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Review

Honey Evaluation Using Electronic Tongues: An Overview

Ana C. A. Veloso [1,2], Mara E. B. C. Sousa [3], Leticia Estevinho [3], Luís G. Dias [3] and António M. Peres [3,4,*]

1 Instituto Politécnico de Coimbra, ISEC, DEQB, Rua Pedro Nunes, Quinta da Nora, 3030-199 Coimbra, Portugal; anaveloso@isec.pt
2 CEB-Centre of Biological Engineering, University of Minho, Campus de Gualtar, 4710-057 Braga, Portugal
3 Centro de Investigação de Montanha (CIMO), ESA, Instituto Politécnico de Bragança, Campus de Santa Apolónia, 5300-253 Bragança, Portugal; mebsousadias@gmail.com (M.E.B.C.S.); leticia@ipb.pt (L.E.); ldias@ipb.pt (L.G.D.)
4 Laboratory of Separation and Reaction Engineering—Laboratory of Catalysis and Materials (LSRE-LCM), ESA, Instituto Politécnico de Bragança, Campus Santa Apolónia, 5300-253 Bragança, Portugal
* Correspondence: peres@ipb.pt; Tel.: +351-273-30-3220

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Abstract: Honey-rich composition in biologically active compounds makes honey a food products highly appreciated due to the nutritional and healthy properties. Food-manufacturing is very prone to different types of adulterations and fraudulent labelling making it urgent to establish accurate, fast and cost-effective analytical techniques for honey assessment. In addition to the classical techniques (e.g., physicochemical analysis, microscopy, chromatography, immunoassay, DNA metabarcoding, spectroscopy), electrochemical based-sensor devices have arisen as reliable and green techniques for food analysis including honey evaluation, allowing in-situ and on-line assessment, being a user-friendly procedure not requiring high technical expertise. In this work, the use of electronic tongues, also known as taste sensor devices, for honey authenticity and assessment is reviewed. Also, the versatility of electronic tongues to qualitative (e.g., botanical and/or geographical origin assessment as well as detection of adulteration) and quantitative (e.g., assessment of adulterants levels, determination of flavonoids levels or antibiotics and insecticides residues, flavonoids) honey analysis is shown. The review is mainly focused on the research outputs reported during the last decade aiming to demonstrate the potentialities of potentiometric and voltammetric multi-sensor devices, pointing out their main advantages and present and future challenges for becoming a practical quality analytical tool at industrial and commercial levels.

Keywords: electronic tongue; potentiometry; voltammetry; lab-made devices; chemometrics; honey analysis; botanical origin assessment; geographical origin evaluation; adulteration evaluation; contaminants detection

1. Introduction

Honey is a natural sweet substance consisting of floral extracts and bee secretions, derived from pollen and nectar and produced by several species of bees [1]. Both polyfloral and monofloral honeys can be found, although the latter is usually preferred by consumers due to their rarity, unique flavors and medicinal properties, being in some cases very expensive [2]. Indeed, several biological properties and therapeutic effects of honey consumption are known [3,4]. Thus, considering the physicochemical and medicinal known properties, their potential use by the pharmaceutical and cosmetic industries has significantly increased. Honey has been used to prevent, and treat patients with, oral mucositis

resulting from radio/chemotherapy [5,6], to reduce esophagitis induced by chemoradiation therapy during the treatment of lung cancer [7], to treat skin ulcer [8,9] and to treat acute irritating cough [10]. Also, due to the recognized antibacterial activity of honey [11–14] its potential application in wound healing and tissue engineering has been studied [15,16], as for example for the treatment of burns and skin disorders [12,17]. Indeed, over the centuries, honey has been an essential ingredient in traditional medicines around the world [1]. On the other hand, the possibility of using honey as a natural sucrose-alternative sweetener in the food industry has been evaluated [18]. Thus, to fulfill the worldwide honey demand and considering the decline of the bee-keeping industry in many parts of the world [1], honey commercialization is prone to several fraudulent practices including adulteration or commercializing of mislabeled low quality honey as higher price honeys.

Several analytical techniques, some of them coupled with traditional melissopalynology analysis, together with chemometric tools have been developed and implemented for honey analysis, namely for:

(i) Verifying honey authenticity, through the identification of botanical, entomological and/or geographical origin [2,19–33].
(ii) Evaluating honey physicochemical parameters as well as antioxidant and antimicrobial activities and therapeutic properties [4,11,13–15,22,23,25,28,34–53].
(iii) Detecting insecticides, pesticides, veterinary drug or multi-class antibiotic residues in honey [54–57].
(iv) Detecting honey adulterations [24,30,58–64].

Researchers usually aim to develop highly sensitive and accurate techniques such as chromatographic methods (e.g., thin-layer chromatography; gas chromatography; high-performance liquid chromatography coupled to electrochemical detection, anion-exchange or tandem mass spectrometry; immunochromatography) and spectroscopy/spectrometry techniques (e.g., front phase fluorometric spectroscopy, near- or mid-infrared spectroscopy, nuclear magnetic resonance spectroscopy, Raman spectroscopy, quadrupole time-of-flight mass spectrometry, inductively coupled plasma atomic emission spectroscopy also referred as inductively coupled plasma optical emission spectrometry), as recently reviewed [30,64,65]. Other less common techniques have also been used for honey analysis, namely, DNA metabarcoding [2,65] or hyperspectral imaging analysis [60]. However, the majority of the analytical techniques reported for honey analysis (e.g., physicochemical characterization, biological and therapeutic activities evaluation) or, the detection of adulterations (e.g., dilution of high-value honey with water, the addition of high-sugar corn syrups or sugar-based adulterants, as well as the filtration of low-value honey to remove its source pollen and spiked with pollen from the 'desired' high-value honey), are usually time-consuming, destructive and expensive techniques, hardly applied in-situ and on-line, being far away from the economic possibilities and technical skills available at the majority of the small and medium bee-keeping industries.

The acknowledgement of this fact has recently attracted attention of the scientific research community, which are developing, building and testing fast, low-cost and user-friendly techniques such as electrochemical sensor devices for honey analysis, which require minimum sample pre-treatment steps and that may be miniaturized allowing their practical in-situ application. Thus, in the last decade electronic noses (E-noses) and electronic tongues (E-tongues) have been proposed for the classification of honeys according to botanical or geographical origins as well as to detect possible honey adulterations or the presence of atypical chemical compounds that have been intentionally incorporated in honey or derive from bee-keeping practices such as the use of non-legal antibiotics to treat different bees' diseases. The fast progress in key fields, which include artificial intelligence, digital electronic sensors design, material sciences, microcircuit design, software innovations, and electronic systems integration, has stimulated the development of electronic sensor technologies applicable to many diverse areas of human activity [66]. E-tongues are electrochemical-based analytical devices comprising single or multi non-specific cross-sensitivity, non-specific and poorly selective sensor arrays coupled to chemometric tools, aiming the establishment of predictive multivariate statistical models that can

relate the sensors signals to their analytical meaning [67–70]. Qualitative and quantitative multivariate models are developed based on the meaningful chemical fingerprint contained in the recorded electrochemical complex data profiles, which are identified after the removal of redundant data through the application of different variable selection statistical techniques (e.g., heuristic or meta-heuristic algorithms). Also, the E-tongue sensors allow the simultaneous determination of several species, with risks related to interferences, drifts and/or non-linearity, minimized or overcome by the use of advanced chemometric tools [71,72]. In some situations, the sensors with different measuring principles (e.g., potentiometry, voltammetry among others) have been applied, requiring the use of sensor data fusion techniques, taking advantage of their specific analytical characteristics, and thus, improving the dataset quality and permitting to develop more robust prediction or decision models [68].

