Electronic noses and tongues to assess food authenticity and adulteration

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ABSTRACT

Background

There is a growing concern for the problem of food authenticity assessment (and hence the detection of food adulteration), since it cheats the consumer and can pose serious risk to health in some instances. Unfortunately, food safety/integrity incidents occur with worrying regularity, and therefore there is clearly a need for the development of new analytical techniques.

Scope and Approach

In this review, after briefly commenting the principles behind the design of electronic noses and electronic tongues, the most relevant contributions of these sensor systems in food adulteration control and authenticity assessment over the past ten years are discussed. It is also remarked that future developments in the utilization of advanced sensors arrays will lead to superior electronic senses with more capabilities, thus making the authenticity and falsification assessment of food products a faster and more reliable process.

Key Findings and Conclusions

The use of both types of e-devices in this field has been steadily increasing along the present century, mainly due to the fact that their efficiency has been significantly improved as important developments are taking place in the area of data handling and multivariate data analysis methods.

Keywords:

Electronic nose; Electronic tongue; Food adulteration; Authenticity assessment; Food analysis

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1. Introduction

The rigorous, objective assessment of food authenticity has become of paramount importance, mainly due to the problem of adulteration (a legal term meaning that a food product fails to meet legal standards, *i.e.* noncompliance with health or safety regulations). Unfortunately, major food adulteration events seem to occur with worrying regularity, and there is no doubt that the concern for this fact will increase concurrent with population pressures. Therefore, there is a growing need for nonstop vigilance, which means research and development of rapid analytical and detection techniques in the field of food authenticity assessment. In this sense, two approaches are emerging as promising tools in the attempt to efficiently address this issue (Borrás et al., 2015), namely: electronic noses (e-noses) and electronic tongues (e-tongues). Both are sensor systems, but they do not look at the same features when applied to a given liquid sample; the former are in contact with its headspace, whereas the latter are immersed in the sample (Cosio et al., 2015).

Electronic noses are devices which mimic the sense of smell. These instruments generally consist of an array of sensors utilized to detect and distinguish odors in complex samples and at low cost. These characteristics make them very useful for different applications in many areas, including food industry. In this context, a lot of papers have appeared in the present century in the literature describing the use of e-noses in food analysis processes.

On the other hand, e-tongues are analytical devices (groups of sensors) mainly employed to identify and classify the tastes of several chemical substances in beverages or liquid phase food samples, their mode of operation “imitating” the human sense of taste. E-tongues can be utilized to characterize multicomponent mixtures for both qualitative and quantitative purposes, hence the increasing attention they are receiving in the field of food analysis, as shown in recent surveys in the literature.

In the last years, many reviews on e-noses and/or e-tongues fundamentals and applications in several research areas have been published in the literature, mainly in the field of food analysis (e.g., Boeker, 2014; Ciosek and Wróblewski, 2011; del Valle., 2012, Escuder-Gilabert and Peris, 2010; Kiani et al., 2016; Loutfi et al., 2015; Peris and Escuder-Gilabert, 2009; Rodríguez-Méndez, 2016; Śliwińska et al., 2014; Tahara and Toko, 2013; Vlasov et al., 2005). This paper will then focus on the employment of both e-noses and e-tongues in food authenticity assessment (and hence the detection of food adulteration). After briefly commenting the fundamentals of this type of devices, the most relevant contributions in this field over the past ten years will be dealt with. In this sense, and as a general overview, in a recent chapter of a book (Karoui, 2012) devoted to food authenticity and fraud, Karoui discusses the relative potential and ease of application of different technologies for the confirmation of food quality and adulteration. Special emphasis is put on e-nose technology (combined with chemometric tools) as a promising technique in this field. Some examples clearly show that there has always been a risk of fraud, since food became a trade object. The chapter also describes the different kinds of food adulteration and related fraudulent practices, with details of detection methods, including the use of e-noses. In a similar way, Cappozzo (2013) has presented recent analytical innovations for quality assurance in the detection of food adulteration through the utilization of e-noses. Panchariya et al. (2013) have reported an overview of the applications of e-noses and e-tongues for classification and authentication of beverages. As far as e-tongues are concerned, Sliwinska et al. (2014) have also dealt with their potential in the authenticity and falsification assessment of foodstuffs.

2. General concepts

Major components of both electronic devices are widely described in the literature and their details are therefore omitted in this paper. Nevertheless, in this section the general concepts of the electrochemical methods applied in these e-systems are briefly mentioned in order to help potential readers to better understand the principles behind these techniques.

2.1. Fundamentals of e-noses

E-noses are designed to detect and distinguish among complex odors (from food samples) making use of a sensor array, which is composed of broadly tuned (non-specific) sensors that are treated with different odor-sensitive (bio)chemical substances. An odor stimulus now yields a characteristic fingerprint (or smellprint) from the group of sensors. These patterns from known odors are then utilized to generate a database that is subjected to multivariate analysis, so that unknown odors can therefore be identified and classified. Nevertheless, it should be remarked that, in recent years, the usual sensor types used for e-nose instruments have been considerably improved by new technologies developed in this field, and either a set of gas sensors or mass spectrometry (or their combination) are commonly utilized for this purpose. Anyway, and in a broader sense, electronic noses basically consist of three elements (Fig. 1a), namely: (i) a sample handling system, (ii) a detection system, and (iii) a data processing system.

The basis of electrochemical gas sensor operation involves interactions between gaseous molecules and sensor-coating materials which modulate electrical current passing through the sensor, detectable by a transducer that converts the modulation into a recordable electronic signal (Rodríguez-Méndez, 2016), which is then amplified and conditioned. Thereafter, a digital converter transforms the signal from electrical (analog) to digital, and finally a computer microprocessor reads the digital signal and displays the output after which the statistical analysis for sample classification or recognition is performed.

There are many different types of electrochemical sensors (e.g. metal-oxide gas sensors, metal-oxide semiconductor field effect transistors, acoustic wave gas sensors, electrochemical gas sensors, quartz crystal microbalance sensors, conducting polymer gas sensors, surface acoustic wave devices, field-effect gas sensors, fiber-optic gas sensors, and others) and many different types of sensor-coating materials which are classified according to additive doping materials, the type and nature of the chemical interactions, the reversibility of the chemical reactions and running temperature. A summary of the types and mechanisms involved with some common gas sensor technologies is contained in the work of Wilson and Baietto (2009).

Transducer recording devices of various types in electronic-nose sensors are ranked according to the nature of the physical signal they measure. The most common methods make use of transduction principles based on electrical measurements, including changes in current, voltage, resistance or impedance, electrical fields and oscillation frequency. Others involve measurements of mass changes, temperature changes or heat generation. Last but not least, optical sensors measure the modulation of light properties or characteristics such as changes in light absorbance, polarization, fluorescence, optical layer thickness, color or wavelength (colorimetric) and other optical properties.

2.2. Fundamentals of e-tongues

E-tongues can be considered the wet counterparts of e-noses. The output of a non-specific array of sensors shows different patterns for the different taste-causing chemical substances and the resulting data are statistically treated. A wide variety of chemical sensors are currently used in the design of e-tongues, the selection of the sensor group being carried out taking into account the chemical nature of the food samples analyzed. Regardless of the type of sensors utilized, an e-tongue is essentially composed of three elements (Fig. 1b): (i) automatic sampler (although not a necessary component), (ii) a set of chemical sensors with different selectivity, and (iii) software with the suitable algorithm to process the signal and get the corresponding results.

The electronic tongue system relies on sensors with only moderate selectivity and having the so-called cross-sensitivity. In this way, each sensor in the array, in principle, delivers information on the concentrations of a number of analytes, the next step being to decode the signals obtained from the sensor array. The sensors in the array can be of different nature, although Ion Selective Electrodes (ISEs) predominate among the various types of sensors utilized, electrodes with chalcogenide glass membranes (Mikhelson, 2013) being particularly suitable for these devices. An ISE generates a difference in electrical potential between itself and a reference electrode, and this output potential is proportional to the activity of the selected analyte in the sample solution according to the well-known Nernst equation (Skoog et al., 2013), although a calibration of the working electrode should be previously carried out using a series of standard solutions with known composition.
The number of sensors in the array can vary, but most typical number is about 10–20. Unlike in the classical measurements with ISEs, the electronic tongue system can work without a reference electrode. In such a setup, the potential difference is measured for all pairs of the electrodes in the array. This is advantageous since reference electrodes often cause problems with the measurements.

The signals obtained from the sensor array are processed using different chemometrical methods, whereas the interpretation and representation of the data is often based on the principal component analysis method. This allows for the characterization of the samples not only in terms of the concentrations (activities) of the particular analytes, but also for the recognition of the nature of the sample, since different types of samples fall into different places in the principal components plot.

3. E-noses in food authenticity/adulteration assessment

Major applications of e-noses in food authenticity assessment (and/or detection of potential food adulteration) found in the literature are summarized in Table 1 and described in this section.

To begin with, of great importance is the report on the effectiveness of 3 fast procedures for the analysis of volatile substances using principal component analysis (PCA) treatment of data in order to discriminate between virgin olive oil (VOO) samples adulterated with hazelnut oil (Mildner-Szkudlarz and Jeleń, 2008). Evaluated methods involved comparison of chromatograms of volatile compounds obtained utilizing solid-phase microextraction fast gas chromatography-flame ionization detector (SPME-fast GC-FID), analysis of volatiles by means of (a) a metal oxide semiconductor (MOS) based electronic nose (HS-Enose^{TM}), and (b) SPME-GC/MS, and determination using SPME-MS. The three tested methods permitted the detection of VOO adulteration with several amounts of hazelnut oil in the range between 5 and 50 % (v/v).

