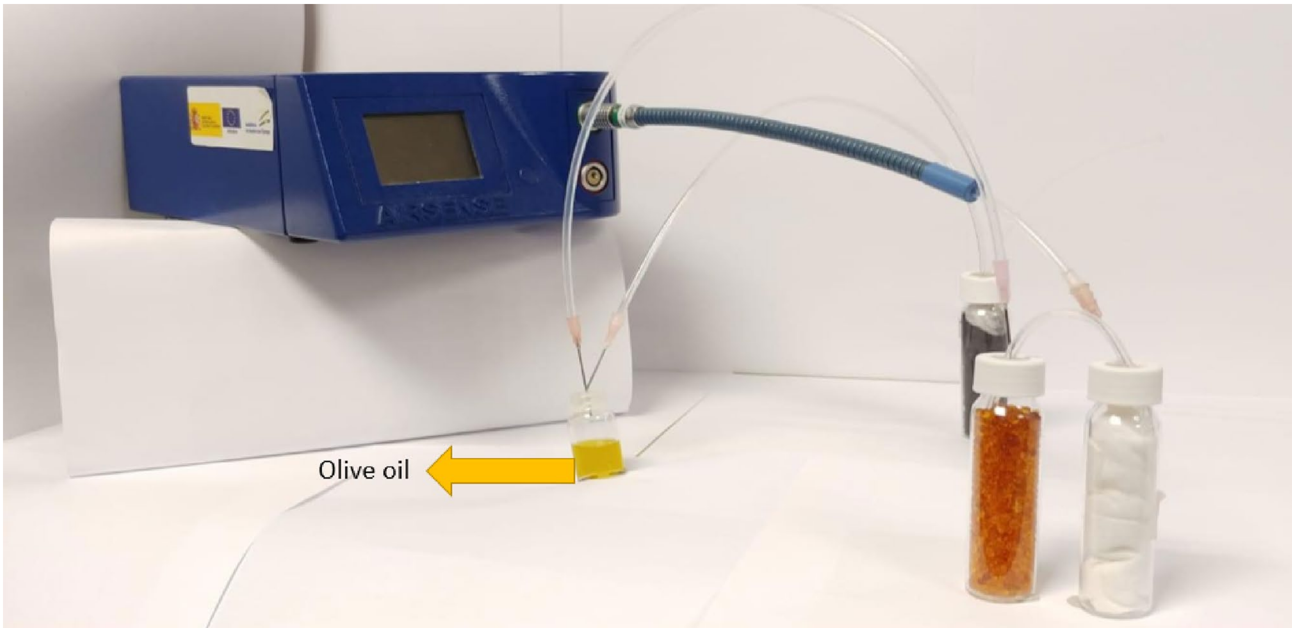

Fig. 2 Setup configured for the olive oil sample measurement process

The second stage of the feature extraction process was to compress the transient response of the sensor array to form a feature vector, useful as an olfactory fingerprint. The olfactory fingerprint was generated using a transient compression method based on the parameter extraction. The extracted features were the median (Eq. 4), sum (Eq. 5), mean (Eq. 6), standard deviation (Eq. 7), confidence (Eq. 8), variance (Eq. 9), minimum value (Eq. 10), maximum value (Eq. 11) and end point (Eq. 12).

$$f[1] = x\left[n/2\right] \tag{4}$$

$$f[2] = \sum_{n=1}^{60} x[n] \tag{5}$$

$$f[3] = \frac{\sum_{n=1}^{60} x[n]}{60} \tag{6}$$

$$f[4] = \sqrt{\frac{\sum_{n=1}^{60} |x[n] - f[3]|^2}{59}} \tag{7}$$

$$f[5] = f[3] \pm 1.96\frac{f[4]}{\sqrt{60}} \tag{8}$$

$$f[6] = \frac{\sum_{n=1}^{60} |x[n] - f[3]|^2}{60} \tag{9}$$

$$\mathrm{f}[7] = \min(\mathrm{x}[n]) \tag{10}$$

$$f[8] = max(x[n]) \tag{11}$$

$$f[9] = x[60] \tag{12}$$

Classification algorithms

In order to evaluate the possibility of using the electronic nose to classify samples of olives fruit batches and oil according to their quality classification based on the ethyl ester content of the final olive oil, three supervised classification algorithms were tested: a Naïve Bayes classifier (NB), an Artificial Neural Network type Multilayer Perceptron (MLP) and a Sequential Minimal Optimization algorithm (SMO), using the data mining procedure of WEKA ver. 8.0 (Statsoft Inc., Tulsa, OK, USA).