The present work intends to summarize the work published during the last decade regarding the use of E-tongue devices for honey assessment. In fact, the versatility of E-tongues and their broad range of applicability for food analysis have been clearly described in the literature. A number of books, book chapters and review papers have been devoted to this important issue [73–78]. Also, their potential use for biomedical applications has been recently reviewed [79]. Thus, in this review a detailed survey and discussion is carried out focusing the problematic and challenge of applying E-tongues for honey evaluation; a food product highly appreciated by consumers due to the physicochemical, nutritional, biological and therapeutic known properties. First, a brief overview of the most common electrochemical techniques is made, aiming to introduce the less known reader to some theoretical basic knowledge concerning electrochemical principles, allowing a better understanding of the E-tongue potentialities as a practical tool within the food analysis field. Since the application of multi-sensor devices results in large datasets, the most used chemometric tools for extracting the valuable information contained in the electrochemical profiles recorded are briefly referred together with the usefulness of applying variable selection algorithms to avoid the use of redundant variables, minimizing the risk of overfitting and consequent overoptimistic estimation results and poor predictive performances. Also, model validation issues are addressed. Later, works reporting the use of E-tongues for honey analysis are introduced and discussed, identifying possible drawbacks and advantages, aiming to demonstrate the usefulness of these sensor-based approaches. Finally, future trends, perspectives and challenges are briefly discussed.

2. Electrochemical Sensor Devices for Honey Evaluation: Overview and Usual Chemometric Tools

In the literature, several research works reported the development and application of E-tongues based on different electrochemical techniques (e.g., potentiometry, voltammetry, impedance, etc.) as well as hybrid E-tongues, which are systems that merge different techniques by applying data fusion approaches with different abstraction levels (i.e., the way how data originated from several analytical techniques or different sources, can be merged, and form a consistent concatenated single data matrix). In which concerns honey analysis, both potentiometric and voltammetric E-tongues have been proposed and applied for both qualitative and quantitative analysis and will be the focus of the present review. At this point, it would be helpful to contextualize the E-tongue meaning. As pointed out by Kirsanov and co-workers [80], the nowadays widely used E-tongue terminology was introduced in the late 90's as an alternative to the more limited "taste sensor" term. In a broader sense, E-tongues are systems composed of one or more arrays of chemical sensors, namely electrochemical, coupled with appropriate multivariate data processing techniques. The basic concepts and principles regarding the two most common electrochemical techniques associated (i.e., potentiometry and voltammetry) to the E-tongues have been recently addressed in detail [79,81].

Similar to other analytical techniques that generate a huge amount of data per sample analysis (e.g., spectroscopy-based techniques), the full application of E-tongues-based strategies requires multivariate data analysis for pattern recognition, classification and quantification purposes. A potentiometric E-tongue comprising multi-sensors (i.e., N sensors) may generate for each sample (M samples) one potentiometric signal per sensor and sensor array (K arrays), resulting in a final

matrix of (M × KN) data. In Figure 1 a scheme is represented aiming to illustrate, as an example, the complexity of the potentiometric data matrix that can be generated by using and E-tongue device with multi-sensors.

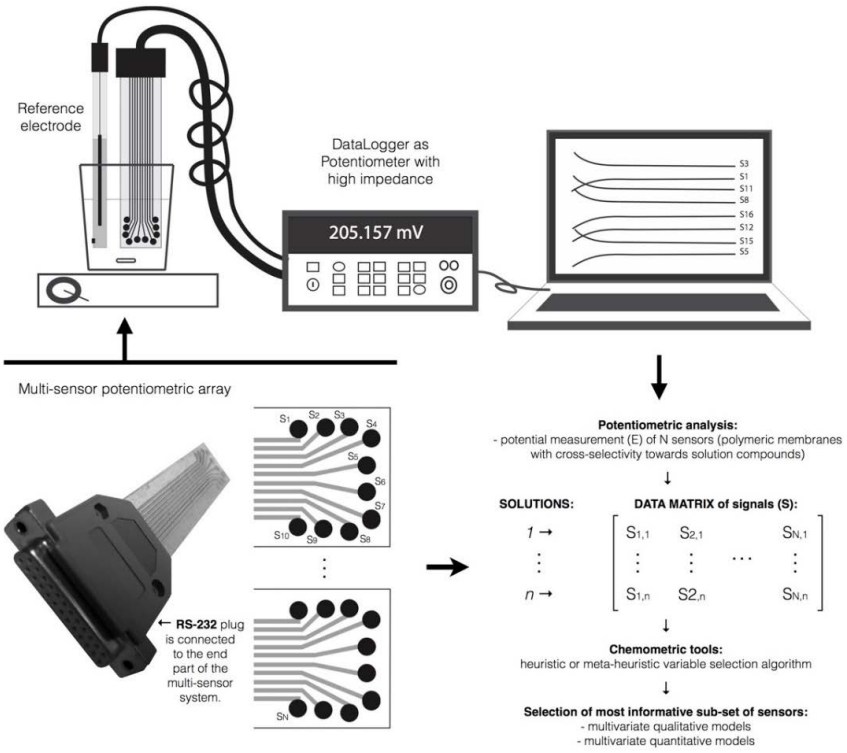


Figure 1. Database of signal profiles generated by a potentiometric E-tongue device comprising K sensor arrays each with N sensors, during the analysis of M samples.

For voltammetric E-tongues, a vector with K voltammetric measures per working electrode may be obtained either for cyclic or square-wave voltammetry. Figure 2 aims to exemplify the possible complexity when using a multi-working electrodes (multi-WEs) voltammetric E-tongue and the need of using variable selection algorithms to extract the most valuable information of the data gathered by the electrochemical device.

For both approaches, taking into account the magnitude and the complexity of the data matrices generated, the use of feature extraction strategies is required. Among them, heuristic or meta-heuristic variable selection algorithms are usually applied, aiming to reduce the number of variables that will be included in the final regression/predictive qualitative or quantitative statistical models and therefore, noise effects or overcoming issues.

Thus, usually E-tongue systems are combined with linear and non-linear qualitative and quantitative chemometric techniques, which allow verifying the capability and versatility of these electrochemical devices. Among linear pattern recognition approaches, the most common are the Principal Component Analysis (PCA), the K-Nearest Neighbor (KNN) and Linear Discriminant Analysis (LDA). For quantitative assessment, Multiple Linear Regression (MLR), Principal Component Regression (PCR) and Partial Least-Squares (PLS) models are often used. On the other hand, concerning qualitative and/or quantitative non-linear strategies, Artificial Neural Networks (ANNs) are the most applied, which include Probabilistic Neural Networks (PNNs) with Radial Basis Functions (RBF) or Feed-Forward Networks with Backpropagation (BP) learning method, Fuzzy Adaptive Resonance Theory Multidimensional Maps (ARTMAP) Neural Networks or Support Vector Machines (SVMs) are quite applied [68].

For supervised statistical classification techniques as well as for multivariate regression models, feature extraction is a key stage, allowing selecting the best set of input variables that will enable to achieve correct a posteriori classification of the data in their a priori groups or the quantitative prediction of a parameter of interest. Feature extraction tools allow identifying the meaningful variables from a set of complex data, avoiding redundancies and overcoming collinearity issues, enabling the establishment of robust mathematical models with good generalization capabilities. Among these tools, heuristic (e.g., forward, backward and stepwise techniques) and meta-heuristic (e.g., genetic algorithms, simulated annealing, etc.) variable selection algorithms are commonly applied. Moreover, to verify the predictive performance of the multivariate statistical models, in general, cross-validation variants (e.g., leave-one-out, repeated K-folds, among others) are usually used. When the dataset size allows, data split techniques (e.g., random, Kennard–Stones algorithm, etc.) are also implemented allowing establishing independent training and testing data subsets, being the latter used to evaluate the real predictive performance of the multivariate qualitative and/or quantitative models established using the former dataset.

