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Additional Information

# 1 **Electronic noses and tongues to assess food authenticity and** 2 **adulteration**

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## 9 10 11 **ABSTRACT**

### 12 13 *Background*

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

### 19 *Scope and Approach*

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

### 27 *Key Findings and Conclusions*

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

### 32 33 *Keywords:*

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

36

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## 41 1. Introduction

42

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

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

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

69 In the last years, many reviews on e-noses and/or e-tongues fundamentals  
70 and applications in several research areas have been published in the literature,  
71 mainly in the field of food analysis (e.g., Boeker, 2014; Ciosek and Wróblewski, 2011;  
72 del Valle., 2012, Escuder-Gilabert and Peris, 2010; Kiani et al., 2016; Loutfi et al.,  
73 2015; Peris and Escuder-Gilabert, 2009; Rodríguez-Méndez, 2016; Śliwińska et al.,  
74 2014; Tahara and Toko, 2013; Vlasov et al., 2005). This paper will then focus on the  
75 employment of both e-noses and e-tongues in food authenticity assessment (and

76 hence the detection of food adulteration). After briefly commenting the fundamentals  
77 of this type of devices, the most relevant contributions in this field over the past ten  
78 years will be dealt with. In this sense, and as a general overview, in a recent chapter  
79 of a book (Karoui, 2012) devoted to food authenticity and fraud, Karoui discusses the  
80 relative potential and ease of application of different technologies for the confirmation  
81 of food quality and adulteration. Special emphasis is put on e-nose technology  
82 (combined with chemometric tools) as a promising technique in this field. Some  
83 examples clearly show that there has always been a risk of fraud, since food became  
84 a trade object. The chapter also describes the different kinds of food adulteration and  
85 related fraudulent practices, with details of detection methods, including the use of e-  
86 noses. In a similar way, Cappozzo (2013) has presented recent analytical  
87 innovations for quality assurance in the detection of food adulteration through the  
88 utilization of e-noses. Panchariya et al. (2013) have reported an overview of the  
89 applications of e-noses and e-tongues for classification and authentication of  
90 beverages. As far as e-tongues are concerned, Sliwinska et al. (2014) have also  
91 dealt with their potential in the authenticity and falsification assessment of foodstuffs.

92

## 93 **2. General concepts**

94

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

100

### 101 2.1. Fundamentals of e-noses

102

103 E-noses are designed to detect and distinguish among complex odors (from  
104 food samples) making use of a sensor array, which is composed of broadly tuned  
105 (non-specific) sensors that are treated with different odor-sensitive (bio)chemical  
106 substances. An odor stimulus now yields a characteristic fingerprint (or smellprint)  
107 from the group of sensors. These patterns from known odors are then utilized to  
108 generate a database that is subjected to multivariate analysis, so that unknown odors  
109 can therefore be identified and classified. Nevertheless, it should be remarked that, in  
110 recent years, the usual sensor types used for e-nose instruments have been

111 considerably improved by new technologies developed in this field, and either a set  
112 of gas sensors or mass spectrometry (or their combination) are commonly utilized for  
113 this purpose. Anyway, and in a broader sense, electronic noses basically consist of  
114 three elements (Fig. 1a), namely: (i) a sample handling system, (ii) a detection  
115 system, and (iii) a data processing system.

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

124 There are many different types of electrochemical sensors (e.g. metal-oxide  
125 gas sensors, metal-oxide semiconductor field effect transistors, acoustic wave gas  
126 sensors, electrochemical gas sensors, quartz crystal microbalance sensors,  
127 conducting polymer gas sensors, surface acoustic wave devices, field-effect gas  
128 sensors, fiber-optic gas sensors, and others) and many different types of sensor-  
129 coating materials which are classified according to additive doping materials, the type  
130 and nature of the chemical interactions, the reversibility of the chemical reactions and  
131 running temperature. A summary of the types and mechanisms involved with some  
132 common gas sensor technologies is contained in the work of Wilson and Baietto  
133 (2009).

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

143

## 144 2.2. Fundamentals of e-tongues

145

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

156 The electronic tongue system relies on sensors with only moderate selectivity  
157 and having the so-called cross-sensitivity. In this way, each sensor in the array, in  
158 principle, delivers information on the concentrations of a number of analytes, the next  
159 step being to decode the signals obtained from the sensor array. The sensors in the  
160 array can be of different nature, although Ion Selective Electrodes (ISEs)  
161 predominate among the various types of sensors utilized, electrodes with  
162 chalcogenide glass membranes (Mikhelson, 2013) being particularly suitable for  
163 these devices. An ISE generates a difference in electrical potential between itself and  
164 a reference electrode, and this output potential is proportional to the activity of the  
165 selected analyte in the sample solution according to the well-known Nernst equation  
166 (Skoog et al., 2013), although a calibration of the working electrode should be  
167 previously carried out using a series of standard solutions with known composition.

168 The number of sensors in the array can vary, but most typical number is about 10–  
169 20. Unlike in the classical measurements with ISEs, the electronic tongue system can  
170 work without a reference electrode. In such a setup, the potential difference is  
171 measured for all pairs of the electrodes in the array. This is advantageous since  
172 reference electrodes often cause problems with the measurements.