The Naïve Bayes classifier [56] is a probabilistic classification method and is based on obtaining the probability of belonging to each class. Since the extracted characteristics are continuous variables and are normally distributed, the distribution of each class can be represented as a Gaussian probability density function in terms of its mean μ_c and standard deviation σ_c. In this way the probability of belonging of each sample to each class will be given by Eqs. 13 and 14.

$$p\left(v_{n,s\times f}|c\right) = g\left(v_{n,s\times f};\mu_c,\sigma_c\right), where \tag{13}$$

$$g\left(v_{n,s\times f};\mu_c,\sigma_c\right) = \frac{1}{\sigma_c\sqrt{2\pi}}e^{-\frac{(v-\mu_c)^2}{2\sigma_c^2}} \tag{14}$$

where $g\left(v_{n,s\times f};\mu_c,\sigma_c\right)$ is the Gaussian probability density function.

Also, the MLP classifier was evaluated [57]. The multilayer perceptron neural network is a neural network formed by multiple layers and has the advantage of being able to solve classification problems where the classes are not linearly separable. For our case, the MLP network was configured with three layers. An input layer with 90 nodes (equal to the number of features extracted multiplied by the number of sensors), a hidden layer with 58 nodes with sigmoidal transfer functions and an output layer with 2 nodes (equal to the number of classes). The training of the network was backpropagation type and the number of times used to train the MLP was 500.

The last classifier was SMO and it is an algorithm for training Support Vector Machines (Cortes and Vapnik, 1995). The main idea is to find the hyperspace where our considered classes could be optimally separated. This approach is based on a decision boundary which can be described as a hyper-plane that is expressed in terms of a linear combination of functions parameterized by support vectors that give the best separating hyper-plane using a kernel function (Eqs. 15 and 16).

$$\min_{\beta,\beta_0} \frac{1}{2}\beta^2 + C\sum_{i=1}^{N}\xi_i \tag{15}$$

$$subject\ to\ y_i\left(x_i^T\beta + \beta_0\right) \geq 1 - \xi_i\ \forall i \tag{16}$$

For ethyl ester class discrimination, the features of the signals of the sensors were used as the inputs in the classification model and the outputs were the class assigned to each model. Two different classifications were tested (see Sect Statistical results for the laboratory analysis).

The criteria based on hold out samples was used to validate the classification models. The proposal is to employ a percentage of samples to train de model and the rest to validate it. In our case three *hold-out* percentages were tested: 33%, 50% and 66%.

The performance of the classification models was assessed in terms of prediction accuracy, Kappa statistic and the area under the curve AUC. The accuracy of the prediction was defined by dividing the number of correctly classified samples in the respective class, by the total number of samples analysed. The Kappa statistic is used to measure the reliability for categorical items and it takes into account the possibility of the accuracy occurring by chance. Finally, the AUC represents the relation between the true positive rate and false positive rate. Higher the AUC and the Kappa values, better the model is at distinguishing between classes.

Results

Statistical results for the laboratory analysis

The laboratory results for the FAEE parameter was the basis for the category assignment of olive oils. These parameter, FAEE, as the others parameters results were submitted to the statistical one-way ANOVA using the HSD Tukey's test, with significant differences estimated at $P < 0.05$. Two different binary classification models were independently evaluated.

Firstly, the Binary Classification I: Following the European legislation for the FAEE content [46], two classes were defined. First class (A), fulfils the requirement for the EVOO class, according to the FAEE. Therefore, the samples selected for this class had less than or equal to 35 mg /kg. It has been considered to include in this class a single sample that has all the parameters to belong to this class except the peroxide index, with a value of 22.5. Instead, for the second class (B), samples with higher values of FAEE ($x \geq 36$ mg/kg) were considered. This two classes statistical comparative results are presented in Table 1.

The results in Table 1, shows that classification according to the European regulation [46] presents statistical differences for the FAEE and also for the free fatty acid and peroxide values. Aside in the K270 and K232 no differences were found, however, this results are accordance to others studies where variations of this two parameters were hardly seen [58, 59]. It must said that apart from the FAEE, the mean value of the other parameters are under the limits of the EVOO category, even though the B class correspond to the VOO assigned category.