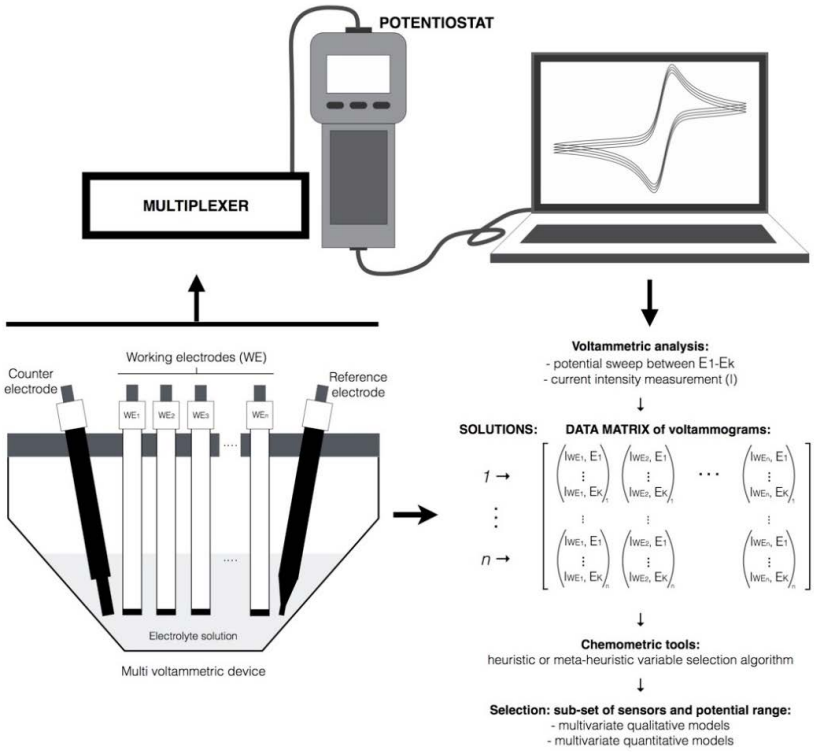


Figure 2. Database of signals profiles generated by a voltammetric E-tongue device comprising K working electrodes, during the analysis of M samples.

3. Electrochemical Sensor Devices for Honey Assessment

The broad range of applicability of E-tongue devices for food analysis has been recently reviewed by different authors [66,71,73–78,82–86]. At this point, a deeper overview is envisaged regarding the potential use of E-tongues for honey assessment, namely potentiometric and/or voltammetric based strategies.

3.1. Potentiometric Electronic Tongues

In the last decade several E-tongue potentiometric approaches have been described for honey evaluation, either based on E-tongue commercial devices (Table 1) or on home-made E-tongue multi-sensor arrays (Table 2).

Table 1. Honey evaluation using commercial potentiometric E-tongue based devices.

E-Tongue Sensors	Type of Application	Chemometric Approach and Performance	Ref.
α-Astree™: ChemFET sensor technology with 7 cross-selective liquid sensors(sensitive to ionic, neutral and chemical compounds responsible for taste) coupled with an Ag/AgCl reference electrode	Honey classification according to floral origin (Acacia, Astragali, Data, Coptis, Vitex, Motherwort, Radix Changll and Buckwheat)	- PCA - DFA - BP-ANN (correct honey classification of 98.43% according to the floral origin)	[87]
α-Astree™: ChemFET sensor technology with 7 cross-selective liquid sensors(sensitive to ionic, neutral and chemical compounds responsible for taste) coupled with an Ag/AgCl reference electrode	Honey classification according to floral origin (Acacia, Astragali, Data, Coptis, Vitex, Motherwort, Radix Changll and Buckwheat) Honey classification according to geographical origin	- PCA - CA (correct honey classification: 90% according to the floral origin and 92% according to geographical origin) - ANN (correct honey classification: 93.75% according to the floral origin and 95% according to geographical origin, for test group)	[88]
Chalcogenide-based device: Ion-selective sensors (iron, cadmium, copper, mercury, titanium, sulfur and chromium ions) plus a Ag/AgCl reference electrode	Monofloral honey classification Identification of honey adulteration	- PCA - LDA (correct classification rate of 96.7%, for LOO-CV) - PNN (90.74% of corrected classifications for test group)	[89]
Chalcogenide-based device (commercial device): Ion-selective sensors (iron, cadmium, copper, mercury, titanium, sulfur and chromium ions) plus a Ag/AgCl reference electrode	Honeys classification (different floral origins, including leaf, durian, maluka, coconut, starfruit, wax apple and tualang; or, of tualang honey from different producers)	LDA with feature extraction or selection methods: LDA plus backward selection: 86.54% of correct classification using signals from 3 sensors LDA plus forward selection: 94.23% of correct classification using signals from 2 sensors LDA plus PCA: 57.70% of correct classification using data from 4 principal components	[90]
α-Astree™: ChemFET sensor technology with 7 potentiometric chemical sensors based on chemically modified field effect transistor technology, with sensors coated with materials sensitive to the basic tastes, coupled with an Ag/AgCl reference electrode	Classification of honey samples according to their botanical (Acacia, Data, Motherwort and Buckwheat) and geographic (4 regions) origins	Pattern recognition techniques with feature extraction (12 variable features), - PCA and DFA: honey samples correctly grouped in 2-D principal components according to the floral origin or geographical origin Quantitative analysis: - PCR, PLSR and LS-SVM models (floral and geographical origins were coded, varying from 1 to 4): prediction R^2 equal to 0.7360, 0.9021 and 0.9447, respectively	[91]
α-Astree™: ChemFET sensor technology with 7 cross-selective liquid sensors(sensitive to ionic, neutral and chemical compounds responsible for taste) coupled with an Ag/AgCl reference electrode	Honey classification according to botanical origin (acacia, chestnut and honeydew) Honey analysis (physicochemical properties: electrical conductivity, acidity, water content, invert and total sugar contents)	Pattern recognition techniques: - PCA - CCA - ANN (downsizing of the model required to avoid overfitting: 100% of correct honey classification according to botanical origin, for the test group) Quantitative analysis: - ANN (models established for physicochemical parameters: 0.982 $\leq$ *R*-value $\leq$ 0.999, for the test group)	[92]

Table 1. *Cont.*

E-Tongue Sensors	Type of Application	Chemometric Approach and Performance	Ref.
α-Astree™: ChemFET sensor technology with 7 cross-selective liquid sensors(sensitive to ionic, neutral and chemical compounds responsible for taste) coupled with an Ag/AgCl reference electrode	Honey botanical origin classification (acacia, jujube and vitex varieties from different geographical origins) Identification of raw honey adulteration (honey adulterated with different percentages of corn or rice syrups)	Sensor data pretreatment: SNV, autoscale, smoothing and derivatives PCA PLSDA classification models: - botanical origin: 91.53% of correct classification for test group - adulteration identification: 100% of correct classification for test group SVMDA classification models: - botanical origin: 100% of correct classification for test group	[93]
α-Astree™: ChemFET sensor technology with 7 cross-selective liquid sensors(sensitive to ionic, neutral and chemical compounds responsible for taste) coupled with an Ag/AgCl reference electrode	Classification of different Sicilian honey varieties: chestnut, eucalyptus, sulla and orange blossom from 7 different provenances	PCA: - effective discrimination of the different honeys according to their botanical origin using the potentiometric data DFA: - overall 70.8% of correct classifications for cross-validation	[94]

ANN: artificial neural networks; BP-ANN: back propagation artificial neural networks; CA: cluster analysis; CCA: canonical correlation analysis; DFA: discriminant function analysis; KNN: k-nearest neighbor; LDA: linear discriminant analysis; LOO-CV: leave-one-out cross-validation procedure; LS-SVM: least squares-support vector machines models; PCA: principal component analysis; PLS: partial least squares models; PLSDA: partial least squares discriminant analysis models; PNN: probabilistic neural network; *R*-value: correlation coefficient; R^2: determination coefficient; SA: simulated annealing variable selection algorithm; SNV: standard normal variate; SVM: support vector machine; SVMDA models: support vector machine discriminant analysis.