173 The signals obtained from the sensor array are processed using different  
174 chemometrical methods, whereas the interpretation and representation of the data is  
175 often based on the principal component analysis method. This allows for the  
176 characterization of the samples not only in terms of the concentrations (activities) of

177 the particular analytes, but also for the recognition of the nature of the sample, since  
178 different types of samples fall into different places in the principal components plot.

179

### 180 **3. E-noses in food authenticity/adulteration assessment**

181

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

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

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



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

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

238 Continuing with the field of edible oils, the zNose™ electronic nose was used  
239 to evaluate the adulteration levels of virgin coconut oil (Marina et al., 2009). This  
240 device was utilized to obtain a fingerprint of volatile compounds occurring in the oil  
241 samples. Virgin coconut oil was adulterated with different contents (ranging from 1 to  
242 20 % w/w) of refined, bleached and deodorized palm kernel olein, the corresponding  
243 peaks being identified in the chromatogram and fitted to a curve using linear  
244 regression. The relationship between the peak initially identified as methyl  
245 dodecanoate and the percentage of palm kernel olein added gave rise to the best  
246 result ( $r = 0.95$ ). On the other hand, correlation coefficient values of  $r = 0.92$  and  $r =$

247 0.89 were achieved between adulterant peak methyl dodecanoate and of the iodine  
248 and peroxide values, respectively. PCA was employed to discriminate between  
249 adulterated and pure samples; the results obtained were satisfactory, with 74 % of  
250 the variation accounted for by the first principal component and 17 % by the second  
251 principal component.

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

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

277 A common adulteration of honey takes place when sugar concentrate is added  
278 to this product; unfortunately, laboratory tests have proved so far to be ineffective in  
279 the detection of this fraud. A Chinese research team (Pei *et al.*, 2015) has developed  
280 a method for rapid detection of Acacia honey adulteration using a FOX 4000<sup>TM</sup> e-  
281 nose. Samples were spiked with different amounts of rape honey and rice syrup, and

282 the information (from the e-nose) on both natural and adulterated honey was  
283 analyzed by PCA. LDA was employed to study the ability of qualitative recognition of  
284 the e-nose for adulterated honey. The results showed that there was a linear  
285 relationship between e-nose signals and the adulteration level. On the other hand,  
286 the minimum amount of rape honey and rice syrup added leading to honey aroma  
287 system changes was 2 % and 1 %, respectively. Therefore, honey aroma system can  
288 easily be changed by adulterant compounds. This also demonstrated that the e-nose  
289 had a strong discriminable ability for honey adulteration. The results concluded that  
290 pure honey and adulterated honey can be distinguished by LDA pattern recognition  
291 algorithms, this fact resulting in a fast and accurate identification of honey  
292 adulteration.

293 An e-nose was also proposed for the detection and differentiation of lard from  
294 other kinds of animal fats as well as from foodstuffs containing lard (Nurjuliana et al.,  
295 2011). The results obtained are displayed in the form of the so-called *VaporPrint*. In  
296 this two-dimensional olfactory image, the radial angles representing the sensor yield  
297 individual patterns (smellprints) of the odor of different animal body fats. PCA was  
298 utilized to interpret the results achieved and gave rise to a satisfactory grouping of  
299 samples (61 % of the variation corresponded to the first principal component, and 29  
300 % to the second principal component). All of the lard-containing samples formed a  
301 separate group from those having no lard. On the other hand, the ability to detect the  
302 presence of lard in food products helps Halal authentication (compliance with Islamic  
303 law).

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

317           The high cost of saffron inevitably (and unfortunately) leads to frequent  
318 attempts to adulterate it. That is why in a recent work (Heidarbeigi et al., 2014) the  
319 odor smellprints of saffron, saffron with yellow styles, safflower, and dyed corn stigma  
320 were recognized by an electronic nose. The characteristics of the obtained results  
321 were extracted and analyzed, PCA being used for this purpose. The corresponding  
322 data were then confirmed by BP-ANN, and showed that the e-nose is able to detect  
323 the saffron adulteration with excellent results. The authors conclude that this e-nose-  
324 based system could yield a good differentiation of the saffron and the adulterated one  
325 (100 and 86.9 % classification accuracy respectively) at adulteration levels over 10 %  
326 using ANN.

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

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

347           Yu et al. (2007) performed a study with a view of monitoring the adulteration of  
348 milk with water or reconstituted milk powder utilizing the PEN 2™ electronic nose  
349 with ten different MOS sensors. For this purpose, a series of experiments were  
350 conducted over 7 days of storage using three types of samples: whole fluid milk,  
351 reconstituted milk powder, and whole fluid milk adulterated with several amounts of

352 water. The data obtained were a consequence of applying two chemometric  
353 techniques: PCA and LDA. According to the authors, the corresponding results  
354 proved that the artificial sense used (the e-nose) was able to differentiate the purity of  
355 milk samples when skimmed milk is diluted with variable amounts of water, and both  
356 LDA and PCA show a regular distribution of the results for the aforementioned three  
357 samples analyzed. Finally, with these two chemometric methods, the electronic nose  
358 could also differentiate between milk samples that had been stored for different  
359 numbers of days.