Two different e-noses have been employed to detect adulteration of extra virgin olive oil (EVOO) samples with sunflower and rapeseed oils (Mildner-Szkudlarz and Jeleń, 2010). As in the previously commented work, the proposed methods included determination of volatiles with HS-Enose^{TM} and solid-phase microextraction coupled to mass spectrometry, as well as SPME-GC/MS. EVOO samples were adulterated with different contents (between 5 and 50 % v/v) of several seed oils, patterns of volatile profiles of all samples being then obtained. Two goals were to be achieved: to get as much chemical information as possible and to find a volatile marker to detect EVOO adulterations; bearing them in mind, PCA and partial least square (PLS) analyses were applied to the corresponding data. This was enough to distinguish the adulterated samples from pure EVOO. Highly satisfactory results were achieved in the prediction of the adulteration degree using PLS analysis. They are even better than those provided by SPME-GC/MS analysis, and with the additional advantage of saving time. Therefore, and as a concluding remark, the two e-noses are straightforward with reliability and rapidity, and enable detection of extra virgin olive oil adulteration.

The evaluation of possible adulterations of sesame oil has also been the subject of study by means of an e-nose (Hai and Wang, 2006a). An array of 10 MOS sensors was utilized to obtain a smellprint of the volatile compounds occurring in the samples. Prior to several supervised chemometric analyses (linear discriminant analysis (LDA), probabilistic neural network (PNN), back-propagation artificial neural network (BP-ANN), and general regression neural network (GRNN)) of the data provided by the electronic nose, the following feature extraction techniques were employed to select a group of optimal discriminant variables: PCA, Fisher linear transformation (FLT), stepwise linear discriminant analysis (SLDA), and selection by Fisher weights (SFW). As for LDA and PNN, FLT turned out to be the best extraction method, whereas SLDA was more suitable for BP-ANN and FLT was more effective for GRNN. Outstanding results were achieved in the prediction of adulteration level in sesame oil by GRNN and BP-ANN, the latter being more precise in quantitative terms after an iterative training.

Another application field of the e-nose was the detection of maize oil adulteration in camellia seed oil and sesame oil (Hai and Wang, 2006b). Multivariate analysis of variance (MANOVA) was carried out and the results obtained showed that there are significant differences among the sensor signals of various types of oil. PCA could be applied to discriminate the adulteration of sesame oil, unlike in the discrimination of adulteration in camellia seed oil. Instead, LDA could be utilized to distinguish the adulteration of both types of oil. Canonical discriminant analysis (CDA) was also performed to test the discrimination ability of LDA, acceptable results being obtained (83.6 % accuracy prediction for camellia seed oil and 94.5 % for sesame oil). The artificial neural network (ANN) model was then utilized to determine the adulteration level in both types of oil, results being satisfactory for sesame oil but not for camellia seed oil.

Continuing with the field of edible oils, the zNose™ electronic nose was used to evaluate the adulteration levels of virgin coconut oil (Marina et al., 2009). This device was utilized to obtain a fingerprint of volatile compounds occurring in the oil samples. Virgin coconut oil was adulterated with different contents (ranging from 1 to 20 % w/w) of refined, bleached and deodorized palm kernel olein, the corresponding peaks being identified in the chromatogram and fitted to a curve using linear regression. The relationship between the peak initially identified as methyl dodecanoate and the percentage of palm kernel olein added gave rise to the best result (r = 0.95). On the other hand, correlation coefficient values of r = 0.92 and r =

0.89 were achieved between adulterant peak methyl dodecanoate and of the iodine and peroxide values, respectively. PCA was employed to discriminate between adulterated and pure samples; the results obtained were satisfactory, with 74 % of the variation accounted for by the first principal component and 17 % by the second principal component.

The literature clearly shows that microbial contamination can easily affect processed tomato; that is why there is a need for the determination of organoleptic adulterations in order to prevent potential health risks for consumers. Therefore, a fast and reliable detection of spoilage, for instance by using e-noses, is required to ensure food safety. In this context, in the work of (Concina *et al*., 2009), canned peeled tomato samples were adulterated with several types of microbial flora and later analyzed using a MOS-based electronic nose. Previous analyses carried out by dynamic-headspace GC/MS demonstrated the existence of significant differences in the semi-quantitative volatile compounds profile of adulterated tomatoes just after few hours from spoilage, which opens the windows to the possibility of utilizing the e-nose for an early detection of microbial presence (always depending on the kind of contaminant) as well as for recognizing spoiled tomato samples.

The detection of adulteration levels in tomato juices by means of e-noses has also been dealt with in a recent paper (Hong et al., 2014a), in which spectral clustering (a recent clustering method) is described and compared with six conventional clustering methods. Three external validation criteria – mutual information criteria (MI), precision, and rand index (RI) – were employed to evaluate clustering performances on three independent e-nose datasets (obtained from tomato juice analyses). The spectral clustering outperformed with statistical significance (alpha = 0.05) the performance of other methods, and the single linkage exhibited the worst (really unacceptable) clustering result. Furthermore, the proposed procedure (cluster validation criteria combined with majority voting) somewhat makes clustering a semi-supervised classification technique. This method enables the comparison clustering-based semi-supervised methods with classification methods to find which procedure is better for discrimination of a given e-nose dataset.

A common adulteration of honey takes place when sugar concentrate is added to this product; unfortunately, laboratory tests have proved so far to be ineffective in the detection of this fraud. A Chinese research team (Pei et al., 2015) has developed a method for rapid detection of Acacia honey adulteration using a FOX 4000™ e-nose. Samples were spiked with different amounts of rape honey and rice syrup, and the information (from the e-nose) on both natural and adulterated honey was analyzed by PCA. LDA was employed to study the ability of qualitative recognition of the e-nose for adulterated honey. The results showed that there was a linear relationship between e-nose signals and the adulteration level. On the other hand, the minimum amount of rape honey and rice syrup added leading to honey aroma system changes was 2 % and 1 %, respectively. Therefore, honey aroma system can easily be changed by adulterant compounds. This also demonstrated that the e-nose had a strong discriminable ability for honey adulteration. The results concluded that pure honey and adulterated honey can be distinguished by LDA pattern recognition algorithms, this fact resulting in a fast and accurate identification of honey adulteration.

An e-nose was also proposed for the detection and differentiation of lard from other kinds of animal fats as well as from foodstuffs containing lard (Nurjuliana et al., 2011). The results obtained are displayed in the form of the so-called *VaporPrint*. In this two-dimensional olfactory image, the radial angles representing the sensor yield individual patterns (smellprints) of the odor of different animal body fats. PCA was utilized to interpret the results achieved and gave rise to a satisfactory grouping of samples (61 % of the variation corresponded to the first principal component, and 29 % to the second principal component). All of the lard-containing samples formed a separate group from those having no lard. On the other hand, the ability to detect the presence of lard in food products helps Halal authentication (compliance with Islamic law).

The detection of potential adulteration in spices can be carried out by applying two different portable multi gas sensors (ion mobility spectrometer (IMS) and electronic nose) and multivariate data analysis (Banach et al., 2012). Headspace above spice mixtures for sausages and saveloy and product falsifications was analyzed making use of a MOS-based e-nose, discrimination being carried out by means of LDA of sensor resistivity data. Simultaneously, an IMS was coupled to the emission chamber to enable the detection of gaseous substances above the spice mixtures. PCA was then utilized to discuss the differences (between the two spice mixtures) provided by the obtained spectra. Both IMS and e-nose permitted to differentiate between the types of spice mixtures and subsequently to highlight product adulteration. Moreover, a headspace gas analysis (using gas chromatography) was carried out to identify major volatiles as well as to lay the chemical basis for the existing differences in the multi gas sensors.

The high cost of saffron inevitably (and unfortunately) leads to frequent attempts to adulterate it. That is why in a recent work (Heidarbeigi et al., 2014) the odor smellprints of saffron, saffron with yellow styles, safflower, and dyed corn stigma were recognized by an electronic nose. The characteristics of the obtained results were extracted and analyzed, PCA being used for this purpose. The corresponding data were then confirmed by BP-ANN, and showed that the e-nose is able to detect the saffron adulteration with excellent results. The authors conclude that this e-nose-based system could yield a good differentiation of the saffron and the adulterated one (100 and 86.9 % classification accuracy respectively) at adulteration levels over 10 % using ANN.

The determination of wine traceability and authenticity is also a critical issue to try to avoid illegal adulteration practices, namely: (i) addition of ethanol, flavoring and coloring compounds, (ii) dilution of wines with water, and (iii) mixing with, or replacement by, cheaper wine. In this sense, the utilization of e-noses along with multivariate statistical methods (mainly PCA, cluster analysis (CA), and SLDA) has led to better means for wine traceability, as well as discrimination and classification of grapes and wines (mainly in terms of grape varieties and geographical origin). Some of the recent advances on wine typification and authentication have been recently reviewed by (Versari *et al*., 2014), who also remark that several challenges need to be solved in order to improve the assessment of wine authenticity and confirm potential adulteration.

The remarkable popularity of whisky frequently involves a certain risk of adulteration. Therefore, authenticity assessment is a critical issue, and is usually carried out by comparing the composition of this alcoholic beverage with other spirits. The paper of (Wiśniewska et al., 2014) summarizes all information related to both the identification of and quality evaluation of whisky, and finally the detection of possible adulterations. In this field, the e-nose turns out to be one of the most promising analytical techniques in combination with the application of chemometric tools such as PCA, DFA, LDA, analysis of variance (ANOVA), soft independent modelling of class analogy (SIMCA), PNN, *k*-nearest neighbors (*k*-NN) and CA.