Secondly, for the Binary Classification II: the aim was to evaluate differences in between EVOO with higher or low content of FAEE. With this purpose two classes, named C and D, both of them with an FAEE content below the EVOO limits, ≤ 35 mg /kg oil, will be compare. In this case class C will consider samples with a maximum of 10 mg/kg of FAEE, a very low content, considered for this work as a *supreme* EVOO. The limited value of the C class was obtained as the median value from all the 84 olive oil samples. The limits of class D were settle by $11 \leq x \leq 35$ mg / kg. This mean that class D correspond to EVOO with higher contents of FAEE, but still remaining inside the EVOO regulated limits for this parameter. The comparative statistical results for this two classes are presented in Table 2.

In Table 2 it can be observed that the Peroxide value as well as K_{270} and K_{232} have non-significant difference, meaning that in terms of these oxidation parameters, classes C,

Table 1 Chemical results of olive oil samples for the Binary Classification I

Binary Classification I						
		Ethyl Ester (mg·kg^{-1})	Free fatty acid %	Peroxide value (meq O2·kg^{-1})	K270	K232
Class A	Max	34.00	0.59	22.50	0.24	2.29
	Mean	13.78^{a}	0.29^{a}	5.61^{a}	0.16^{a}	1.53^{a}
	Min	3.00	0.12	2.10	0.09	1.26
	SD	8.27	0.10	4.06	0.04	0.17
	SEM	1.15	0.01	0.57	0.005	0.02
Class B	Max	104.00	0.79	17.20	0.17	1.53
	Mean	65.17^{b}	0.61^{b}	11.05^{b}	0.13^{a}	1.41^{a}
	Min	36.00	0.38	5.10	0.10	1.28
	SD	21,22	0.16	4.83	0.02	0.10
	SEM	8.66	0.06	1.97	0.009	0.04

Maximum (max), mean, minimum (min), standard deviation (SD) and standard error of the mean (SEM) values are presented by class. Statistical differences (Tuckey´s, $p \leq 0.05$), between classes are shown when different letters are presented in the mean value

the *supreme* EVOO, and D are not different from each other. For the free fatty acid parameter and the FAEE content, however, statistical differences are shown between this two classes. The class C, *supreme* EVOO, has significant differences of oxidative compounds, from the normal EVOO, class D. Indeed these results are to be expected, because the synthesis of FAEE requires free fatty acid for their esterification [51]. The use of deteriorated olives fruit could be associated with the increase of these two parameters [48]. This results confirm that the FAEE content could as the adequate parameter to stablish a superior class of EVOO, which has also been supported by [11, 36].

This statistical analysis results, confirm the suitability of the FAEE parameter, to generate a classification methodology for the further machine learning analysis, of the electronic nose volatile response.

Algorithm classification results over olive oils samples

The machine learning process was performed with the array of features extracted from the e-nose response, according to Sect Materials and Methods. These features were analyzed by the tree different algorithm, (NB, MLP, SMO), all of them describe in Sect Materials and Methods.

Binary classifications I and Binary classifications II were tested independently in each of the tree algorithms. In all of them results will be presented by the *hold out*.

Table 2 Chemical results of olive oil samples for the Binary Classification II

Binary Classification II						
		Ethyl Ester (mg·kg^{-1})	Free fatty acid %	Peroxide value (meq O2·kg^{-1})	K270	K232
Class C	Max	10.00	0.48	11.30	0.28	1.92
	Mean	6.30^{a}	0.24^{a}	4.39^{a}	0.16^{a}	1.50^{a}
	Min	3.00	0.12	2.10	0.09	1.26
	SD	1.85	0.08	1.75	0.04	0.15
	SEM	0.40	0.01	0.38	0.008	0.03
Class D	Max	34.00	0.59	22.50	0.24	1.97
	Mean	18.50^{b}	0.33^{b}	6.22^{a}	0.16^{a}	1.56^{a}
	Min	11.00	0.15	2.40	0.10	1.28
	SD	6.74	0.11	4.69	0.03	0.16
	SEM	1,23	0.02	0.85	0.005	0.03

Maximum (max), mean, minimum (min), standard deviation (SD) and standard error of the mean (SEM) values are presented by class. Statistical differences (Tuckey´s, $p \leq 0.05$), between classes are shown when different letters are presented in the mean value

Results for the Binary classification I are presented in Table 3. It is possible to see that the first derivative of the features, successfully work to classified class A and B with an accuracy and precision of 100%, with SMO and the MLP algorithm.