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

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

380 Two food adulteration cases (a pure variety of green coffee beans and pure  
381 cayenne with bell pepper powder) were studied by (Rodríguez et al., 2014) with the  
382 goal of reporting the improvements achieved in the discrimination of complex aroma  
383 samples with very small differences in odor pattern. For this purpose, they utilized a  
384 portable e-nose consisting of a sensor array which records changes in conductivity  
385 as a function of time when aroma molecules reach the sensors. The proposed  
386 method is then based on the application of unfolded cluster analysis to selected time

387 windows within the temporal evolution of the aroma profile (recorded by the sensor  
388 array), providing an efficient, rapid, and reliable data analysis tool. The results  
389 obtained showed that this procedure enables to discriminate highly similar samples,  
390 thus decreasing the probability of a wrong grouping due to the use of doubtful data.  
391 The automation of this type of analysis is easy and enhances the efficiency of the e-  
392 nose in a significant way, what implies reducing the time of sensor's signal recording  
393 that is required for a reliable assessment of the studied system. The results were  
394 validated by clustering the sample component scores that are obtained by applying  
395 parallel factor analysis to the original three-dimensional data array.

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

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

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

422 SLDA and PCA results obtained from e-nose data demonstrated that the device used  
423 did not perform very well in the discrimination of samples according to their  
424 geographical origin (success rates over 75 % and 37 % in training and cross-  
425 validation steps, respectively). Nevertheless, the e-nose exhibited an excellent ability  
426 to classify samples according to their growing altitude (100 % success rates in both  
427 calibration and cross-validation steps). On the other hand, e-nose data provided poor  
428 to moderate prediction of sensory attributes by PLS modeling ( $0.59 \leq r \leq 0.89$  and  
429  $0.45 \leq r \leq 0.72$ , in training and cross-validation steps, respectively). Finally, sensory  
430 analysis proved that an average tea consumer (without specialized and intensive  
431 training) can hardly differentiate Sri Lanka teas from different geographical origins.  
432 As regards the results obtained from the e-tongue data, they are described in the  
433 next section.

434

#### 435 **4. E-tongues in food authenticity/adulteration assessment**

436

437 E-tongues are also emerging as promising supplemental techniques to  
438 classical analytical methods for a fast and low cost detection of malpractices. As  
439 shown in Table 2, e-tongues have shown their capability in the detection of food  
440 adulteration as well as in the authenticity assessment of different types of foodstuffs,  
441 such as honeys (Dias et al., 2008; Escriche et al., 2012; Garcia-Breijo et al., 2013;  
442 Major et al., 2011; Sousa et al., 2014; Wei and Wang, 2011; Wei et al., 2009), milk  
443 and dairy products (Dias et al., 2009; Paixão and Bertotti, 2009), alcoholic beverages  
444 (Gutiérrez et al., 2010; Gutiérrez-Capitán et al., 2013; Moreno-Codinachs et al.,  
445 2008; Novakowski et al., 2011; Parra et al., 2004; Parra et al., 2006; Pigani et al.,  
446 2008; Rodríguez-Méndez et al., 2008a; Rudnitskaya et al., 2007; Rudnitskaya et al.,  
447 2010), edible oils (Apetrei and Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et  
448 al., 2005; Apetrei et al., 2007; Apetrei, 2012; Dias et al., 2014; Oliveri et al., 2009;  
449 Rodríguez-Méndez et al., 2008b), and teas (Chen et al., 2008; He et al., 2009;  
450 Kovács et al., 2010).

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

456 2014; Wei and Wang, 2011; Wei et al., 2009) and geographical origin (Wei et al.,  
457 2009).

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

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

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

485 The aforementioned commercial e-tongue  $\alpha$ -Astree™ has also been applied  
486 for botanical classification and prediction of physicochemical properties of 12  
487 samples from three botanical origins (Major et al., 2011). PCA analysis of e-tongue  
488 data showed a very good clustering of samples according to their botanical origin. On  
489 the other hand, a 100 % accuracy was obtained in the botanical classification of  
490 honey samples using ANN. Satisfactory ( $r = 0.979 - 0.999$ ) ANN models for the



491 prediction of studied physicochemical properties (electrical conductivity) and content  
492 of different chemical parameters (acidity, water content, invert sugar, and total sugar)  
493 from the sensors outputs were obtained, which clearly highlights the potential of the  
494 e-tongue for a fast honey analysis.

495 Escriche et al. (2012) used another potentiometric e-tongue, comprising seven  
496 metals and metallic compounds, for quality control and authenticity assessment of  
497 honey. They resorted to PCA and ANN based on a Fuzzy ARTMAP algorithm and  
498 leave-one-out cross-validation to show the usefulness of the proposed e-tongue for  
499 discriminating samples according to their four botanical origins (ANN classification  
500 success exceeding 93 %), although it was not able to clearly differentiate among the  
501 three thermal treatments used. On the other hand, acceptable to good ( $r = 0.74 -$   
502  $0.96$ ) PLS correlations between the e-tongue data and seven physicochemical  
503 parameters were obtained. Nevertheless, poor ( $r = 0.49 - 0.73$ ) PLS correlations  
504 between e-tongue data and volatiles concentrations were observed. In a further work  
505 (Garcia-Breijo et al., 2013), data were dealt with using ANN based on a simplified  
506 Fuzzy ARTMAP (SFA) and graphical user interface (GUI) for MATLAB® cross-  
507 validation. For botanical origin classification, 100 % recognition rates were obtained  
508 in the supervised phase, as well as when utilizing data of four selected electrodes.  
509 For thermal treatment of samples, up to 83 % recognition rate was achieved. In the  
510 non-supervised phase, a recognition rate of 69 % was obtained for four new test  
511 samples. This rate was increased up to 75 % by reducing the number of electrodes  
512 to four.