Yu et al. (2007) performed a study with a view of monitoring the adulteration of milk with water or reconstituted milk powder utilizing the PEN 2™ electronic nose with ten different MOS sensors. For this purpose, a series of experiments were conducted over 7 days of storage using three types of samples: whole fluid milk, reconstituted milk powder, and whole fluid milk adulterated with several amounts of water. The data obtained were a consequence of applying two chemometric techniques: PCA and LDA. According to the authors, the corresponding results proved that the artificial sense used (the e-nose) was able to differentiate the purity of milk samples when skimmed milk is diluted with variable amounts of water, and both LDA and PCA show a regular distribution of the results for the aforementioned three samples analyzed. Finally, with these two chemometric methods, the electronic nose could also differentiate between milk samples that had been storaged for different numbers of days.

The detection of the adulteration of mutton was achieved using classical procedures (pH and color evaluation) as well as an electronic nose to develop a model capable of detecting and estimating the adulteration of minced mutton with pork (Tian et al., 2013). An MOS-based e-nose was utilized to collect volatile compounds occurring in the samples. Feature extraction methods, PCA, loading analysis, and SLDA were used to obtain the optimum data matrix. Among the discriminant analysis methods employed to evaluate the results achieved, SLDA turned out to be the most effective procedure. Then CDA was utilized as PR technique for the authentication of meat. BP-ANN, PLS, and multiple linear regression (MLR) were used to build a model for estimation of the amount of pork in minced mutton, the best results being obtained with the model constructed by BP-ANN.

Li et al. (2014) developed a procedure for the fast identification of poultry meat species and detection of meat adulteration. For this purpose, they carefully examined the relationship between the heating temperature and the volatiles of duck, chicken, and goose meats. An electronic nose was then utilized for the detection of the different heating temperature of those poultry meats, with the help of LDA and DFA. The results obtained clearly show that this electronic device is able to distinguish between the different kinds of poultry meat, what leads the way in detecting the adulteration of meat products.

Two food adulteration cases (a pure variety of green coffee beans and pure cayenne with bell pepper powder) were studied by (Rodríguez et al., 2014) with the goal of reporting the improvements achieved in the discrimination of complex aroma samples with very small differences in odor pattern. For this purpose, they utilized a portable e-nose consisting of a sensor array which records changes in conductivity as a function of time when aroma molecules reach the sensors. The proposed method is then based on the application of unfolded cluster analysis to selected time windows within the temporal evolution of the aroma profile (recorded by the sensor array), providing an efficient, rapid, and reliable data analysis tool. The results obtained showed that this procedure enables to discriminate highly similar samples, thus decreasing the probability of a wrong grouping due to the use of doubtful data. The automation of this type of analysis is easy and enhances the efficiency of the e-nose in a significant way, what implies reducing the time of sensor's signal recording that is required for a reliable assessment of the studied system. The results were validated by clustering the sample component scores that are obtained by applying parallel factor analysis to the original three-dimensional data array.

It is quite clear to everyone that rice is the staple food for most Southeast Asian countries. In this sense, Jasmine rice is produced from varieties Khao Dowk Mali 105 and Kor Kho 15. Unfortunately, adulteration of Jasmine rice with other varieties such as Pathum Tani 1 and Chai Nat 1 is a common practice, as well as a major problem regarding Thailand rice export. To solve this problem, a Thai researcher (Masiri, 2006) has evaluated potential indices to adulteration of Jasmine rice with Pathum Tani 1 by using an e-nose consisting of two standard arrays of six MOS sensors. He found that PCA could classify adulteration of Jasmine rice efficiently, excellent results being obtained.

Tea (currently produced in nearly 50 countries around the world) is also an important target for fraudulent activities. Thus, methodologies to authenticate the geographical origin of tea and avoid incorrect labeling are becoming important tools to monitor illegal practices. On the other hand, the quality of the tea depends on the climate of the planting geographical areas as well as on the processing technique. In recent years there have been some attempts in the development of reliable methods for evaluating the quality of tea by chemical analysis (Cubero-Leon et al., 2014).

Kovács et al. (2010) used an electronic tongue, an electronic nose, and sensory panel assessment for geographical origin identification of Sri Lanka black teas. Five black tea samples from different regions and latitudes were studied. The electronic devices used were: the commercial α-Astree^{TM} e-tongue and the commercial e-nose NST3320^{TM} consisting of an array of twelve metal MOS sensors and ten MOSFET (MOS field effect transistor). It should be remarked that -in order to get a representative sample of the average tea consumer- panelists for sensory analysis were not specifically trained to tea. The corresponding data were analyzed by means of SLDA, PCA, and one-way ANOVA (in the case of sensory analysis). PLS regression was utilized to estimate the sensory attributes by the e-devices.

SLDA and PCA results obtained from e-nose data demonstrated that the device used did not perform very well in the discrimination of samples according to their geographical origin (success rates over 75 % and 37 % in training and cross-validation steps, respectively). Nevertheless, the e-nose exhibited an excellent ability to classify samples according to their growing altitude (100 % success rates in both calibration and cross-validation steps). On the other hand, e-nose data provided poor to moderate prediction of sensory attributes by PLS modeling ($0.59 \leq r \leq 0.89$ and $0.45 \leq r \leq 0.72$, in training and cross-validation steps, respectively). Finally, sensory analysis proved that an average tea consumer (without specialized and intensive training) can hardly differentiate Sri Lanka teas from different geographical origins. As regards the results obtained from the e-tongue data, they are described in the next section.

4. E-tongues in food authenticity/adulteration assessment

E-tongues are also emerging as promising supplemental techniques to classical analytical methods for a fast and low cost detection of malpractices. As shown in Table 2, e-tongues have shown their capability in the detection of food adulteration as well as in the authenticity assessment of different types of foodstuffs, such as honeys (Dias et al., 2008; Escriche et al., 2012; Garcia-Breijo et al., 2013; Major et al., 2011; Sousa et al., 2014; Wei and Wang, 2011; Wei et al., 2009), milk and dairy products (Dias et al., 2009; Paixão and Bertotti, 2009), alcoholic beverages (Gutiérrez et al., 2010; Gutiérrez-Capitán et al., 2013; Moreno-Codinachs et al., 2008; Novakowski et al., 2011; Parra et al., 2004; Parra et al., 2006; Pigani et al., 2008; Rodríguez-Méndez et al., 2008a; Rudnitskaya et al., 2007; Rudnitskaya et al., 2010), edible oils (Apetrei and Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et al., 2005; Apetrei et al., 2007; Apetrei, 2012; Dias et al., 2014; Oliveri et al., 2009; Rodríguez-Méndez et al., 2008b), and teas (Chen et al., 2008; He et al., 2009; Kovács et al., 2010).

In the last years, several papers have been published dealing with honey authentication by applying multivariate chemometric techniques to e-tongues data. These studies clearly point out that e-tongues can be utilized as efficient and practical tools to classify honeys according to their botanical (Dias et al., 2008; Escriche et al., 2012; Garcia-Breijo et al., 2013; Major et al., 2011; Sousa et al., 2014; Wei and Wang, 2011; Wei et al., 2009) and geographical origin (Wei et al., 2009).

A potentiometric e-tongue comprising 20 all-solid-state electrodes with polymeric membranes was developed for the discrimination of 52 honey samples with different pollen profiles by Dias et al. (2008). LDA and PCA were used for multivariate analysis of e-tongue data. Results indicated that the electronic device exhibited a good ability (84 % and 72 % classification accuracies in calibration and cross-validation, respectively) for classification of honeys as a function of the primary pollen type.

Wei et al. (2009) employed a commercial potentiometric e-tongue (α-Astree™) to classify honeys from different floral and geographical origins. PCA on e-tongue data obtained from 192 samples (same geographical origin) yielded full differentiation of all the eight monofloral origins studied; additionally, the position of the samples on the PCA score plots was related to the degree of ‘‘sweetness”. ANN and CA gave rise to satisfactory results as well (94 % and 90 % classification rates, respectively). On the other hand, PCA did not lead to complete differentiation of the five geographical origins of Acacia honeys tested, but the position of the samples on the PCA score plot could also be related to conductance. In this last case, CA and ANN provided good results (92 % and 95 % classification rates, respectively).

In a further work, the same research group (Wei and Wang, 2011) developed a voltammetric e-tongue -based on multifrequency large amplitude pulse voltammetry (MLAPV)- composed of six metallic working electrodes. The purpose was to discriminate 42 certified monofloral honeys of seven different floral origins. It was found that all honey samples were correctly classified by DFA, PCA, and CA on voltammetric data. Furthermore, the efficient working sensors and frequencies were selected for the further study. It must be noticed that the use of combined data from different working electrodes and different frequencies gave rise to much better classification ability of samples than that observed using data from an individual working electrode with a single frequency.

The aforementioned commercial e-tongue α-Astree™ has also been applied for botanical classification and prediction of physicochemical properties of 12 samples from three botanical origins (Major et al., 2011). PCA analysis of e-tongue data showed a very good clustering of samples according to their botanical origin. On the other hand, a 100 % accuracy was obtained in the botanical classification of honey samples using ANN. Satisfactory (r = 0.979 - 0.999) ANN models for the prediction of studied physicochemical properties (electrical conductivity) and content of different chemical parameters (acidity, water content, invert sugar, and total sugar) from the sensors outputs were obtained, which clearly highlights the potential of the e-tongue for a fast honey analysis.