Results presented in Table 3 expose the highly sensitive response of the e-nose sensor to the volatile emission from the olive oil that could be related to the FAEE parameter. This e-nose is able to determine the belonging commercial class A as EVOO and class B as VOO, according to the FAEE content of the oil. These results are even better than a similar classification by other oxidative parameter presented by [53]. Therefore, it confirms that the FAEE compounds has a volatile expression able to be recognize by the e-nose and a proper feature extraction analyses under an specific algorithm, for this case SMO or MLP.

In Table 4 results are presented for the Binary classification II. An even better performance is observed reaching a 100% for correct classification, with a holdout of 66% by the SMO algorithm. In this case only raw data was needed to the best classification results, and so it was excluding the first and the second derived results, as were not supporting any further analysis. The perfect classification of oils under this categories, show the potential of the e-nose as a perfect sensor to recognize EVOO that have small quantities of FAEE compounds, the *supreme* EVOO, as we have named class C.

The feasibility of the e-nose to identify these *supreme* EVOO, encourage the applicability of this sensor as a quick scan to categorized those oil that could have an extended shelf life, conserving the oil oxidative properties under the limits of EVOO, covering with this a need which has been recently presented by the industry [51].

Algorithm classification results over olive fruit batches

The same approach was applied for the e-nose volatile pattern respond applied over olive fruits. The two binary classification were evaluated with the same algorithm.

Table 3 Classification Performance of the three algorithms (Naïve Bayes classifier NB, Artificial Neural Network type Multilayer Perceptron (MLP) and Sequential Minimal Optimization algorithm SMO) tested in predicting the belonging of olive oils' samples to the two classes defined by the Binary Classification I, using the extracted features of the sensors'response (($x'[n]$)), their first ($x'[n]$) and their second derivative (($x'[n]$)) with different hold out percentages (33, 50, 66%)

Sensor response	Classification algorithm	"Hold out" %	Correctly classified %	Kappa statistic	AUC value
$x'[n]$	NB	33	78.57	0.22	0.66
		50	66.67	0.16	0.70
		66	65.52	0.21	0.66
	MLP	33	69.64	0.07	0.60
		50	76.19	0.05	0.76
		66	79.31	0.37	0.80
	SMO	33	78.57	0	0.50
		50	76.19	0.29	0.66
		66	79.31	0.20	0.57
$\dot{x}'[n]$	NB	33	80.36	0.31	0.67
		50	69.05	0.19	0.65
		66	65.52	0.14	0.59
	MLP	33	98.21	0.94	1.00
		50	100	1	1
		66	100	1	1
	SMO	33	100	1	1
		50	100	1	1
		66	100	1	1
$\ddot{x}'[n]$	NB	33	76.79	0.19	0.63
		50	66.67	0.16	0.63
		66	58.62	0.05	0.57
	MLP	33	69.64	0.13	0.60
		50	57.14	-0.08	0.59
		66	68.97	0.19	0.60
	SMO	33	67.86	0.32	0.55
		50	64.29	-0.01	0.49
		66	65.52	0.06	0.53

Table 4 Classification Performance of the three algorithms (Naïve Bayes classifier NB, Artificial Neural Network type Multilayer Perceptron (MLP) and Sequential Minimal Optimization algorithm SMO) tested in predicting the belonging of olive oils' samples to classes C and D defined by the binary classification II, using the extracted features of the sensors'response ($(x'[n])$). With different hold out percentages (33, 50, 66%)

Sensor response	Classification algorithm	"Hold out" %	Correctly classified %	Kappa statistic	AUC value
$x'[n]$	NB	33	88.46	0.77	0.92
		50	89.74	0.80	0.95
		66	8519	0.70	0.98
	MLP	33	82.69	0.65	0.89
		50	87.18	0.74	0.92
		66	92.59	0.85	1.00
	SMO	33	80.77	0.61	0.80
		50	87.18	0.74	0.87
		66	100.00	1.00	1.00

The results for the Binary classification I, are presenting in Table 5, although only raw data results is presented, because by introducing the 1st and 2nd derivative of the sensor responses, no improvements were registered, therefore these data are not show.

There is a high response of the e-nose to the volatiles that merge from olives fruit batches. Results show a high accuracy of relating the volatile pattern of olives fruit to the FAEE content of their corresponding oils. These classification categories based on the legislation requirements for FAEE, and adopting the MLP approach, with a "hold out" validation percentage of 66%, it was possible to discriminate the two classes with a correct classification of 88%. Similar percentages could be obtained also with a lower "hold out" percentage (33%), both for the MLP and NB classification algorithms.