513 Another successful application of e-tongues for ensuring monofloral honey  
514 authenticity is the study reported by Sousa et al. (2014). A specifically designed  
515 potentiometric e-tongue consisting of two replicated groups of 20 all-solid-state  
516 sensors with different cross-sensitivity membranes was used. 65 Portuguese  
517 monofloral honey samples (harvested in different years) were studied. LDA based on  
518 the e-tongue signal profiles from 13 sensors -selected with a simulated annealing  
519 (SA) variable selection algorithm- resulted in the fact that 91 % (for both original data  
520 and leave-one-out cross-validation) of the honey samples were correctly classified  
521 according to three main color groups. For each of these color groups, all honeys  
522 were correctly classified according to their floral origin using a variable selection of e-  
523 tongue data combined with a LDA leave-one-out cross-validation strategy. However,  
524 LDA model using all samples data gave rise to a poor classification (in cross-  
525 validation) of honey samples according to their floral origin. The authors pointed out

526 that the use of selected variables increased the accuracy performance of the LDA  
527 models, hence the importance of using variable selection algorithms in this type of  
528 studies. More recently, authors from this research group have published a book  
529 chapter focused on the applications of electrochemical sensors to evaluate  
530 antioxidant capacity of bee hives products (Peres et al., 2016).

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

539 Detection of milk adulteration has also been addressed by Paixão and Bertotti  
540 (2009). The authors developed disposable integrated voltammetric e-tongues  
541 composed of bare and Prussian Blue modified electrodes. PCA inspection of Au and  
542 Prussian Blue-modified gold electrodes data allowed to discriminate milk samples  
543 adulterated with H<sub>2</sub>O<sub>2</sub> as well as to differentiate several pasteurization processes of  
544 samples. Disposable electrodes are of particular interest in milk analysis since  
545 adsorption onto the sensor surface of substances present in high concentrations in  
546 the sample matrix can alter the voltammetric signal.

547 Fraudulent practices are increasingly performed in several alcoholic  
548 beverages, most notably in wines. Both the quality control of wines and grape juices  
549 and the quantitation of different compounds have a paramount significance in wine  
550 production. The stages in the wine-making process have to be carefully monitored to  
551 control potential adulterations as well as to determine the concentration of some key  
552 components for the final quality of the product. In this context, e-tongues have proven  
553 their usefulness to authenticate geographical origin (Parra et al., 2004; Pigani et al.,  
554 2008), grape variety (Gutiérrez et al., 2010; Gutiérrez-Capitán et al., 2013; Moreno-  
555 Codinachs et al., 2008; Pigani et al., 2008; Rodríguez-Méndez et al., 2008a), vintage  
556 (Moreno-Codinachs et al., 2008) and age (Parra et al., 2004; Rudnitskaya et al.,  
557 2007; Rudnitskaya et al., 2010) of wines or grape juices, and to detect adulterated  
558 wines and whiskeys (Novakowski et al., 2011; Parra et al., 2006).

559 In this framework, Parra et al. (2004) developed a voltammetric sensor array  
560 composed of rare-earth bisphthalocyanine carbon paste electrodes (CPEs) for the

561 discrimination of six Spanish red wines from the same variety of grape but from three  
562 different origin designations and ageing stages. PCA applied to e-tongue data  
563 showed a good discrimination of the tested samples. The discrimination ability of the  
564 e-tongue was similar to that obtained using eight chemical variables coming from  
565 chemical analysis. In a further work (Rodríguez-Mendez et al., 2008a), some authors  
566 from the same research group built an e-tongue consisting of CPEs modified with  
567 rare-earth bisphthalocyanines and perylenes. PCA study of data obtained for six  
568 white wines demonstrated the e-tongue ability to discriminate among the grape  
569 varieties utilized for sample preparation.

570 The effectiveness of three different poly(3,4-ethylenedioxythiophene) (PEDOT)  
571 conducting polymer modified electrodes to analyze similar matrices (six white wines  
572 of different grape varieties and geographical areas) was tested by Pigani et al.  
573 (2008). Voltammetric responses of each sensor separately and the combined  
574 responses of two or three sensors were used in the chemometric analysis. Partial  
575 least square-discriminant analysis (PLS-DA) on voltammetric data proved that all the  
576 electrodes could successfully classify (nearly 100 % correct classifications) samples  
577 according to their variety.

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

591 In the paper by Gutiérrez et al. (2010), the capability of a hybrid e-tongue for  
592 the classification of wines according to the grape varieties and vintage as well as for  
593 the prediction of some chemical and optical parameters of interest in wine quality  
594 control is demonstrated. The proposed e-tongue included an array of electrochemical  
595 microsensors and a colorimetric optofluidic system. Both of them can be integrated

596 into the same platform, thus providing portable, rapid, and feasible equipment for *in-*  
597 *situ* measurements. E-tongue data obtained for red (samples from the same vintage  
598 but different grape varieties) and white (two vintages and four grape varieties) wines  
599 were processed by means of PLS and PCA. In spite of the small number of samples,  
600 the potential of the developed e-tongue in this kind of studies is clearly demonstrated.