Escriche et al. (2012) used another potentiometric e-tongue, comprising seven metals and metallic compounds, for quality control and authenticity assessment of honey. They resorted to PCA and ANN based on a Fuzzy ARTMAP algorithm and leave-one-out cross-validation to show the usefulness of the proposed e-tongue for discriminating samples according to their four botanical origins (ANN classification success exceeding 93 %), although it was not able to clearly differentiate among the three thermal treatments used. On the other hand, acceptable to good (r = 0.74 - 0.96) PLS correlations between the e-tongue data and seven physicochemical parameters were obtained. Nevertheless, poor (r = 0.49 - 0.73) PLS correlations between e-tongue data and volatiles concentrations were observed. In a further work (Garcia-Breijo et al., 2013), data were dealt with using ANN based on a simplified Fuzzy ARTMAP (SFA) and graphical user interface (GUI) for MATLAB® cross-validation. For botanical origin classification, 100 % recognition rates were obtained in the supervised phase, as well as when utilizing data of four selected electrodes. For thermal treatment of samples, up to 83 % recognition rate was achieved. In the non-supervised phase, a recognition rate of 69 % was obtained for four new test samples. This rate was increased up to 75 % by reducing the number of electrodes to four.

Another successful application of e-tongues for ensuring monofloral honey authenticity is the study reported by Sousa et al. (2014). A specifically designed potentiometric e-tongue consisting of two replicated groups of 20 all-solid-state sensors with different cross-sensitivity membranes was used. 65 Portuguese monofloral honey samples (harvested in different years) were studied. LDA based on the e-tongue signal profiles from 13 sensors -selected with a simulated annealing (SA) variable selection algorithm- resulted in the fact that 91 % (for both original data and leave-one-out cross-validation) of the honey samples were correctly classified according to three main color groups. For each of these color groups, all honeys were correctly classified according to their floral origin using a variable selection of e-tongue data combined with a LDA leave-one-out cross-validation strategy. However, LDA model using all samples data gave rise to a poor classification (in cross-validation) of honey samples according to their floral origin. The authors pointed out that the use of selected variables increased the accuracy performance of the LDA models, hence the importance of using variable selection algorithms in this type of studies. More recently, authors from this research group have published a book chapter focused on the applications of electrochemical sensors to evaluate antioxidant capacity of bee hives products (Peres et al., 2016).

Dairy products (especially milk) are frequently also subject to adulterations. In the paper by Dias et al. (2009), a potentiometric e-tongue with two units of all-solid-state polymeric membranes electrodes is proposed to detect raw goat milk adulterations with raw cow milk. LDA results showed that this e-tongue had an adequate ability (97% and 87 % classification rates in calibration and cross-validation, respectively) to evaluate the potential adulterations. Nevertheless, the authors claim that the sensitivity of the sensor array towards milk composition changes has to be improved before this device can be used as a routine tool.

Detection of milk adulteration has also been addressed by Paixão and Bertotti (2009). The authors developed disposable integrated voltammetric e-tongues composed of bare and Prussian Blue modified electrodes. PCA inspection of Au and Prussian Blue-modified gold electrodes data allowed to discriminate milk samples adulterated with H_2O_2 as well as to differentiate several pasteurization processes of samples. Disposable electrodes are of particular interest in milk analysis since adsorption onto the sensor surface of substances present in high concentrations in the sample matrix can alter the voltammetric signal.

Fraudulent practices are increasingly performed in several alcoholic beverages, most notably in wines. Both the quality control of wines and grape juices and the quantitation of different compounds have a paramount significance in wine production. The stages in the wine-making process have to be carefully monitored to control potential adulterations as well as to determine the concentration of some key components for the final quality of the product. In this context, e-tongues have proven their usefulness to authenticate geographical origin (Parra et al., 2004; Pigani et al., 2008), grape variety (Gutiérrez et al., 2010; Gutiérrez-Capitán et al., 2013; Moreno-Codinachs et al., 2008; Pigani et al., 2008; Rodríguez-Méndez et al., 2008a), vintage (Moreno-Codinachs et al., 2008) and age (Parra et al., 2004; Rudnitskaya et al., 2007; Rudnitskaya et al., 2010) of wines or grape juices, and to detect adulterated wines and whiskeys (Novakowski et al., 2011; Parra et al., 2006).

In this framework, Parra et al. (2004) developed a voltammetric sensor array composed of rare-earth bisphthalocyanine carbon paste electrodes (CPEs) for the discrimination of six Spanish red wines from the same variety of grape but from three different origin designations and ageing stages. PCA applied to e-tongue data showed a good discrimination of the tested samples. The discrimination ability of the e-tongue was similar to that obtained using eight chemical variables coming from chemical analysis. In a further work (Rodríguez-Mendez et al., 2008a), some authors from the same research group built an e-tongue consisting of CPEs modified with rare-earth bisphthalocyanines and perylenes. PCA study of data obtained for six white wines demonstrated the e-tongue ability to discriminate among the grape varieties utilized for sample preparation.

The effectiveness of three different poly(3,4-ethylenedioxythiophene) (PEDOT) conducting polymer modified electrodes to analyze similar matrices (six white wines of different grape varieties and geographical areas) was tested by Pigani et al. (2008). Voltammetric responses of each sensor separately and the combined responses of two or three sensors were used in the chemometric analysis. Partial least square-discriminant analysis (PLS-DA) on voltammetric data proved that all the electrodes could successfully classify (nearly 100 % correct classifications) samples according to their variety.

An integrated potentiometric multisensor composed of six ion-selective field effect transistors (ISFETs) and a flow injection analysis (FIA) system has been applied to grape juice and wine analysis (Moreno-Codinachs et al., 2008). PCA and SIMCA results demonstrated the ability of the e-tongue under batch conditions for differentiating four grape varieties in grape juices. Furthermore, PCA and SIMCA on FIA/e-tongue data allowed for the discrimination of wine samples (two different groups of wines) according to the grape variety and the vintage year. Finally, PLS regression of grape juice and wine samples data showed that those devices were able to quantify diverse physicochemical parameters and concentrations of different sample components (prediction errors under 10 %) with good correlations with traditional analytical methods ($r > 0.99$). The miniaturization of the flow cell is possible due to the use of an integrated multisensor built with the help of microelectronic technology.

In the paper by Gutiérrez et al. (2010), the capability of a hybrid e-tongue for the classification of wines according to the grape varieties and vintage as well as for the prediction of some chemical and optical parameters of interest in wine quality control is demonstrated. The proposed e-tongue included an array of electrochemical microsensors and a colorimetric optofluidic system. Both of them can be integrated into the same platform, thus providing portable, rapid, and feasible equipment for *in-situ* measurements. E-tongue data obtained for red (samples from the same vintage but different grape varieties) and white (two vintages and four grape varieties) wines were processed by means of PLS and PCA. In spite of the small number of samples, the potential of the developed e-tongue in this kind of studies is clearly demonstrated.

In a later work (Gutiérrez-Capitán et al., 2013), some authors from the aforementioned research group used a similar hybrid e-tongue for the analysis of white grape juices (different white *Vitis* genotypes with comparable characteristics). A PCA model from e-tongue data of samples belonging to three reference genotypes was used to estimate some basic properties of the other varieties. On the other hand, SIMCA method on the reference genotypes allowed these reference varieties and the other samples to be differentiated. The authors claimed that this e-tongue could be used for adulteration detection in wines since the results for a non-*vinifera* genotype (a hybrid prohibited in several European countries for wine production) appeared as outliers in the PCA and SIMCA models. Moreover, this electronic device could also be of great utility in the wine production process to estimate grape juice properties.

In the same way as other wines with controlled origin, the Portuguese Port and Madeira wines are affected by frauds, so they require strict quality and authenticity control. There is a considerable interest in the development of fast methods for the age estimation of these wines, since their quality and price are directly proportional to their age. In this sense, a potentiometric e-tongue has shown similar ability to conventional analytical techniques to predict the age of a great deal of Port wine samples of different types and ages (PLS prediction accuracy under 6 years) (Rutnitskaya et al., 2007). Orthogonal signal correction (OSC) was utilized as data pre-processing strategy to eliminate the temporary drift in e-tongue responses. An analogous e-tongue has also been applied in the case of Madeira wine age estimation and determination of the concentration of several components, mainly phenolic compounds and organic acids (Rutnitskaya et al., 2010).

With respect to the use of e-tongues to detect adulteration of alcoholic drinks, Parra et al. (2006) proposed a voltammetric e-tongue composed of two kinds of sensors (phthalocyanine-based CPEs and conducting polypyrrole polymers based electrodes) for the detection of the most common adulterants of red wines. The e-tongue data (PLS regression) yielded good estimations (prediction errors between 1 and 15 %) of the concentrations of seven chemical compounds (common adulterants) added to the samples. Additionally, PCA on e-tongue data exhibited a good performance for distinguishing the different kinds of adulterations. The inclusion of those two types of sensors in the e-tongue increased their discrimination ability.

Nowakowski et al. (2011) made use of a disposable, integrated voltammetric e-tongue fabricated using gold and copper substrates to classify wine and whisky samples. In case of wines, PCA inspection of e-tongue data revealed that this electronic instrument was able to differentiate four types of wine from the same brand. When different types and brands of wines were included in the study, PCA score plot of e-tongue data showed an almost complete clustering of the different samples. On the other hand, PCA models applied to data obtained from a single copper electrode allowed to distinguish not only several whisky brands but also different adulteration processes of the whiskies. The authors highlight the fact that the combination of an extensive database of whiskies and wines coupled with the developed disposable system could be helpful in forensic analysis to detect unidentified or adulterated samples.