Table 2. Honey evaluation using lab-made potentiometric E-tongue based devices.

E-Tongue Sensors	Type of Application	Chemometric Approach and Performance	Ref.
All-solid-state sensors device: 20 polymeric membranes (additive + plasticizer + PVC) applied on solid conducting silver-epoxy supports plus a Ag/AgCl reference electrode	Honey classification according to floral origin (Erica, Echium and Lavandula)	- PCA - LDA coupled with heuristic variable selection algorithms (stepwise, backward and forward) allowed 72% of correct classifications, for LOO-CV procedure	[95]
Multi-electrode device (metallic electrodes): Pure metals (Au, Ag and Cu) and metal compound electrodes (Cu_2O, Ag_2O, AgCl, Ag_2CO_3 and Ag_2SO_4) plus a Ag/AgCl reference electrode.	Honey classification according to floral origin (citrus, rosemary, polyfloral and honeydew—forest origin) Honey physical treatment (raw, liquefied and pasteurized honeys)	- PCA - ANN (Fuzzy-ARTMAP network; correct honey classification: 83.3% according to the floral origin and 58.3% according to physical treatment)	[96]

Table 2. *Cont.*

E-Tongue Sensors	Type of Application	Chemometric Approach and Performance	Ref.
Multi-electrode device: Pure metals (e.g., gold, silver and copper) and metallic compounds (e.g., AgO_2, CuO_2, AgCl and Ag_2CO_3) plus a Ag/AgCl reference electrode.	Honey botanical origin classification (citrus, rosemary, polyfloral and honeydew) Honey physical treatment (raw, liquefied and pasteurized honeys)	Pattern recognition techniques: - PCA - Fuzzy-ARTMAP neural networks (correct classification rates, for LOO-CV, of 94% and 42% for botanical origin and physical treatment, respectively) Quantitative analysis: - PLS (satisfactory performance for mmPfund color scale, color coordinate L* and diastase activity; $R^2 \geq 0.926$)	[97]
Multi-electrode device (metallic electrodes): Pure metals (e.g., gold, silver and copper) and metallic compounds (e.g., AgO_2, CuO_2, AgCl and Ag_2CO_3) plus a Ag/AgCl reference electrode.	Honey floral origin classification (citrus, rosemary, polyfloral and forest) Honey physical treatment (raw, liquefied and pasteurized honeys)	Fuzzy ARTMAP neural networks SFAM networks coupled or not to heuristic variable selection algorithms (stepwise, backward and forward) Better recognition performance for floral origin compared to physical treatment Maximum recognition rate of 75% for a test group	[98]
Multi-sensor arrays: 20 lipid membrane sensors and respective replicas (combinations of different lipid additives and plasticizers with PVC)	Honey classification according to color (white, amber and dark) Honey classification according to botanical origin (*Castanea* sp., *Echium* sp., *Erica* sp., *Lavandula* sp., *Prunus* sp. and *Rubus* sp.)	LDA coupled with feature selection (meta-heuristic SA variable selection algorithm): - color classification: 91% of corrected classified honey samples for LOO-CV - floral origin classification: 100% of correctly classified samples for LOO-CV after color split	[99]
Multi-sensor arrays: 20 lipid membrane sensors and respective replicas (combinations of different lipid additives and plasticizers with PVC)	Honey pollen profile assessment (i.e., quantification of pollen percentage in honey samples): monofloral honey of *Castanea* sp., *Echium* sp., *Erica* sp., *Eucalyptus* sp., *Lavandula* sp., *Prunus* sp., *Rubus* sp. and *Trifolium* sp.; and polyfloral honeys	MLR models coupled with feature selection (meta-heuristic SA variable selection algorithm): - pollen percentage quantification: MLR-SA models with mean R^2 values ($\pm$SD) between 0.91 $\pm$ 0.15 and 0.996 $\pm$ 0.010, for repeated K-fold-CV, after color split (keeping more than 10% of data for prediction purposes)	[100]
Sensor array: Eight metallic electrodes including noble metals (gold, platinum, iridium and rhodium) and non-noble metals (copper, silver, nickel and cobalt)	Honey classification: orange blossom, rosemary, thyme, sunflower, winter savory and honeydew honey. Honey physicochemical evaluation: water activity, conductivity, moisture, color and antioxidant activity.	Pattern recognition techniques: - PCA - Fuzzy ARTMAP artificial neural networks: - 100% honey type classification success for the test group Quantitative MLR models: - Predicted *R*-value of 0.9666 and 0.8959 for antioxidant activity and electrical conductivity, respectively	[101]

ANN: artificial neural networks; CA: cluster analysis; CCA: canonical correlation analysis; DFA: discriminant function analysis; Fuzzy-ARTMAP: fuzzy adaptive resonance theory multidimensional maps; K-fold-CV: repeated K folds cross-validation procedure; LDA: linear discriminant analysis; LOO-CV: leave-one-out cross-validation procedure; MLR model: multiple linear regression model; PCA: principal component analysis; PLS models: partial least squares models; R^2: determination coefficient; *R*-value: correlation coefficient; SA: simulated annealing variable selection algorithm; SD: standard deviation; SFAM: simplified fuzzy adaptive resonance theory map.

The success of this emerging electronic sensor technology is mainly related to the ability of merging different key fields like artificial intelligence, digital electronic sensors design, material sciences and electronic systems integration [66], allowing to develop fast and cost-effective complementary analytical devices which on-line and in-situ applications may be foreseen. Nevertheless, it should be remarked that few E-tongue devices are being commercialized, being in general different home-made solutions developed by each research team. The low number of commercial E-tongues may be partially attributed to the significant time effort and resources spent during calibration and recalibration of a new system as well as to the difficulty in establishing generalized models valid over various systems [102]. Indeed, commercial and home-made devices incorporate different chemical sensors, such as pure metals and metallic compounds, ion-selective sensors, cross-selective liquid sensors or lipid membranes.

The potentiometric E-tongues, coupled with different chemometric tools (e.g., PCA, LDA, ANN, etc.), have been mainly applied for qualitative honey analysis, namely as practical and successful tools for honey classification according to color, botanical or geographical origins, as well as, for honey adulteration identification [87–90,92–99,101]. Although in a few cases, some works also reported the satisfactory quantitative performance of potentiometric E-tongue devices (using, MLR, PLS and ANN models) for the determination of honey physicochemical levels or honey pollen profile assessment [92,97,100,101], confirming the broad versatility and potential of potentiometric E-tongues for honey evaluation. Some of these studies, also pointed out the advantages of using variable selection algorithms with multi-sensor potentiometric E-tongues, which allow minimizing noise effects arising from the use redundant sensor signal data [90,92,98–100].