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

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

624 With respect to the use of e-tongues to detect adulteration of alcoholic drinks,  
625 Parra et al. (2006) proposed a voltammetric e-tongue composed of two kinds of  
626 sensors (phthalocyanine-based CPEs and conducting polypyrrole polymers based  
627 electrodes) for the detection of the most common adulterants of red wines. The e-  
628 tongue data (PLS regression) yielded good estimations (prediction errors between 1  
629 and 15 %) of the concentrations of seven chemical compounds (common  
630 adulterants) added to the samples. Additionally, PCA on e-tongue data exhibited a

631 good performance for distinguishing the different kinds of adulterations. The inclusion  
632 of those two types of sensors in the e-tongue increased their discrimination ability.

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

645 Authenticity assessment of vegetable oils is another research area of interest  
646 for e-tongues (Apetrei and Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et al.,  
647 2005; Apetrei et al., 2007; Apetrei, 2012; Dias et al., 2014; Oliveri et al., 2009;  
648 Rodríguez-Méndez et al., 2008b). The high price of EVOOs and VOOs make them  
649 ideal “candidates” for adulteration, be it mislabeling or blending with cheaper olive  
650 and seed oils. Some VOOs and EVOOs are certified as protected designation of  
651 origin, which are partly related to olive oil production and processing made in a  
652 specific geographical origin. Furthermore, label authentication of olive cultivar is of  
653 paramount importance due to marketing of high-quality (and high-price) monovarietal  
654 EVOOs. Label information about the geographical origin affects consumers’  
655 acceptability, while information about cultivar significantly impacts on the expectation  
656 of pungency and bitterness for olive oils. As shown in Table 2, papers reported in the  
657 literature on this topic have demonstrated the effectiveness of e-tongues for the  
658 discrimination among different vegetable oils (Apetrei et al., 2005; Apetrei and  
659 Apetrei, 2014; Oliveri et al., 2009), quality grades of olive oils (Apetrei et al., 2005),  
660 olive cultivars (Dias et al., 2014) and geographical origin of EVOOs (Oliveri et al.,  
661 2009). E-tongues have also shown their ability for quantitative prediction of different  
662 physicochemical parameters utilized for EVOOs characterization (Apetrei and  
663 Apetrei, 2013; Apetrei and Apetrei, 2014; Apetrei et al., 2007; Apetrei, 2012;  
664 Rodríguez-Méndez et al., 2008b), as well as to detect EVOOs adulterations with  
665 seed oils (Apetrei and Apetrei, 2014). Finally, Rodríguez-Méndez et al. (2010) have

666 authored a chapter book including some applications of e-tongues purposely  
667 designed for the characterization of olive oils.

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

680 In a more recent work (Apetrei and Apetrei, 2014), the usefulness of this type  
681 of e-tongues to detect EVOOs adulteration is demonstrated. PCA, PLS-DA and PLS  
682 applied to e-tongue data showed the capability of this device (a) to discriminate pure  
683 oils (an EVOO and three seed oils) according to their botanical origins; (b) to predict  
684 total polyphenolic content of binary mixtures of EVOO and seed oils ( $r$  values near  
685 0.99, in both PLS calibration and validation); (c) to classify the adulterated EVOOs  
686 when the concentration of adulterant oil was over 5 %; and (d) to estimate the  
687 composition of EVOO and seed oil mixtures within the range 2 – 25 % range ( $r > 0.99$   
688 in both PLS calibration and validation).

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

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

701 PLS-DA exploration of the data obtained from such systems demonstrated their  
702 ability to discriminate six Spanish EVOOs according to their sensorial and chemical  
703 bitterness, as well as to predict bitterness degree and chemical parameters such as  
704 peroxide value, free acidity, bitterness index, and K indexes ( $r > 0.87$  in both,  
705 calibration and validation of the PLS models) (Apetrei, 2012). On the other hand,  
706 classification of 18 EVOOs according to their total polyphenolic content was  
707 successfully accomplished using PCA, PLS-DA and SIMCA (Apetrei and Apetrei,  
708 2013). A good PLS model for the quantification of this chemical parameter was  
709 obtained ( $r > 0.98$  for both the training and the test set). Furthermore, e-tongue data  
710 for spiked EVOO emulsion samples having different individual phenolic compounds  
711 permitted PCA discrimination and an error-free PLS-DA classification of these  
712 substances.

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

722 As commented on previously, label authentication of monovarietal EVOOs is  
723 of primary importance. In the paper by Dias et al. (2014), a potentiometric e-tongue  
724 with different cross-sensitivity membrane sensors was fabricated to discriminate 18  
725 Portuguese and Spanish monovarietal EVOOs according to the olive cultivar. The e-  
726 tongue is analogous to that used for honey classification (Sousa et al., 2014). In a  
727 similar way to that paper, the most informative potentiometric sensor signal profiles  
728 were selected using an SA algorithm to establish LDA models with the best leave-  
729 one-out cross-validation predictive performance. Hydro-ethanolic extracts of EVOOs  
730 were utilized to solve the problem of electrochemical assays in oils. E-tongue data  
731 gave rise to outstanding classification results according to the olive cultivar for  
732 EVOOs of each country (100 % and 97.5-100 % correct classification for the original  
733 data and for cross-validation, respectively). Notwithstanding, no simultaneous  
734 discrimination of all the six Spanish and Portuguese cultivars could be achieved (92  
735 % and 43 % correct classification for original data and cross-validation, respectively).