This methodology generates expectations due to possible applications for the industry. The use of the e-nose could provide early information of the expected quality of olive oils even before processing the fruit. The early prediction of this kind of parameters has only been explore successfully by visual inspection system, considering for its purpose ripeness and healthiness of olives fruits as classification features[54].

Furthermore the performance of the algorithm in the Binary classification II is even better, reaching up to a 90.9% of correctly classification of categories define for class C and D, results presented in Table 6. This best algorithm for this classification was the SMO, with a holdout of 50%. First and second derivative were also performed, nevertheless, no improvements were obtained (data not presented).

The increase in accuracy exposed in these results, reflects the higher variability that might be found in the best commercial labeling class according to the FAEE. The applicability of this model to sort between olives batches, could ensure homogenous lots for milling process seeking to obtain an EVOO with the lowest content of FAEE, a *supreme* EVOO, avoiding to fall in the borderline for this parameter, which has been demonstrate to rise even at fine storage or along the shelf life [11].

Table 5 Classification Performance of the three algorithms (Naïve Bayes classifier NB, Artificial Neural Network type Multilayer Perceptron (MLP) and Sequential Minimal Optimization algorithm SMO) tested in predicting the belonging of olives' samples to the two class defined by the Binary Classification I, using the extracted features of the sensors'response ($(x'[n])$), with different hold out percentages (33, 50, 66%)

Sensor response	Classification algorithm	"Hold out" (%)	Correctly classified (%)	Kappa statistic	AUC value
$x'[n]$	NB	33	87.50	0.68	0.89
		50	83.13	0.58	0.86
		66	78.95	0.46	0.92
	MLP	33	86.61	0.57	0.89
		50	84.34	0.54	0.90
		66	87.72	0.68	0.94
	SMO	33	82.14	0.36	0.64
		50	78.31	0.27	0.61
		66	80.70	0.40	0.66

Table 6 - Classification Performance of the three algorithms (Naïve Bayes classifier NB, Artificial Neural Network type Multilayer Perceptron (MLP) and Sequential Minimal Optimization algorithm SMO) tested in predicting the belonging of olives' samples to the three class defined by the Binary Classification II, using the extracted features of the sensors'response $((x'[n]))$, with different hold out percentages (33, 50, 66%)

Sensor response	Classification algorithm	"Hold out" %	Correctly classified %	Kappa statistic	AUC value
$x'[n]$	NB	33	75.96	0.54	0.90
		50	77.92	0.56	0.88
		66	66.04	0.38	0.88
	MLP	33	85.58	0.71	0.96
		50	88.31	0.76	0.96
		66	88.68	0.76	0.98
	SMO	33	84.62	0.69	0.85
		50	90.90	0.80	0.90
		66	86.79	0.72	0.87

Conclusions

In this study two different binary classification models were proved successfully for olive oil and for olive fruit batches. Firstly, Binary Classification I, obtained 100% of precision and accuracy for olive oils, using the Sequential Minimal Optimization (SMO) and the Multilayer Perceptron (MLP) approaches with all the "hold out" percentage (33–50-66%). In this case both classes were defined by the European legislation limit for the FAEE parameter. Although this model performed over olives batches reached an accuracy of only 88% (MLP; 66% hold out), which is still very significant as no research has been performed with the aim of predicting FAEE, starting from the volatile emission of the raw materials (olive fruit batches) by means of an e-nose.

Secondly, outstanding results were obtaining under the Binary Classification II, reaching 100% for the oils samples, with SMO algorithm, 66% hold out. The exposed results could generate futures applications by detecting an outstanding EVOO class, corresponding to those oils outside the borderline of the limits of FAEE, thereby minimizing the risk to exceed the regulated limits along the estimated shelf life with harmful economic consequence. A similar approach might be suitable for olive batches, where Binary Classification II obtained up to 91% of correct classification performing the SMO algorithm at 50% hold out. The methodology of this work show that the e-nose sensor not only work fine to classify olive oil categories, but also the suitability to recognize volatiles expression of oxidative compounds by its application directly over olives fruit batches, and so to be use a prediction sensor of the oil quality. These results suggest the use of the e-nose as a on-line sensor in the EVOO production line, and even to control the evolution of the FAEE in storage conditions.

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