3.2. Voltammetric Electronic Tongues

Similarly to the potentiometric E-tongues several voltammetric devices have been successfully applied for qualitative and quantitative honey analysis, using self-assembled or lab-made (with modified WEs with biofilms or nanoparticles) devices. These works usually reported the use of conventional three-electrode systems (one single WE coupled with one reference electrode (RE) and one counter electrode (CE)) or multi-WE devices (combined with one RE and one CE). In general, the WE include noble metals (e.g., platinum, gold, palladium), non-noble metals (e.g., copper, glassy carbon, nickel) and/or reactive noble metal (e.g., silver). Also, the RE is either a Ag/AgCl electrode (saturated with KCl or NaCl) or a saturated calomel electrode (SCE). The CE, is usually a platinum wire or electrode. From a qualitative (i.e., classification/discrimination) point of view, the majority of the literature works addressed the possibility of classifying honey samples according to the botanical or geographical origins as well as to identify honey adulterations or the adulteration level [81,91,103–113]. A substantial number of works reported the satisfactory quantitative performance of voltammetric E-tongues used to predict chemical and biochemical honey composition as well as the levels of adulterants and/or contaminants [91,104,106,113–125]. As can be easily inferred from Table 3 (commercial devices) and Tables 4 and 5 (self-assembled lab-made conventional or multi-sensors devices), the use of voltammetric E-tongues for honey analysis is a more recent practice (from 2011) compared to the potentiometric approaches (from 2008) being largely used together with different multivariate statistical techniques (e.g., multiple linear regression models (MLRM), PLS, ANN, among others) as successful quantitative analytical tools. Only one work reported the use of a commercial conventional three-component device [114]. In contrast all the other studies, reported, as previously stated, the development and/or use of lab-made devices comprising a single WE [81,105–111,115–125] or more WEs [91,103,104,112,113], some of them modified incorporated porous films or nanoparticles [99–101,115,117–125]. Within these applications, different voltammetric techniques have been applied namely cyclic voltammetry (CV, the most common), square-wave voltammetry (SWV) and square-wave cathodic stripping voltammetry (SWCSV), differential pulse voltammetry (DPV) and multifrequency large amplitude pulse voltammetry (MLAPV) as well as linear sweep voltammetry (LSV). Overall, all the above-mentioned works demonstrate the versatility and feasibility of applying voltammetric E-tongues as alternative/complementary analytical tool for honey analysis, allowing in some cases in-situ assays due to the potential portable nature of these electronic device [105].

Table 3. Honey evaluation using commercial voltammetric E-tongue based device.

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
CHI660D electrochemical analyzer: - one WE (glassy carbon) - one RE (saturated calomel electrode, saturated with KCl solution) - one CE (platinum wire)	CV	Detection of honey adulteration with rice syrups	Pattern recognition techniques with feature extraction (12 variable features), - PCA: pure and adulterated honey samples were completely distinguished using the two first PCs - PCA-LDA and LDA: recognition rates of 100% for both calibration and prediction sets Quantitative analysis, - MLR model with 12 PCs: $R_{prediction}$ = 0.898 - PCR model with 3 PCs $R_{prediction}$ = 0.881 - PLS with 4 PCS: $R_{prediction}$ = 0.898	[114]

CE: counter electrode; CV: cyclic voltammetry; LDA: linear discriminant analysis; MLR model: multiple linear regression model; PCA: principal component analysis; PCR: principal components regression; PCs: principal components; PLS: partial least squares model; R^2: determination coefficient; RE: reference electrode; *R*-value: correlation coefficient; WE: working electrode.

Table 4. Honey analysis using self-assembled lab-made conventional three-electrodes voltammetric devices.

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- one WE: platinum electrode - one RE: Ag/AgCl saturated KCl - one CE: platinum electrode	CV	Discrimination of monofloral honeys: Eucalyptus, Til, Leechu and Khalisa	PCA used as a pattern recognition classifier: successful recognition of floral origin	[107]
- one WE: platinum electrode - one RE: Ag/AgCl saturated KCl - one CE: platinum electrode	CV	Identification of floral honey origin: Eucalyptus *(Eucalyptus globulus)* Til *(Sesamum indicum)* Leechu *(Litchi chinensis)* Khalisa (regional name)	- PCA (with relative $scale_2$ method): Til and Eucalyptus honeys grouped into two distinct clusters while honey samples from Kholisa and Leechi overlapped - LDA (with autoscale method): 100% of corrected classified samples (original grouped samples) - BP-MLP neural network (with range scale method): 93.42% of correct classifications for a validation dataset - RBF neural network (with baseline subtraction method): 82.50% of correct classifications for a validation dataset	[108]
- one WE: NiO/Nps modified carbon paste electrode - one RE: Ag/AgCl saturated KCl - one CE: platinum wire	CV	Floral characterization of honey (Eucalyptus, Til, Lecchi, Pumpkin, Mustard and polyfloral) with the same geographical origin	PCA: honey samples correctly grouped according to the floral origin in 2-D dimensional planes, being polyfloral honey samples, a mixture of eucalyptus and mustard honeys, grouped closely to mustard and eucalyptus honey groups	[109]

Table 4. *Cont.*

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- one WE: gold electrode - one RE: saturated calomel electrode - one CE: platinum electrode	DPV LSV CV SWV	Detection of honey adulteration with sugar syrups Quantification of adulteration percentage	Pattern recognition methods: - PCA: allows distinguishing honey samples according to the adulteration percentage (from 0% up to 70%) - RBF: 83.33% of honey samples correctly classified according to adulteration level (from 0% to 70%) - FKNN: 88.89% of honey samples correctly classified according to adulteration level (from 0% to 70%) - Fuzzy ARTMAP: 94.40% of honey samples correctly classified according to adulteration level (from 0% to 70%) Quantitative analysis: - PLS: honey adulteration percentage satisfactorily predicted (R-value = 0.8442)	[106]
Portable device with integrated chemometrics tools: - one WE: gold disk electrode - one RE: Ag/AgCl electrode - one CE: gold disk electrode	CV	Classification of honey samples according to their botanical (*quince, orange, and coffee*) and geographic 3 regions) origins	PCA (four-components model based on 408 variables, with decomposed signals): successfully applied to fingerprint honey samples according to their botanical and geographic origins.	[105]
- one WE: silver electrode - one RE: Ag/AgCl electrode - one CE: platinum electrode	CV	Differentiation of monofloral honeys according to botanical origin (*Castanea* sp., *Echium* sp., *Rubus* sp., *Lavandula* sp., *Prunus* sp., *Erica* sp., *Trifolium* sp.) Monofloral honey differentiation according to color scale	Qualitative approach: - honey samples from the same color group, anodic peak currents and anodic areas differ with floral origin of honeys - similar oxidation potentials and overall voltammetric profiles observed for *Lavandula* sp. honeys, regardless honey color - anodic peak current and anodic curve area of *Lavandula* sp. honeys increase with darkness increasing of *Lavandula* sp. honeys (mmPfund values versus anodic peak current intensity, R = 0.9680)	[81]
- one WE: glassy carbon electrode disk - one RE: KCl saturated calomel electrode - one CE: platinum foil	CV SWV	Determination of antiseptic agents (eugenol, carvacrol and thymol) in honey samples	Multivariate calibration tools developed based on SWV data, with baseline correction and signal alignment: - PLS: poor predictive capability (validation set: $0.19 \leq R^2 \leq 0.76$ with relative errors of prediction greater than 30%) - ANN (feed-forward network with Levenberg-Marquardt back propagation training): - validation set: $0.968 \leq R^2 \leq 0.997$ with relative errors of prediction of 5–7% and limits of detection between 0.010 and 0.240 mg L^{-1}	[116]
- one WE: modified platinum thin-film microelectrode with o-phenylenediamine - one RE: platinum electrode - one CE: platinum electrode	CV SWV	Determination of antibiotics in honey: chloramphenicol (CAP)	CAP dynamic range: from 0.9 to 10 nM (R = 0.992) CAP detection limit: 0.39 nM CAP recovery assays: from 89 to 107.3%	[117]