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

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

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

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

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



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

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

793

## 794 **5. E-noses and e-tongues data fusion in food authenticity/adulteration** 795 **assessment**

796

797 In some applications, and due to the high complexity of food samples, the  
798 employment of just e-nose or e-tongue data is insufficient, and multisensor data  
799 fusion techniques, e.g. combination of e-nose with e-tongue and/or spectroscopic  
800 data, have been utilized as efficient characterization methodologies. Nevertheless, in  
801 most cases, a variable selection seems to be imperative for the application of sensor  
802 array data -particularly when a data fusion strategy is used- in order to remove  
803 response variables or sensors that are redundant, noisy, or irrelevant for qualitative  
804 or quantitative purposes. In this sense, a very interesting review about analytical  
805 techniques and strategies employed in data fusion methodologies for food and

806 beverage authentication and quality assessment has recently been published (Borràs  
807 *et al.*, 2015). The present section is then just focused on applications based on the  
808 joint use of e-noses and e-tongues. The main features of such studies are  
809 summarized in Table 3 and are briefly described below.

810 In the literature there are some examples in which multisensor data fusion  
811 techniques have given rise to enhanced results for honey analysis (Maamor *et al.*,  
812 2014; Subari *et al.*, 2012; Ulloa *et al.*, 2013; Zakaria *et al.*, 2011). The combination of  
813 data obtained from both an e-nose and an e-tongue to discriminate between different  
814 honeys, sugar syrups, and sugar adulterated honey samples has also been proposed  
815 (Zakaria *et al.*, 2011). Samples were analyzed by means of the Cyranose320<sup>TM</sup> e-  
816 nose (32 non-selective sensors of different types of polymer matrix, blended with  
817 carbon black), as well as by a potentiometric e-tongue consisting of seven  
818 chalcogenide-based ion selective electrodes (ISEs). By combining the data obtained  
819 from both e-systems, the discrimination ability for all the analyzed samples was  
820 significantly enhanced. The best results turned out to be those using LDA (100 %  
821 success rates in both training and cross-validation steps).

822 It is also worth reporting the comparison between data obtained from single  
823 modality and fusion methods in the classification of pure or adulterated Tualang  
824 honeys (Subari *et al.*, 2012). Ten different brands of certified pure Tualang honey  
825 from Malaysia and Sumatra were blended with different concentrations of cane and  
826 beet sugar solutions and analyzed by means of the Cyranose320<sup>TM</sup> e-nose and  
827 Fourier transform infrared spectroscopy (FTIR). The best classification rate (92.2 %  
828 in validation) was obtained when using normalized low-level FTIR and e-nose fusion  
829 data by means of SLDA. In a further work Maamor *et al.* (2014) (authors from the  
830 same research group) demonstrated that the discrimination ability between pure and  
831 sugar-adulterated Tualang honeys could be improved by utilizing combined data from  
832 FTIR, an e-nose, and a potentiometric e-tongue. PCA was then used to reduce high  
833 dimensional features of these three techniques. Among the different classification  
834 methods studied, *k*-NN provided the best performance for the training and test sets  
835 (100 and 96.4 % correct classification, respectively).

836 Ulloa *et al.* (2013) have tried to classify four commercial brands of Portuguese  
837 honey according to their botanical origin by means of sensor fusion of an impedance  
838 e-tongue and visible–near infrared (Vis–NIR) and ultraviolet–visible (UV–Vis)  
839 spectroscopies assisted by PCA and CA. 13 heterogeneous Portuguese honey  
840 nectar samples were analyzed. Different chemometric tools showed that fusion of the

841 corresponding data obtained yielded a better discrimination ability than that achieved  
842 with the three individual techniques. In this sense, multi-way PCA (MPCA) proved to  
843 be an excellent (100 % classification success) alternative for data fusion, unlike  
844 simple concatenation of all matrices. Last but not least, a variable selection method  
845 based on one-dimensional clustering was developed to define two new strategies,  
846 both of them giving rise to even better defined sample clusters. Notwithstanding, the  
847 authors make it clear that the aim of the work was the demonstration of the proposed  
848 methods and there is clearly a need for further research work with a larger number of  
849 samples.

850 Multisensor data fusion techniques have also demonstrated to be of great  
851 utility for the analysis of some types of olive oils (Apetrei et al., 2010; Haddi et al.,  
852 2013). In the paper of Apetrei et al. (2010), the so-called electronic panel (data fusion  
853 of three systems, namely: an e-nose, an e-tongue, and an e-eye) was employed to  
854 characterize the organoleptic properties of 25 EVOOs from three different olive  
855 varieties. The e-nose consisted of a set of 13 MOS sensors whereas the e-tongue  
856 was based on modified CPE voltammetric sensors. PCA and PLS-DA of data  
857 showed that the combined system had a higher capability of sample discrimination  
858 according to the olive variety than that obtained with the three instruments used  
859 separately. Finally, PLS regression models provided good correlation coefficients  
860 between e-tongue data and bitterness scores (PLS1,  $r > 0.97$  in both model  
861 calibration and cross-validation) as well as between the electronic panel data and the  
862 concentrations of 20 polyphenolic compounds (PLS2,  $r > 0.9$  in calibration and cross-  
863 validation).