Authenticity assessment of vegetable oils is another research area of interest for e-tongues (Apetrei and Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et al., 2005; Apetrei et al., 2007; Apetrei, 2012; Dias et al., 2014; Oliveri et al., 2009; Rodríguez-Méndez et al., 2008b). The high price of EVOOs and VOOs make them ideal “candidates” for adulteration, be it mislabeling or blending with cheaper olive and seed oils. Some VOOs and EVOOs are certified as protected designation of origin, which are partly related to olive oil production and processing made in a specific geographical origin. Furthermore, label authentication of olive cultivar is of paramount importance due to marketing of high-quality (and high-price) monovarietal EVOOs. Label information about the geographical origin affects consumers’ acceptability, while information about cultivar significantly impacts on the expectation of pungency and bitterness for olive oils. As shown in Table 2, papers reported in the literature on this topic have demonstrated the effectiveness of e-tongues for the discrimination among different vegetable oils (Apetrei et al., 2005; Apetrei and Apetrei, 2014; Oliveri et al., 2009), quality grades of olive oils (Apetrei et al., 2005), olive cultivars (Dias et al., 2014) and geographical origin of EVOOs (Oliveri et al., 2009). E-tongues have also shown their ability for quantitative prediction of different physicochemical parameters utilized for EVOOs characterization (Apetrei and Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et al., 2007; Apetrei, 2012; Rodríguez-Méndez et al., 2008b), as well as to detect EVOOs adulterations with seed oils (Apetrei and Apetrei, 2014). Finally, Rodríguez-Méndez et al. (2010) have authored a chapter book including some applications of e-tongues purposely designed for the characterization of olive oils.

The analysis of oils using electrochemical methods is rather troublesome due to the lack of conductivity, the viscosity, and the low solubility in the standard solvents employed for oil samples. To overcome this difficulty, the research group of Apetrei and co-workers has tested the usefulness of chemically modified CPEs with the oil sample (Apetrei et al., 2005; Apetrei et al., 2007; Apetrei and Apetrei, 2014). In this case, voltammetric data obtained when immersing the oil-based electrodes in different electrolytic solutions are used in the chemometric analyses. In the first work of the series (Apetrei et al., 2005), the study of the electrochemical data by means of PCA provided a clear discrimination among vegetable oils of three diverse origins and among olive oils of five different quality grades. A further paper has proven the usefulness of these oil modified CPEs for the evaluation of bitterness of EVOOs (Apetrei et al., 2007).

In a more recent work (Apetrei and Apetrei, 2014), the usefulness of this type of e-tongues to detect EVOOs adulteration is demonstrated. PCA, PLS-DA and PLS applied to e-tongue data showed the capability of this device (a) to discriminate pure oils (an EVOO and three seed oils) according to their botanical origins; (b) to predict total polyphenolic content of binary mixtures of EVOO and seed oils (*r* values near 0.99, in both PLS calibration and validation); (c) to classify the adulterated EVOOs when the concentration of adulterant oil was over 5 %; and (d) to estimate the composition of EVOO and seed oil mixtures within the range 2 – 25 % range ($r > 0.99$ in both PLS calibration and validation).

The research group of Rodríguez-Méndez et al. (2008b) developed a voltammetric e-tongue consisting of chemically modified electrodes with several electroactive materials (phthalocyanine derivatives and polypyrroles doped with different doping substances) to evaluate the phenolic content of six EVOOs. Sample pretreatment involved dissolution in hexane, extraction of the phenolic fraction with a methanol-water mixture and drying and redissolution of the extract in potassium chloride aqueous solution. High correlation coefficient values ($r > 0.99$) for the PLS regression models between the e-tongue data and several parameters (bitterness index, polyphenol content, and bitterness degree) were achieved.

Voltammetric e-tongues based on polypyrrole sensors have also been proposed for the authenticity assessment of EVOOs (Apetrei, 2012; Apetrei and Apetrei, 2013), with a previous preparation of oil emulsions in surfactants. PCA and

PLS-DA exploration of the data obtained from such systems demonstrated their ability to discriminate six Spanish EVOOs according to their sensorial and chemical bitterness, as well as to predict bitterness degree and chemical parameters such as peroxide value, free acidity, bitterness index, and K indexes (r > 0.87 in both, calibration and validation of the PLS models) (Apetrei, 2012). On the other hand, classification of 18 EVOOs according to their total polyphenolic content was successfully accomplished using PCA, PLS-DA and SIMCA (Apetrei and Apetrei, 2013). A good PLS model for the quantification of this chemical parameter was obtained (r > 0.98 for both the training and the test set). Furthermore, e-tongue data for spiked EVOO emulsion samples having different individual phenolic compounds permitted PCA discrimination and an error-free PLS-DA classification of these substances.

In another attempt to solve the aforementioned problem of electrochemical analysis of oils, Oliveri et al., (2009) recommended the use of suitable room temperature ionic liquids, added to oils as supporting electrolytes to provide these low-polarity samples with conductivity. The following step consisted of voltammetric measurements with a platinum microelectrode, which were carried out directly in those edible oil/ionic liquid mixtures. Voltammetric data were then processed by means of PCA and a *k*-NN classification method, and showed that EVOOs having different nature (maize and olive) or geographical origin (from different Italian and Spanish regions) can be differentiated.

As commented on previously, label authentication of monovarietal EVOOs is of primary importance. In the paper by Dias et al. (2014), a potentiometric e-tongue with different cross-sensitivity membrane sensors was fabricated to discriminate 18 Portuguese and Spanish monovarietal EVOOs according to the olive cultivar. The e-tongue is analogous to that used for honey classification (Sousa et al., 2014). In a similar way to that paper, the most informative potentiometric sensor signal profiles were selected using an SA algorithm to establish LDA models with the best leave-one-out cross-validation predictive performance. Hydro-ethanolic extracts of EVOOs were utilized to solve the problem of electrochemical assays in oils. E-tongue data gave rise to outstanding classification results according to the olive cultivar for EVOOs of each country (100 % and 97.5-100 % correct classification for the original data and for cross-validation, respectively). Notwithstanding, no simultaneous discrimination of all the six Spanish and Portuguese cultivars could be achieved (92 % and 43 % correct classification for original data and cross-validation, respectively).

The performance of the e-device to differentiate each Spanish cultivar from the three Portuguese cultivars was satisfactory to poor (89–100 % and 61-98 % of correct classifications for the original data and cross-validation, respectively). The discriminant ability was related to the polar compound contents of EVOOs and therefore, indirectly, to organoleptic properties. This last issue was addressed in a further work (Veloso et al., 2016), a similar e-tongue being then used for analyzing the hydro-ethanolic extracts of a great number of EVOOS from eleven different cultivars and two crop years. LDA-SA applied to e-tongue data yielded a good discrimination of samples of each crop year according to their overall intensity perception levels (100 %, 91 % and ~ 80 % of correct classifications for the original data, leave-one-out and K-fold cross-validation, respectively). Consequently, the authors of this paper proposed their e-tongue as an auxiliary tool for trained sensory panels.

As previously remarked in section 3, new methodologies to authenticate the geographical origin and quality of tea as well as to avoid incorrect labeling are becoming important tools to monitor frauds and other illegal practices. In this framework, e-tongues have been employed for the recognition of tea grade level (Chen et al., 2008; He et al., 2009) and for geographical origin authentication of tea samples (Kovács et al., 2010; He et al., 2009).

Chen et al. (2008) utilized the commercial α-Astree^{TM} e-tongue to identify Chinese green tea grade level. A large group of samples belonging to four different grades were investigated and divided into training and test sets to build identification models. BP-ANN modeling resulted in higher identification rates (100 % in both the training and the test set) than *k*-NN modeling (97.5 % and 100 % for the training and the test set, respectively).

The α-Astree^{TM} e-tongue was also used by He et al. (2009) for the differentiation of Chinese tea. Eight samples of green tea and eight samples of black tea were studied. Different PCA models demonstrated the ability of the e-tongue data to discriminate between black and green teas, as well as the geographical origins of green or black teas. Moreover, the e-tongue sensors that were best correlated with ten sensory attributes in tea taste were determined.

As commented in section 3, the research goal of Kovács et al. (2010) was to evaluate the possible application of both the NST3320^{TM} e-nose and the α-Astree^{TM} e-tongue for geographical origin identification of Sri Lanka black tea. PCA and SLDA results indicated that the e-tongue showed a much better performance (100 %

success rates in both training and cross-validation steps) than the e-nose for sample discrimination according to their geographical origin. Nevertheless, the e-nose exhibited a slightly better ability to classify samples according to their growing altitude (e-tongue success did not reach 100 % in both training and cross-validation steps). On the other hand, these two e-devices provided poor to acceptable PLS prediction of sensory attributes, although the e-tongue performance was slightly better ($0.65 \leq r \leq 0.92$ and $0.47 \leq r \leq 0.86$, in training and cross-validation steps, respectively) than that of the e-nose.