Table 4. *Cont.*

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- one WE: modified glassy carbon electrode using an isoreticular carbon porous metal-organic framework - one RE: saturated calomel electrode - one CE: platinum wire	SWV	Determination of antibiotics in honey: chloramphenicol (CAP)	CAP dynamic range: from 10 nM to 1 μM (R^2 ≥0.991) CAP detection limit: 2.9 nM CAP (0.1 to 1.5 μM) recovery assays: from 96 to 110%	[121]
- one WE: bare glassy carbon electrode or modified electrode (MIL-101(Cr)/XC-72/GCE sensor) - one RE: saturated calomel electrode - one CE: platinum wire	CV DPV	Determination of antibiotics in honey: chloramphenicol (CAP)	CAP dynamic range: from 10 nM to 20 μM (R = 0.985) CAP detection limit: 1.5 nM CAP (0.2 to 1.0 μM) recovery assays: from 95 to 101%	[118]
- one WE: functionalized carbon black nanospheres hybrid with MoS2 nanocluster - one RE: Ag/AgCl saturated KCl - one CE: platinum wire	CV DPV	Determination of antibiotics in honey: chloramphenicol (CAP)	CAP dynamic range: from 0.015 to 1370 μM (R^2 = 0.989) CAP detection limit: 0.002 μM CAP (25 and 50 μM) recovery assays: from 93.0 to 96.2% CAP sensitivity: 3400 $\mu A \mu M^{-1} cm^{-2}$	[120]
- one WE: glassy carbon electrode modified (or not) with ordered mesoporous carbon@polydopamine and β-cyclodextrin - one RE: saturated calomel electrode - one CE: platinum wire	CV SWV	Determination of antibiotics in honey: chloramphenicol (CAP)	CAP dynamic range: from 0.5 μM to 0.5 mM (R^2 = 0.9992) CAP detection limit: 0.2 μM CAP recovery assays (5 to 50 μM): 80.0 to 93.0%.	[122]
- one WE: glassy carbon electrode modified with electro-polymerized poly(pyrrole-3-carboxy acid) and electrochemically reduced graphene oxide - one RE: saturated calomel electrode - one CE: platinum electrode	CV DPV	Determination of antibiotics in honey: streptomycin (STR)	STR dynamic range: 2 nM to 1 μM (R > 0.99) STR detection limit: 0.5 nM STR (25 nM to 1 μM) recovery assays: 96 to 104%	[123]
- one WE: antimony film coating a glassy carbon electrode - one RE: Ag/AgCl saturated NaCl - one CE: platinum wire	CV SWCSV	Determination of antibiotics (tetracyclines) in honey samples	Quantitative analysis using a LR model based on SWCSV: - linear range: 0.40–3.00 μM - sensitivity: 1.46 μA μM^{-1} - detection limit: 0.15 μM - recoveries: from 91.81% to 109.69%	[115]
- one WE: ZrO_2NPs with modified carbon paste electrode and paraffin oil - one RE: Ag/AgCl saturated KCl - one CE: platinum wire	CV	Floral characterization of honey with different floral origins (Eucalyptus, Til, Pumpkin and Mustard) from different apiaries of the same geographical region	PCA (data preprocessed: scaled): honey samples correctly grouped according to the floral origin in 2-D dimensional planes	[110]
- one WE: carbon paste electrode modified with zinc oxide nanoparticles - one RE: Ag/AgCl saturated KCL - one CE: platinum wire	CV	Discrimination of the floral origin of honey: *Eucalyptus globulus*, *Cucurbita maxima*, *Litchi chinensis*, *Brassica juncea*, *Sesamum indicum*	Pattern recognition techniques: - PCA: allowed the discrimination among the different floral types - ANN (BP-MLP and RBF): classification model with more than 90% accuracy (86 to 97% of correct classification according to each honey floral type)	[111]

Table 4. *Cont.*

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- one WE: carbon paste electrode modified with magnetic Fe_3O_4@NiO core/shell nanoparticles - one RE: Ag/AgCl electrode - one CE: platinum rod	CV DPV	Determination of Quercetin (Q, flavonoid) and Tryptophan (Trp, essential aminoacid) in honey samples	Q dynamic range: 0.08–60 μM (R^2 = 0.9845) Trp dynamic range: 0.1–120 μM (R^2 = 0.9893) Q detection limit: 2.18 nM Trp detection limit: 14.23 nM	[124]
- one WE: modified nanohybrid glassy carbon electrode with highly porous polypyrrole (MIP/MIL-101 (Cr)/MoS2/GCE sensor) - one RE: saturated calomel electrode - one CE: platinum foil	CV DPV	Determination of Quercetin (Q, flavonoid) in honey samples	Q dynamic range: 0.1 to 700 μM (R^2 = 0.999) Q detection limit: 0.06 μM in phosphate buffer solution (PBS, pH = 3.5) Q recovery assays (1.1 to 1.5 μM): 97.3 to 101.3%	[119]
- one WE: glassy carbon electrode modified with ß-cyclodextrin and graphene oxide - one RE: Ag/AgCl electrode - one CE: platinum wire	CV SWV	Determination of neonicotinoids (insecticides): imidacloprid (IMP), clothianidin (CLT) and thiamethoxam (TMX)	IMP dynamic range: 0 to 165 μM CLT dynamic range: 7.5 to 80 μM TMX dynamic range: 10 to 70 μM IMP detection limit: 8.92 μM CLT detection limit: 4.72 μM TMX detection limit: 7.45 μM Recovery assays (added 20 μM): 108.75, 107.75 and 116% for IMP, CLT and TMX, respectively.	[125]

ANN: artificial neural networks; BP-MLP: back-propagation multi-layer perceptron algorithm; CE: counter electrode; CV: cyclic voltammetry; DPV: differential pulse voltammetry; FKNN: fuzzy k-nearest neighbor algorithm; Fuzzy ARTMAP: Fuzzy adaptive resonance theory multidimensional map; LDA: linear discriminant analysis; LR: linear regression; LSV: linear sweep voltammetry; Nps: nanoparticles; PCA: principal component analysis; PLS: partial least squares; R^2: determination coefficient; RBF: radial basis function; RE: reference electrode; *R*-value: correlation coefficient; SWCSV: square wave cathodic stripping voltammetry; SWV: square wave voltammetry; WE: working electrode.

Table 5. Honey analysis using self-assembled lab-made multi-working electrodes voltammetric E-tongues.

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- six WEs: gold, silver, platinum, palladium, tungsten, and titanium - one RE: Ag/AgCl saturated KCl - one CE: platinum electrode	MLAPV	Classification of honey samples, from the same geographical area, according to their botanical (*Acacia, Astragali, Buckwheat, Coptis, Data, Motherwort and Vitex*)	Pattern recognition techniques with feature extraction: - PCA, DFA and CA: the three methods based on the two databases have similar discrimination performances and the difference between the two databases has no effect to the separation ability	[112]
- six WEs: gold, silver, platinum, palladium, tungsten and titanium - one RE: Ag/AgCl saturated KCl - one CE: platinum electrode	MLAPV	Classification of honey samples according to their botanical (*Acacia, Data, Motherwort and Buckwheat*) and geographic (4 regions) origins	Pattern recognition techniques with feature extraction (12 variable features): - PCA and DFA: honey samples correctly grouped in 2-D principal components according to the floral origin or geographical origin Quantitative analysis: - PCR, PLSR and LS-SVM models (floral and geographical origins were coded, varying from 1 to 4): prediction R^2 equal to 0.8924, 0.9887 and 0.9985, respectively	[91]