864 E-tongue and e-nose data fusion also gave rise to a better performance than  
865 the independent e-devices in the characterization of Moroccan VOOs (Haddi et al.,  
866 2013). A certain number of VOOs (from the same variety and harvested in the same  
867 year) from five different regions of Morocco were analyzed using a voltammetric e-  
868 tongue and a MOS-based e-nose. PCA and CA applied to a reduced subset  
869 containing optimal variables (selected with the help of a recently developed variable  
870 selection strategy based on ANOVA) improved the classification of the VOOs with  
871 respect to the use of all the variables. Support vector machines (SVM) performed on  
872 the reduced subset confirmed the correct identification of all VOOs.

873 The so-called biosensor-based multisensorial system for mimicking nose,  
874 tongue and eyes (BIONOTE) developed by Santonico et al. (2015) has proven  
875 excellent results for detecting EVOOs adulteration. This device includes gas and

876 liquid sensors based on anthocyanins sensing interfaces. Quartz micro balances  
877 (QMBs) were used as transducers for the gas sensor array, while the sensor liquid  
878 array was composed of screen-printed gold voltammetric electrodes. Data fusion of  
879 sensors data allowed the discrimination of twelve EVOOs from different cultivars and  
880 geographical origins, the detection of adulteration of EVOOs with other four  
881 vegetable oils up to concentrations lower than 5 % as well as the prediction of  
882 common chemical parameters related to the quality of the EVOOs.

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

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

901

## 902 **6. Conclusions**

903

904 In this review, the most relevant applications of e-noses and e-tongues in food  
905 authenticity assessment –in many cases leading to the detection of food adulteration-  
906 have been examined. This subject area is particularly (and increasingly) important in  
907 these last years, since it not only concerns the constant fight with food adulterers, but  
908 also relatively new aspects such as bioterrorism or food security. All these problems  
909 clearly highlight the need for further development and refinement of the existing  
910 analytical techniques. In this sense, the use of “artificial senses” such as those

911 discussed in the present paper will undoubtedly contribute to overcome the  
912 shortcomings of other analytical techniques still in use. Furthermore, future  
913 developments in the use of advanced sensors arrays will lead to superior electronic  
914 senses with more capabilities, thus making the authenticity and falsification  
915 assessment of food products a faster and more reliable process.

916

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918

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## References

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922 **Abbreviations**

923

- 924 • **ANN:** artificial neural network
- 925 • **ANOVA:** analysis of variance
- 926 • **ASCA:** ANOVA-simultaneous component analysis
- 927 • **BIONOTE:** biosensor-based multisensorial system
- 928 • **BP-ANN:** back propagation-artificial neural network
- 929 • **CA:** cluster analysis
- 930 • **CCA:** canonical correlation analysis
- 931 • **CDA:** canonical discriminant analysis
- 932 • **CPE:** carbon paste electrode
- 933 • **DFA:** discriminant function analysis
- 934 • **EDA:** exploratory data analysis
- 935 • **e-nose:** electronic nose
- 936 • **e-tongue:** electronic tongue
- 937 • **EVOO:** extra virgin olive oil
- 938 • **FIA:** flow injection analysis
- 939 • **FID:** flame ionization detector
- 940 • **FLT:** Fisher linear transformation
- 941 • **FTIR:** Fourier transform infrared spectroscopy
- 942 • **GC/MS:** gas chromatography-mass spectrometry
- 943 • **GC:** gas chromatography
- 944 • **GRNN:** general regression neural network
- 945 • **GUI:** graphical user interface
- 946 • **IMS:** ion mobility spectrometer
- 947 • **ISE:** ion-selective electrode
- 948 • **ISFET:** ion-selective field effect transistor
- 949 • **k-NN:** *k*-nearest neighbors
- 950 • **LDA:** linear discriminant analysis
- 951 • **LDA-SA:** linear discriminant analysis-simulated annealing
- 952 • **Lib-SVM:** support Vector Machines
- 953 • **MANOVA:** multivariate analysis of variance
- 954 • **MI:** mutual information criteria
- 955 • **MLAPV:** multifrequency large amplitude pulse voltammetry
- 956 • **MLR:** multiple linear regression