Finally, another important issue in which e-tongues have proven their usefulness is the detection of possible contaminations of foodstuffs with gliandins, proteins primarily responsible for gluten intolerance (Peres et al., 2011). In this work, a potentiometric e-tongue comprising 36 polymeric membranes was used. This device is similar to that previously commented in Peres et al., 2009. 15 samples from five different kinds of foodstuffs and two different gluten levels (gluten-free and gluten-containing samples) as well as a gluten-free sample contaminated with different amounts of gliandins (gluten-free, low-gluten content and gluten-containing samples) were analyzed by means of the e-tongue system. A stepwise multivariate technique and LDA were used for variables/sensors selection/reduction and for samples classification into the studied gluten levels, respectively. Leave-one-out cross-validation classification results of LDA models on selected sensors data were satisfactory (84 % and 77 % success rates of uncontaminated and contaminated samples, respectively).

5. E-noses and e-tongues data fusion in food authenticity/adulteration assessment

In some applications, and due to the high complexity of food samples, the employment of just e-nose or e-tongue data is insufficient, and multisensor data fusion techniques, e.g. combination of e-nose with e-tongue and/or spectroscopic data, have been utilized as efficient characterization methodologies. Nevertheless, in most cases, a variable selection seems to be imperative for the application of sensor array data -particularly when a data fusion strategy is used- in order to remove response variables or sensors that are redundant, noisy, or irrelevant for qualitative or quantitative purposes. In this sense, a very interesting review about analytical techniques and strategies employed in data fusion methodologies for food and beverage authentication and quality assessment has recently been published (Borràs *et al.*, 2015). The present section is then just focused on applications based on the joint use of e-noses and e-tongues. The main features of such studies are summarized in Table 3 and are briefly described below.

In the literature there are some examples in which multisensor data fusion techniques have given rise to enhanced results for honey analysis (Maamor et al., 2014; Subari et al., 2012; Ulloa et al., 2013; Zakaria et al., 2011). The combination of data obtained from both an e-nose and an e-tongue to discriminate between different honeys, sugar syrups, and sugar adulterated honey samples has also been proposed (Zakaria et al., 2011). Samples were analyzed by means of the Cyranose320^{TM} e-nose (32 non-selective sensors of different types of polymer matrix, blended with carbon black), as well as by a potentiometric e-tongue consisting of seven chalcogenide-based ion selective electrodes (ISEs). By combining the data obtained from both e-systems, the discrimination ability for all the analyzed samples was significantly enhanced. The best results turned out to be those using LDA (100 % success rates in both training and cross-validation steps).

It is also worth reporting the comparison between data obtained from single modality and fusion methods in the classification of pure or adulterated Tualang honeys (Subari et al., 2012). Ten different brands of certified pure Tualang honey from Malaysia and Sumatra were blended with different concentrations of cane and beet sugar solutions and analyzed by means of the Cyranose320^{TM} e-nose and Fourier transform infrared spectroscopy (FTIR). The best classification rate (92.2 % in validation) was obtained when using normalized low-level FTIR and e-nose fusion data by means of SLDA. In a further work Maamor et al. (2014) (authors from the same research group) demonstrated that the discrimination ability between pure and sugar-adulterated Tualang honeys could be improved by utilizing combined data from FTIR, an e-nose, and a potentiometric e-tongue. PCA was then used to reduce high dimensional features of these three techniques. Among the different classification methods studied, *k*-NN provided the best performance for the training and test sets (100 and 96.4 % correct classification, respectively).

Ulloa et al. (2013) have tried to classify four commercial brands of Portuguese honey according to their botanical origin by means of sensor fusion of an impedance e-tongue and visible–near infrared (Vis–NIR) and ultraviolet–visible (UV–Vis) spectroscopies assisted by PCA and CA. 13 heterogeneous Portuguese honey nectar samples were analyzed. Different chemometric tools showed that fusion of the corresponding data obtained yielded a better discrimination ability than that achieved with the three individual techniques. In this sense, multi-way PCA (MPCA) proved to be an excellent (100 % classification success) alternative for data fusion, unlike simple concatenation of all matrices. Last but not least, a variable selection method based on one-dimensional clustering was developed to define two new strategies, both of them giving rise to even better defined sample clusters. Notwithstanding, the authors make it clear that the aim of the work was the demonstration of the proposed methods and there is clearly a need for further research work with a larger number of samples.

Multisensor data fusion techniques have also demonstrated to be of great utility for the analysis of some types of olive oils (Apetrei et al., 2010; Haddi et al., 2013). In the paper of Apetrei et al. (2010), the so-called electronic panel (data fusion of three systems, namely: an e-nose, an e-tongue, and an e-eye) was employed to characterize the organoleptic properties of 25 EVOOs from three different olive varieties. The e-nose consisted of a set of 13 MOS sensors whereas the e-tongue was based on modified CPE voltammetric sensors. PCA and PLS-DA of data showed that the combined system had a higher capability of sample discrimination according to the olive variety than that obtained with the three instruments used separately. Finally, PLS regression models provided good correlation coefficients between e-tongue data and bitterness scores (PLS1, $r > 0.97$ in both model calibration and cross-validation) as well as between the electronic panel data and the concentrations of 20 polyphenolic compounds (PLS2, $r > 0.9$ in calibration and cross-validation).

E-tongue and e-nose data fusion also gave rise to a better performance than the independent e-devices in the characterization of Moroccan VOOs (Haddi et al., 2013). A certain number of VOOs (from the same variety and harvested in the same year) from five different regions of Morocco were analyzed using a voltammetric e-tongue and a MOS-based e-nose. PCA and CA applied to a reduced subset containing optimal variables (selected with the help of a recently developed variable selection strategy based on ANOVA) improved the classification of the VOOs with respect to the use of all the variables. Support vector machines (SVM) performed on the reduced subset confirmed the correct identification of all VOOs.

The so-called biosensor-based multisensorial system for mimicking nose, tongue and eyes (BIONOTE) developed by Santonico et al. (2015) has proven excellent results for detecting EVOOs adulteration. This device includes gas and liquid sensors based on anthocyanins sensing interfaces. Quartz micro balances (QMBs) were used as transducers for the gas sensor array, while the sensor liquid array was composed of screen-printed gold voltammetric electrodes. Data fusion of sensors data allowed the discrimination of twelve EVOOs from different cultivars and geographical origins, the detection of adulteration of EVOOs with other four vegetable oils up to concentrations lower than 5 % as well as the prediction of common chemical parameters related to the quality of the EVOOs.

It should also be remarked that, according to Cosio et al. (2006), data from four selected e-nose sensors provided better results in the verification of the geographical origin of EVOOs than those from the fusion of chemical variables or an e-tongue. In this study, a commercial e-nose (model 3320 Applied Sensor Lab Emission Analyzer) based on 22 MOS and MOSFET sensors was utilized for the analyses, along with an amperometric e-tongue. The group of analyzed samples included 36 Garda (Italy) oils and 17 oils from other regions.

To conclude, in the literature there are also some examples in which multisensor data fusion techniques have proved to be useful for recognition and quantitative analysis of fresh cherry tomato juices adulterated with different levels of overripe tomato juices (Hong et al., 2014b; Hong and Wang, 2014). In these papers, the PEN 2^{TM} commercial e-nose and the α-Astree^{TM} commercial e-tongue were utilized for sample analysis. Two e-nose measurements (with and without desiccant) were carried out, and the corresponding results indicated that there is no need to use a desiccant prior to e-nose measurement. Several fusion procedures of e-nose and e-tongue data were tested. This work showed that simultaneous utilization of both instruments could guarantee a better performance provided that proper data fusion approaches are used.

6. Conclusions

In this review, the most relevant applications of e-noses and e-tongues in food authenticity assessment –in many cases leading to the detection of food adulteration- have been examined. This subject area is particularly (and increasingly) important in these last years, since it not only concerns the constant fight with food adulterers, but also relatively new aspects such as bioterrorism or food security. All these problems clearly highlight the need for further development and refinement of the existing analytical techniques. In this sense, the use of “artificial senses” such as those discussed in the present paper will undoubtedly contribute to overcome the shortcomings of other analytical techniques still in use. Furthermore, future developments in the use of advanced sensors arrays will lead to superior electronic senses with more capabilities, thus making the authenticity and falsification assessment of food products a faster and more reliable process.

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Abbreviations

- **ANN:** artificial neural network
- **ANOVA:** analysis of variance
- **ASCA:** ANOVA-simultaneous component analysis
- **BIONOTE:** biosensor-based multisensorial system
- **BP-ANN:** back propagation-artificial neural network
- **CA:** cluster analysis
- **CCA:** canonical correlation analysis
- **CDA:** canonical discriminant analysis
- **CPE:** carbon paste electrode
- **DFA:** discriminant function analysis
- **EDA:** exploratory data analysis
- **e-nose:** electronic nose
- **e-tongue:** electronic tongue
- **EVOO:** extra virgin olive oil
- **FIA:** flow injection analysis
- **FID**: flame ionization detector
- **FLT:** Fisher linear transformation
- **FTIR:** Fourier transform infrared spectroscopy
- **GC/MS:** gas chromatography-mass spectrometry
- **GC:** gas chromatography
- **GRNN:** general regression neural network
- **GUI:** graphical user interface
- **IMS:** ion mobility spectrometer
- **ISE:** ion-selective electrode
- **ISFET:** ion-selective field effect transistor
- ***k*-NN:** *k*-nearest neighbors
- **LDA:** linear discriminant analysis
- **LDA-SA:** linear discriminant analysis-simulated annealing
- **Lib-SVM:** support Vector Machines
- **MANOVA:** multivariate analysis of variance
- **MI:** mutual information criteria
- **MLAPV:** multifrequency large amplitude pulse voltammetry
- **MLR:** multiple linear regression

- **MOS:** metal oxide semiconductors
- **MOSFET:** metal oxide semiconductor field effect transistor
- **MPCA:** Multi-way PCA
- **MS:** mass spectrometry
- **OSC:** orthogonal signal correction
- **PCA:** principal component analysis
- **PCR:** principal component regression
- **PEDOT:** poly(3,4-ethylenedioxythiophene)
- **PLS:** partial least square
- **PLS-DA:** partial least square-discriminant analysis
- **PNN:** probabilistic neural network
- **PVC:** poly(vinyl chloride)
- **QMB:** quartz micro balance
- **RI:** rand index
- **SA:** simulated annealing
- **SAW:** surface acoustic wave
- **SFA:** simplified Fuzzy ARTMAP
- **SFA-ANN:** simplified Fuzzy ARTMAP-artificial neural network
- **SFW**: selection by Fisher weights
- **SIMCA:** soft independent modelling of class analogy
- **SLDA:** stepwise linear discriminant analysis
- **SPME:** solid-phase microextraction
- **SPME-fast GC-FID:** solid-phase microextraction fast gas chromatography-flame ionization detector
- **SPME-GC/MS:** solid-phase microextraction - gas chromatography/mass spectrometry
- **SPME-MS:** solid-phase microextraction-mass spectrometry.
- **SVM:** support vector machine
- **UV–Vis:** ultraviolet–visible
- **Vis–NIR:** visible-near infrared
- **VOO:** virgin olive oil

Table 1
Applications of e-noses in food authenticity/adulteration assessment.

Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
Virgin olive oil	Detection of adulteration with hazelnut oil	MOS sensors	PCA	Mildner-Szkudlarz and Jeleń (2008)
Extra virgin olive oil	Detection of adulteration with rapeseed and sunflower oils	MOS sensors and SPME-MS	PCA, PLS	Mildner-Szkudlarz and Jeleń (2010)
Sesame oil	Detection of adulteration	10 MOS sensors	LDA, PNN, BP-ANN, GRNN	Hai and Wang (2006a)
Camelia seed and sesame oil	Detection of adulteration with maize oil	MOS sensors	LDA, ANN, CDA	Hai and Wang (2006b)
Virgin coconut oil	Detection of adulteration	zNose™ (SAW)	PCA	Marina et al. (2010)
Canned tomato	Detection of adulteration	EOS835™ e-nose	PCA, EDA, k-NN	Concina et al. (2009)
Tomato juice	Detection of adulteration	MOS sensors	Spectral clustering	Hong et al. (2014a)
Acacia honey	Detection of adulteration with rape honey and rice syrup	FOX 4000™ e-nose	PCA, LDA	Pei et al. (2015)
Palm olein	Detection of adulteration with lard	zNose™ (SAW)	PCA	Nurjuliana et al. (2011)
Spices	Detection of adulteration	Portable multi gas sensors	PCA, LDA	Banach et al. (2012)
Saffron	Detection of adulteration with yellow styles, safflower, and dyed corn	MOS sensors	PCA, BP-ANN, ANN	Heidarbeigi et al. (2015)
Wine	Authenticity assessment	MOS sensors	PCA, CA, SLDA	Versari et al. (2014)
Whisky	Authenticity assessment	Predominantly MOS sensors	PCA, DFA, LDA, ANOVA, SIMCA, PNN, *k*-NN, CA	Wiśniewska et al. (2014)
Milk	Detection of adulteration with water and milk powder	PEN 2™ e-nose (10 MOS sensors)	PCA, LDA	Yu et al. (2007)
Mutton	Detection of adulteration with pork	MOS sensors	PCA, SLDA, CDA	Tian et al. (2013)
Poultry meats	Detection of adulteration	MOS sensors	LDA, DFA	Li et al. (2014)
Coffee and pepper	Detection of adulteration	Portable e-nose	Unfolded CA	Rodríguez et al. (2014)
Jasmine rice	Detection of adulteration with other varieties	Two standard arrays of six MOS sensors	PCA	Masiri (2006)
Black tea infusions	Discrimination between geographical origins. Discrimination between growing altitudes. Prediction of sensory attributes	NST3320™ e-nose (10 MOSFET and 12 MOS sensors)	PCA, SLDA, ANOVA, PLS	Kovács et al. (2010)

Acronyms used: ANOVA, Analysis of variance; ANN, Artificial neural network; BP-ANN, Back propagation-artificial neural network; CA, Cluster analysis; CDA, Canonical discriminant analysis; DFA, Discriminant function analysis; EDA, Exploratory data analysis; GRNN, General regression neural network; *k*-NN, *k*-Nearest neighbor; LDA, Linear discriminant analysis; MOS, Metal oxide semiconductor; MOSFET, metal oxide semiconductor field effect transistor; PCA, Principal component analysis; PCR, Principal component regression; PLS, Partial least square; PNN, Probabilistic neural network; SAW, surface acoustic wave; SIMCA, Soft independent modelling class analogy; SLDA, Stepwise linear discriminant analysis.

Table 2
Applications of e-tongues in food authenticity/adulteration assessment.

Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
Honey	Discrimination between samples accordingly to the most predominant pollen type	20 all-solid-state electrodes with PVC polymeric membranes applied on solid conducting silver-epoxy supports	PCA, LDA	Dias et al. (2008)
Honey	Discrimination between samples from different monofloral origin. Discrimination between samples from different geographical origin	α-Astree™ e-tongue (7 ISFETs based on polymer membranes)	PCA, ANN, CA	Wei et al. (2009)
Honey	Discrimination between samples from different monofloral origin	6 metal wires electrodes (Au, Ag, Pt, Pd, W and Ti)	PCA, DFA, CA	Wei and Wang (2011)
Honey	Discrimination between samples from different botanical origin. Prediction of physicochemical parameters	α-Astree™ e-tongue (see further details in Wei et al. (2009))	PCA, CCA, ANN	Major et al. (2011)
Honey	Discrimination between samples from different botanical origin. Prediction of physicochemical parameters	7 metallic wire electrodes (Au, Ag, Cu, Ag_2O, AgCl, Ag_2CO_3 and Cu_2O)	PCA, ANN, PLS	Escriche et al. (2012)
Honey	Discrimination between samples from different botanical origin and with different thermal treatments	The same as in Escriche et al. (2012)	SFA-ANN	Garcia-Breijo et al. (2013)
Honey	Discrimination between colors of monofloral honey samples. For honeys of the same color group: discrimination of monofloral honeys according to their floral origin	2 units of 20 all-solid-state electrodes with different pre-established mass combinations of 4 lipidic, 5 plasticizers and PVC high molecular weight polymers	LDA-SA	Sousa et al. (2014)
Goat milk	Detection of adulteration of samples with cow milk	2 units of 20 all-solid-state electrodes with PVC polymeric membranes, applied on solid conducting silver-epoxy supports	LDA	Dias et al. (2009)
Milk	Discrimination of samples adulterated with hydrogen peroxide. Discrimination between samples with different pasteurization process	Au and Prussian Blue-modified gold electrodes	PCA	Paixão and Bertotti (2009)
Red wine	Discrimination between origin denominations and between ageing stages	CPEs modified with 3 rare-earth bisphthalocyanines	PCA, kernel variable reduction	Parra et al. (2004)
White wine	Discrimination between grape varieties	CPEs modified with 3 rare-earth bisphthalocyanines and 3 perylenes	PCA, kernel variable reduction	Rodríguez-Méndez et al. (2008a)
White wine	Discrimination of grape varieties and geographical origins	1 PEDOT conducting polymer, composite materials of Au and Pt nanoparticles embedded in a PEDOT layer	PCA, PLS-DA	Pigani et al. (2008)
Grape juice and wine	Discrimination between grape varieties. Discrimination between vintages. Determination of different parameters and components	6 ISFETs based on polymeric membranes and chalcogenide glass membranes	PCA, SIMCA, PLS	Moreno-Codinachs et al. (2008)
White and red wine	Discrimination between grape varieties. Prediction of chemical and optical parameters	Hybrid electrochemical-optical e-tongue: - Electrochemical sensors: 6 ISFET potentiometric	PCA, PLS	Gutiérrez et al. (2010)

Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
		sensors, 1 conductivity sensor, a redox potential sensor and 2 amperometric sensors - Colorimetric optofluidic system: MIR configuration, the optical fiber was connected to a spectrophotometer that covered the 200-1100 nm range		
White grape juice	Discrimination between grape varieties	Hybrid electrochemical-optical e-tongue similar to those used in Gutiérrez et al. (2010) but including 7 ISFET sensors	PCA, SIMCA	Gutiérrez-Capitán et al. (2013)
Wine	Prediction of wine age	27 plasticized PVC and chalcogenide glass sensors and 1 glass pH electrode	PCA, OSC, PLS	Rudnitskaya et al. (2007)
Wine	Prediction of wine age. Determination of organic acids and phenolic compounds	25 plasticized PVC and chalcogenide glass sensors and 1 glass pH electrode	PCA, PLS, ASCA	Rudnitskaya et al. (2010)
Red wine	Correlation with chemical parameters. Discrimination between chemical adulterants	3 CPEs modified with phthalocyanines, 6 polypyrrole conducting polymers and 1 bare CPE	PLS, PCA	Parra et al. (2006)
Wine and whisky	Discrimination between different brands and types of wines. Discrimination between different brands of whisky. Discrimination between non-adulterated and adulterated whiskies	Au and Cu electrodes	PCA	Novakowski et al. (2011)
Vegetable oils	Discrimination between olive oils of different qualities and discrimination between different vegetable oils	CPEs modified with 6 vegetable oils	PCA, kernell variable reduction	Apetrei et al. (2005)
Extra virgin olive oil	Discrimination between samples of different bitterness degree. Prediction of sensorial bitterness degree obtained by a panel of experts. Prediction of chemical parameters (bitterness index, peroxide index, K indexes and stability)	CPEs modified with 9 olive oils	PCA, PLS-DA, PLS, kernell variable reduction	Apetrei et al. (2007)
Vegetable oils	Discrimination between different vegetable oils of different nature. Prediction of total polyphenolic content. Discrimination between pure and adulterated oils. Prediction of the composition of seed oils and extra virgin olive oil mixtures	CPEs modified with each edible oil studied	PCA, PLS-DA, PLS, kernel variable reduction	Apetrei and Apetrei (2014)
Extra virgin olive oil	Discrimination of samples according to their phenolic content and bitterness index. Correlation with the polyphenol content, the bitterness index (analyzed by chemical methods) and the bitterness degree (determined by a panel of experts)	5 CPEs modified with lanthanide bisphthalocyanines, 6 polypirroles conducting polymers and 1 unmodified CPE	PCA, PLS-DA, PLS, kernel variable reduction	Rodríguez-Méndez et al. (2008b)
Extra virgin olive oil	Discrimination between samples of different bitterness degree. Prediction of sensorial bitterness degree obtained by a panel of experts. Prediction of chemical parameters (bitterness index, free acidity, peroxide index and K indexes)	6 polypyrrole based electrodes	PCA, PLS-DA, PLS, kernel variable reduction	Apetrei (2012)
Extra virgin olive oil	Discrimination between samples of different total polyphenolic content. Prediction of total polyphenolic content. Discrimination between samples with different individual polyphenolic	6 polypyrrole based electrodes with different doping agents	PCA, PLS-DA, SIMCA, PLS, kernel variable reduction	Apetrei and Apetrei (2013)

Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
	compounds			
Maize and extra virgin olive oils	Discrimination between different vegetable oils of different nature. Discrimination between geographical origins of extra virgin olive oils	Pt microelectrodes	PCA, *k*-NN	Oliveri et al. (2009)
Extra virgin olive oil	Discrimination between olive cultivars	2 units of 20 all-solid-state electrodes with different pre-established mass combinations of 4 lipidic, 5 plasticizers and PVC high molecular weight polymer	LDA-SA	Dias et al. (2014)
Extra virgin olive oils	Discrimination between intensity sensory perception levels	2 units of 20 all-solid-state electrodes with different pre-established mass combinations of 4 lipidic, 5 plasticizers and PVC high molecular weight polymer	LDA-SA	Veloso et al. (2016)
Green tea infusions	Discrimination between quality grades	α-Astree^{TM} e-tongue (see further details in Wei et al. (2009))	PCA, BP-ANN	Chen et al. (2008)
Black and green tea infusions	Discrimination between black and green teas. Discrimination between geographical origins. Discrimination between quality grades	α-Astree^{TM} e-tongue (see further details in Wei et al. (2009))	PCA	He et al. (2009)
Black tea infusions	Discrimination between geographical origins. Discrimination between growing altitudes. Prediction of sensory attributes	α-Astree^{TM} e-tongue (see further details in Wei et al. (2009))	PCA, SLDA, ANOVA, PLS	Kovács et al. (2010)
Gluten-free and gluten-containing foodstuffs	Classification of samples according their gluten (or gliandins) level	2 units of 36 all-solid-state electrodes with PVC lipid polymeric membranes	LDA	Peres et al. (2011)

Acronyms used: ANOVA, Analysis of variance; ANN, Artificial neural network; ASCA, ANOVA-Simultaneous component analysis; BP-ANN, Back propagation-artificial neural network; CA, Cluster analysis; CCA, Canonical correlation analysis; CPE, Carbon paste electrode; DFA, Discriminant function analysis; FTIR, Fourier transform infrared spectroscopy; ISE, Ion-selective electrode; ISFET, Ion-selective field effect transistor; *k*-NN, *k*-Nearest neighbor; LDA, Linear discriminant analysis; LDA-SA, Linear discriminant analysis simulated annealing; MIR, Multiple internal reflection; MLR, Multiple linear regression; MOS, Metal oxide semiconductor; MOSFET, Metal oxide semiconductor field effect transistor; MPCA: Multi-way PCA; OSC, Orthogonal signal correction; PCA, Principal component analysis; PCR, Principal component regression; PEDOT, Poly(3,4-ethylenedioxythiophene); PLS, Partial least square; PLS-DA, Partial least square–discriminant analysis; PNN, Probabilistic neural network, PVC, poly(vinyl chloride); SFA, Simplified Fuzzy ARTMAP; SIMCA, Soft independent modelling class analogy; SLDA, Stepwise linear discriminant analysis.

Table 3

Applications of e-noses and e-tongues data fusion techniques in food authenticity/adulteration assessment.

Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
Honey	Discrimination between monofloral, polyfloral honeys, sugar-syrups and honeys adulterated with sugar-syrups	Combination of data from an e-nose and an e-tongue: - Cyranose320™ e-nose (32 non-selective polymer sensors blended with carbon black) - E-tongue (7 chalcogenide-based ISEs)	PCA, LDA, PNN	Zakaria et al. (2011)
Tualang honey	Detection of adulteration with beet and cane sugar	Combination of data from FTIR and an e-nose: - Cyranose320™ e-nose (see further details in Zakaria et al. (2011))	PCA, LDA, SLDA	Subari et al. (2012)
Honey	Discrimination between pure and sugar-adulterated honeys	Combination of data from FTIR, an e-nose and an e-tongue: - No details are provided about the e-nose device. - E-tongue (8 chalcogenide-based metallic electrodes and 1 a pH electrode)	PCA, LDA, PNN, SVM, *k*-NN	Maamor et al. (2014)
Honey	Discrimination between samples from different botanical origin	Combination of data from an e-tongue and Vis-NIR and UV-Vis spectroscopies: - E-tongue (Al, Au, Pt and indium thin oxide impedance sensors)	PCA, CA, MPCA combined with variable selection algorithms	Ulloa et al. (2013)
Extra virgin olive oil	Discrimination between samples from different olive cultivars. Prediction of sensorial bitterness degree obtained by a panel of experts. Prediction of the concentration of 20 polyphenolic compounds	Combination of data from an e-nose, an e-tongue and an e-eye (electronic panel): - E-nose (13 MOS sensors) - E-tongue (CPEs modified with 25 olive oils) - E-eye (transmittance spectra recorded by LEDs in the 780-380 nm for calculation of color coordinates)	PCA, PLS-DA, PLS, kernell variable reduction	Apetrei et al. (2010)
Virgin olive oil	Discrimination between geographical origins	Combination of data from an e-nose and an e-tongue: - E-nose (5 different tin-dioxide gas sensors) - E-tongue (Pt, Au, glassy carbon and indium tin oxide voltammetric sensors)	PCA, CA, SVM, ANOVA variable selection algorithm	Haddi et al. (2013)
Extra virgin olive oil	Discrimination between samples from different cultivars and geographical origins. Prediction of percentage of adulteration. Prediction of chemical parameters	Combination of gas and liquid anthocyanins based sensors (BIONOTE): - Gas sensors (6 QMBs) - Liquid sensors (screen-printed Au voltammetric electrodes)	PCA, PLS-DA	Santonico et al. (2015)
Extra virgin olive oil	Discrimination between geographical origins	Combination of data from an e-nose, an e-tongue and chemical variables: - Model 3320 Applied Sensor Lab Emission Analyser™ e-nose (10 MOS and 10 MOSFET sensors) - E-tongue (a dual and a single glassy carbon amperometric sensors)	PCA, ANN	Cosio et al. (2006)
Cherry tomato juice	Discrimination between non-adulterated and adulterated samples	Combination of data from an e-nose and an e-tongue: - PEN 2™ e-nose (10 different MOS sensors) - α-Astree™ e-tongue (7 ISFETs based on polymer membranes)	PCA, CA, PCR, MLR (ANOVA or stepwise variable selection algorithms)	Hong et al. (2014b)
Cherry tomato juice	Detection of adulteration with overripe tomato juices	Combination of data from an e-nose and an e-tongue: - PEN 2™ e-nose (see further details in Hong et al. (2014b)) - α-Astree™ e-tongue (see further details in Hong et al. (2014b))	PCA, CDA, Lib-SVM, PCR combined with variable selection algorithms	Hong and Wang (2014)

Acronyms used: ANOVA, Analysis of variance; ANN, Artificial neural network; BIONOTE, biosensor-based multisensorial system for mimicking nose, tongue and eyes; CA, Cluster analysis; CPE, Carbon paste electrode; FTIR, Fourier transform infrared spectroscopy; ISE, Ion-selective electrode; ISFET, Ion-selective field effect transistor; *k*-NN, k-Nearest neighbor; LDA, Linear discriminant analysis; Lib-SVM, Library Support Vector Machines; MLR, Multiple linear regression; MOS, Metal oxide semiconductors; MOSFET, Metal oxide semiconductors field effect transistor; MPCA: Multi-way PCA; PCA, Principal component analysis; PCR, Principal component regression; PLS, Partial least square; PLS-DA, Partial least square–discriminant analysis; PNN, Probabilistic neural network; QMBs, quartz micro balances; SLDA, Stepwise linear discriminant analysis; SVM, support vector machine, UV-Vis, Ultraviolet-visible, Vis-NIR, Visible-near infrared.

Figure Captions:

Fig. 1 – Schematic representation of (a) an electronic nose, and (b) an electronic tongue.