Table 5. *Cont.*

E-Tongue Sensors	Technique	Type of Application	Chemometric Approach and Performance	Ref.
- seven WEs: noble metals (platinum, gold, palladium), non-noble metals (copper, glassy carbon, nickel) and reactive noble metal (silver) - one RE: Ag/AgCl electrode - one CE: platinum electrode	CV	Classification of honeys according to geographical (9 countries) and botanical (*Lime green, Thyme, Rosemary, Natural blueberry, Saracen, Carob, Jujube, Mountain, Eucalyptus, Spurge, Orange and Polyfloral*) origins Detection of honeys' adulteration with sugar syrups	Pattern recognition techniques with feature extraction: - PCA: allowed to correctly discriminate honeys according to geographical or botanical origins, as well as to recognize all adulteration levels. - SVM: 100% success rate in the recognition of honeys of different geographical origins as well as of different botanical origins, for LOO-CV, as well as for the identification of adulterated honey - HCA: no errors or misclassifications of honey samples according to geographical or botanical origins as well as to distinguish between different classes of adulterated honey	[103]
- seven WEs: noble metals (platinum, gold, palladium), non-noble metals (copper, glassy carbon, nickel) and reactive noble metal (silver) - one RE: Ag/AgCl electrode - one CE: platinum electrode	CV	Classification of polyfloral honeys according to geographical origin (2 countries: Morocco and France) and type Quantitative prediction of biochemical and physicochemical profiles of honey samples (protein content, color intensity, phenols content, lactonic acidity, free acidity, total acidity, HMF (hydroxymethylfurfural) content, reducing sugars, total sugar, sucrose content)	Pattern recognition techniques with feature extraction (3 variable features): - PCA: successful discrimination of honeys according to geographical or botanical origins. - SVM: 100% success rate in the recognition of honeys of different geographical origins as well as of different botanical origins, for LOO-CV. - HCA: no errors or misclassifications of honey samples according to geographical or botanical origins. Quantitative analysis: - PLS: $0.821 \leq R^2 \leq 0.998$ $0.015 \leq$ NRMSE ≤ 0.184 $2.306 \leq$ RPD ≤ 7.658	[104]
- four WEs: iridium, rhodium, platinum, gold - one RE: saturated calomel electrode - one CE: stainless steel circular piece	PV	Detection of honey adulteration with sugar syrups: monofloral honeys (heather, orange blossom and sunflower), syrup (rice, barley and corn), and adulterated honey (2.5, 5, 10, 20 and 40% of syrup)	Pattern recognition techniques: - PCA: voltammetric data allowed distinguishing pure honey, syrup, and different levels of adulterants Quantitative analysis: - PLS analysis: allowed to predict the level of the adulterants in each honey (sunflower honey adulterated with barley, corn or brown rice syrup: $0.949 \leq R^2 \leq 0.997$; orange blossom honey adulterated with barley, corn or brown rice syrup: $0.879 \leq R^2 \leq 0.993$; and, heather honey adulterated with barley, corn or brown rice syrup: $0.763 \leq R^2 \leq 0.997$)	[113]

CA: clusters analysis; CE: counter electrode; CV: cyclic voltammetry; DFA: discriminant actor analysis; HCA: hierarchical cluster analysis; LOO-CV: leave-one-out cross-validation procedure; LS-SVM: least squared-support vector machines; MLAPV: multifrequency large amplitude pulse voltammetry; NRMSE: normalized root-mean-square error; PCA: principal component analysis; PCR: principal component regression; PLS: partial least squares; PLSR: partial least squares regression; PV: pulse voltammetry; R^2: determination coefficient; RE: reference electrode; RPD: ratio of performance to deviation (ratio of the standard error in prediction to the standard deviation of the samples); SVM: support vector machines; WE: working electrode.

4. Advantages, Limitations and Drawbacks of the Two Most Common Electronic Tongues Variants

As pointed out (Tables 1–5), E-tongues have suffered an increasing application in honey screening analysis, which broad number of qualitative and quantitative applications, reported in the literature, are summarized in Figures 3 and 4, respectively.

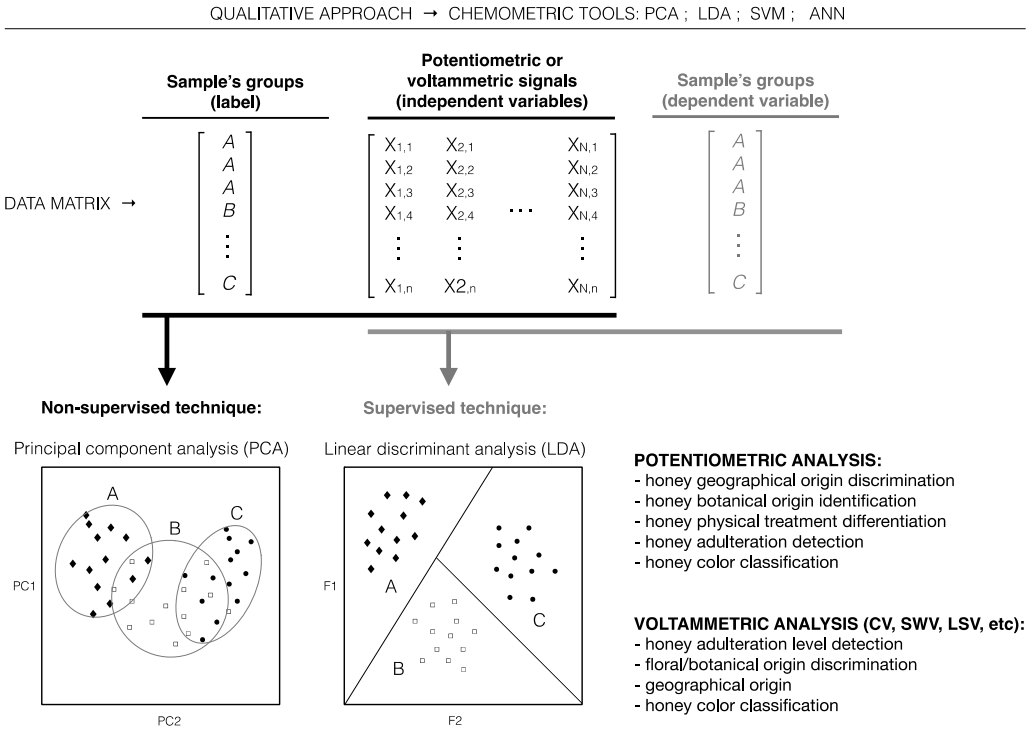
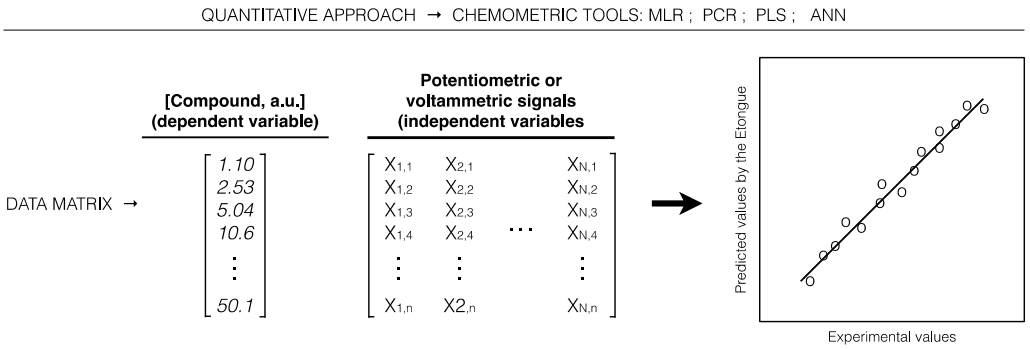


Figure 3. Common qualitative applications of potentiometric and voltammetric E-tongues coupled with chemometric tools for honey analysis, according to the literature survey.