- 957 • **MOS:** metal oxide semiconductors
- 958 • **MOSFET:** metal oxide semiconductor field effect transistor
- 959 • **MPCA:** Multi-way PCA
- 960 • **MS:** mass spectrometry
- 961 • **OSC:** orthogonal signal correction
- 962 • **PCA:** principal component analysis
- 963 • **PCR:** principal component regression
- 964 • **PEDOT:** poly(3,4-ethylenedioxythiophene)
- 965 • **PLS:** partial least square
- 966 • **PLS-DA:** partial least square-discriminant analysis
- 967 • **PNN:** probabilistic neural network
- 968 • **PVC:** poly(vinyl chloride)
- 969 • **QMB:** quartz micro balance
- 970 • **RI:** rand index
- 971 • **SA:** simulated annealing
- 972 • **SAW:** surface acoustic wave
- 973 • **SFA:** simplified Fuzzy ARTMAP
- 974 • **SFA-ANN:** simplified Fuzzy ARTMAP-artificial neural network
- 975 • **SFW:** selection by Fisher weights
- 976 • **SIMCA:** soft independent modelling of class analogy
- 977 • **SLDA:** stepwise linear discriminant analysis
- 978 • **SPME:** solid-phase microextraction
- 979 • **SPME-fast GC-FID:** solid-phase microextraction fast gas chromatography-flame  
980 ionization detector
- 981 • **SPME-GC/MS:** solid-phase microextraction - gas chromatography/mass  
982 spectrometry
- 983 • **SPME-MS:** solid-phase microextraction-mass spectrometry.
- 984 • **SVM:** support vector machine
- 985 • **UV-Vis:** ultraviolet-visible
- 986 • **Vis-NIR:** visible-near infrared
- 987 • **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	$\alpha$ -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	$\alpha$ -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 <sub>2</sub> O, AgCl, Ag <sub>2</sub> CO <sub>3</sub> and Cu <sub>2</sub> O)	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 polypyrroles 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, <i>k</i> -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	$\alpha$ -Astree <sup>TM</sup> 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	$\alpha$ -Astree <sup>TM</sup> 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	$\alpha$ -Astree <sup>TM</sup> 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)

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*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.

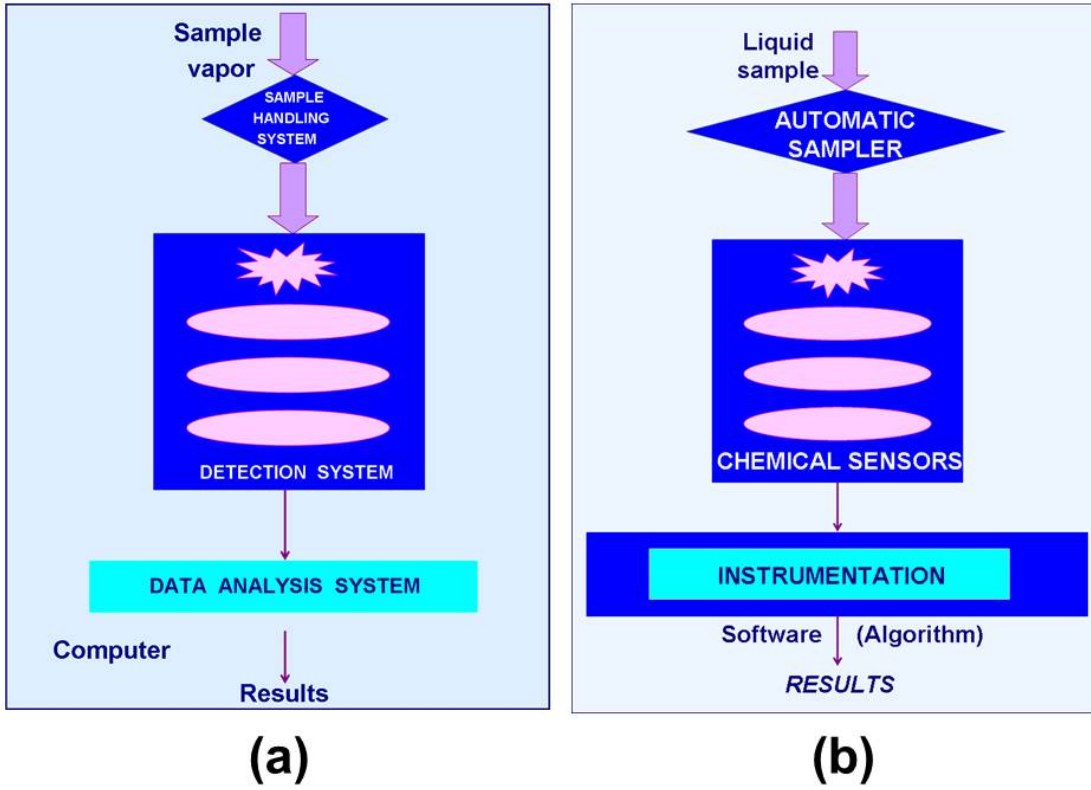
Sample	Type of study	Chemical sensors	Data processing algorithm	Ref.
Honey	Discrimination between monofloral, polyfloral honeys, sugar-syrups and honeys adulterated with sugar-syrups	Combination of data from an e-nose and an e-tongue: - Cyranose320 <sup>TM</sup> e-nose (32 non-selective polymer sensors 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 <sup>TM</sup> 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, <i>k</i> -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 <sup>TM</sup> 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 <sup>TM</sup> e-nose (10 different MOS sensors) - $\alpha$ -Astree <sup>TM</sup> 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 <sup>TM</sup> e-nose (see further details in Hong et al. (2014b)) - $\alpha$ -Astree <sup>TM</sup> 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.

1017 **Figure Captions:**

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1019 **Fig. 1** – Schematic representation of (a) an electronic nose, and (b) an electronic  
1020 tongue.



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