These electrochemical devices have emerged as an innovative sensing technology, supported by the development of different scientific areas such as artificial intelligence, digital electronic sensors design, material sciences, microcircuit design, software innovations, and electronic systems integration. Also, the increase interest relies on several known advantages of electrochemical devices over other conventional analytical methods. Besides being fast, flexible, cost-effective, sensitive, accurate and user-friendly techniques, the use of E-tongues does not require specialized staff neither complex sample pre-treatments. In fact, some potentiometric multi-sensor arrays may be directly immersed into the honey sample, allowing a direct measurement and, in other cases (depending on the sample's viscosity); it is only necessary to previously dissolve a known mass of honey into a pre-defined volume of distilled water, leading to the change of the membrane potentials in response to the different sample chemical compositions [94,95,99,100]. In some cases, prior to the potentiometric analysis the E-tongue sensors may need to be conditioned and calibrated using, in general, an aqueous acid solution [94]. Regarding the voltammetric devices, the honey analysis requires its previous dissolution using an electrolyte solution (e.g., KCl or phosphate buffer saline solution, PBS) [120,124] or in some specific cases, extraction/centrifugation steps [119]. As pointed out by several researchers, both methodologies would require some special washing procedures, between the measurements or after a set of assays, in order to remove all sample leftovers from the sensors surface membranes, ensuring stable and repeatable signal profiles [103,113], although the voltammetric devices may also require the electrodes surfaces to be polished. Depending on the type of sample, sensor membranes may be negatively or positively charged and so, an acid or basic washing solutions are usually

used, respectively, although in some cases only a washing step with ultrapure water is reported. Voltammetric analysis may further require a deoxygenation step by purging the sample solution with an inert gas like nitrogen, turning out into a more complex sample pretreatment compared to the potentiometric analysis [123]. In general, as reported in the literature, both E-tongues show long-term electrochemical response stability and repeatability over time and after storage, being potentiometric devices be more prune to signal drift issues, which may be minimized or overcome by the washing procedures or by the subsequent use of statistical treatments for signal drifts corrections [126–131]. Moreover, the majority of the assays can be carried out at room temperature. Furthermore, the E-tongue profiles together with chemometric tools allow assessing honey physicochemical and biochemical parameters using the electrochemical fingerprints recorded in a single experimental run, which avoids the need of applying several different analytical techniques. Additionally, E-tongues may be easily miniaturized, handled and cleaned, have low power consumption as well as an intrinsic portable characteristic enabling in-situ and continuous analysis, even in harsh industrial environments.



POTENTIOMETRIC ANALYSIS:
- assessment of physicochemical properties (electrical conductivity, acidity, water activity, invert and total sugar contents, moisture, color intensity and antioxidant activity)
- pollen profile assessment

VOLTAMMETRIC ANALYSIS (CV, SWV, LSV, etc):
- Assessment of honey adulterants contents
- detection/quantification of antiseptic agents in honey samples
- detection/quantification of antibiotics in honey samples
- detection/quantification of flavonoids and amino acids in honey samples
- detection/quantification of insecticides in honey samples
- assessment of biochemical and physicochemical profiles of honey samples (protein content, color intensity, phenols content, lactonic acidity, free acidity, total acidity, HMF content, reducing sugars, total sugar, sucrose content)

Figure 4. Common quantitative applications of potentiometric and voltammetric E-tongues coupled with chemometric tools for honey analysis, according to the literature survey.

Nevertheless, several authors still have concerns regarding the lack of specific odor and taste sensors or the difficulty validating the multivariate models established due to the lack of establishing large databases [74]. To address these concerns, new sensors with improved selectivity, including nanosensors and biosensors, have been the focus of several research groups. Also, efforts are being carried out aiming to establish international databases that would allow assembling a large number of well characterized samples to carry out an appropriate training and validation [74]. The occurrence of signal drifts and/or noise effects when the electrochemical analysis is carried out during a long period of time is also a problem that has precluded a broad adoption of E-tongues as routine analytical tools [126]. Indeed, E-tongue's calibration lifetime is typically limited due to the changes of sensor materials related to several physical and chemical phenomena like adsorption of sample components, temperature deviations, surface chemical reactions, among others [126]. Also, it is known that even if two electrochemical devices are sensitive towards the same family of chemical compounds, the different devices can hardly operate in the framework of a single unified calibration model, which would

enable interpreting simultaneously the responses of both systems [102]. Several strategies have been recently developed aiming to overcome these drawbacks. Recently, it has been experimentally verified the feasibility of calibration transfer between voltammetric and potentiometric multi-sensor arrays, which showed the possibility of transforming potentiometric data into voltammetric format, and vice versa, allowing modeling a system response using multivariate regression models built with data from another type of multi-sensor system [102]. Mathematical sensor drift correction procedures have been successfully used to overcome problems related to sensor readings' drift that can invalidate corresponding multivariate calibrations [126–131]. These mathematical procedures minimize the need of E-tongue frequent recalibrations and thus allow maximizing the related investment of time and experimental effort, being of utmost importance for unique and expensive samples. In fact, these works pointed out that it is possible to extend the calibration lifetime in multi-sensor analysis of real complex samples by mathematical drift correction, instead of trying to take into account these issues within the framework of each regression model. Furthermore, as pointed out by Panchuk and co-workers [126], the particular standardization method should be used taking into account the sensor array structure and the analytical task. If a strong correlation in sensor responses towards target parameter is expected, the use of multivariate standardization methods is recommended. If the sensors comprised in the E-tongue show dissimilar signal profiles, univariate single sensor standardization could be the right choice.

The previous discussion clearly points out the difficulty in choosing one E-tongue approach over the other, for honey analysis. Indeed, both potentiometric and voltammetric devices show emerging advantages, posing some limitations and disadvantages. Still, it could be concluded that potentiometric E-tongues may deliver a broader chemical fingerprint of a specific honey sample, since they may detect the presence of any chemical compound that may impose a potential shift of the sensors' membranes due to, for example, electrostatic or hydrophobic interactions [132], not being limited to the analysis of redox chemical compounds. Also, in general, potentiometric devices require less complex sample pre-treatments compared to the voltammetric ones. On the contrary, potentiometric sensor arrays are mainly used for qualitative evaluations, allowing the richness information of the voltammograms a deeper analysis including both qualitative and quantitative perspectives. Moreover, signal drifts are usually more relevant in potentiometric analysis requiring subsequent complex statistical analysis. Thus, the although the capabilities and advantages of E-tongues for honey analysis is evident and straightforward for the majority of the researchers within the electrochemistry field, it is not an easy task to prioritize the best strategy, which will mostly depend on the researcher familiarity with this subject as well as of the equipment availability.

Finally, the overall analytical (qualitative and quantitative) satisfactory performance of E-tongue systems together with the possibility of overcoming issues such as signal's drifts, may envisage a broader routine application in day-to-day laboratory and industrial practices.

5. Conclusions

In conclusion, this review examined and demonstrated the theoretical and practical feasibility and versatility of both potentiometric and voltammetric E-tongues for botanical and geographical origin identification and contaminant detection as well as pollen profile assessment and chemical composition determination. The vast number of research works available in the literature clearly pointed out that these devices are very promising tools for honey analysis, profiting of their portability, miniaturization and possible compatible with smartphone technology, in-situ and on-line operation as well as of the user-friendly and green potentialities. Furthermore, these devices may be very effective tools especially in combination with appropriate chemometric techniques, with the use of improved feature extraction techniques for electronic sensor response analysis, which is a key issue.

Nevertheless, more research is required to develop and take full advantage of E-tongue instruments, bringing them to the full potential of capabilities for industrial applications, overcoming typical concerns of the real world; namely, contributing to shortening the distance

between the optimism of the researchers and the skepticism of the industry and retailers. At present, the main challenge relies in reaching the market, which is obvious considering the scarcity of commercially available E-tongue devices. Indeed, the key challenge would be to build E-tongues with repeatable electrical or electrochemical properties, negligible ageing and temperature effects, as well as the irreversible binding of substances on the materials used as sensing units in some applications., requiring sensor units' replacement and thus, leading to time-consuming re-calibration steps. These drawbacks have prevented the wide use of E-tongues in the market. So, in the future, strategies must comprise the design of arrays formed by new sensing (nano)materials with improved selectivity and sensitivity.

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