



# Analysis of spices & herbs and its phenolic content by means of an electronic tongue

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## ABSTRACT

The drying process to which herbs and spices are submitted has a significant impact on the flavour and quality of those as many compounds might be degraded or transformed. This is especially relevant for most aromatic and bioactive compounds (e.g., phenolic compounds), which values have been reported to diminish during the drying process. In this direction, herein we explore the usage of an electronic tongue (ET) as a tool to assist on the monitoring of herbs and spices quality. The proposed approach is based on the use of a simple extraction method prior to the analysis of the samples with an array of voltammetric sensors in combination with pattern-recognition methods. Firstly, a partial least squares regression (PLS) model was built, which allowed the quantification of the total phenolic content ( $R^2 > 0.985$ ), an important indicator of their preservation. Subsequently, principal component analysis (PCA) of the obtained responses allowed also to qualitatively discriminate the different herbs and spices.

## 1. Introduction

According to the United States Department of Agriculture (USDA) (United States Department of Agriculture, 2023; Vázquez-Fresno et al., 2019), spices and herbs are defined as plant derived substances that add flavour to any dish. Despite both names might be incorrectly used indistinguishably to refer to aromatic parts of plants, the term herb refers to the leafy part of a plant, while spices derive from different parts of the plant (e.g. seeds, leaves, flowers, fruits).

Unlike vegetables and fruits, which are consumed mostly due to their macronutrients, herbs (including spices) are mainly used in very small quantities to flavour, colour or garnish foods. This is because herbs and spices contain chemical compounds that impart unique sensory attributes (Siddiq & Uebersax, 2018; Vázquez-Fresno et al., 2019). Such compounds include terpenes, alkaloids, flavonoids, phenolic compounds (including polyphenols) and salicylates, between others (Shan, Cai, Sun, & Corke, 2005). Beyond the clear sensory impact, herbs and spices have been reported to reduce lipid oxidation during heating of different food products (de Oliveira et al., 2022; Van Hecke, Ho, Goethals, & De Smet, 2017). This is because many of those also exhibit antioxidant activity as scavengers of several reactive oxygen species (ROS, e.g., polyphenols) or

provide anti-inflammatory and pain-relieving benefits (e.g., salicylates) (Martínez-Tomé et al., 2001; Swain, Dutton, & Truswell, 1985; Vázquez-Fresno et al., 2019). For this reason, many herbs are also used for medicinal purposes, or even for the preparation of fragrances.

Although herbs and spices might be used fresh, they are most commonly sold in dried form (either whole or powdered). However, the drying process has a significant impact on the flavour and quality of those, as many compounds might be degraded or transformed during the process. This is especially relevant for most aromatic and bioactive compounds (e.g., phenolic compounds), whose value has been reported to diminish during the drying process (Mouhoubi et al., 2022; Thamkaew, Sjöholm, & Galindo, 2021). In this direction, different methods for drying have been evaluated over the last years in order to better preserve the initial properties of the different herbs (Thamkaew et al., 2021). However, it has also been found that the extent of the chemical changes and the optimum drying conditions are very dependent on the botanical structure of the different herbs (the type of herb), the harvesting season, the age of the plant or the storage conditions, between others. Alternatively, extraction of the essential oils has also been evaluated, but again, the composition of the extracts is greatly affected by the extraction method, both qualitatively (in terms of the compounds

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present) and quantitatively (concentration of those) (Giacometti et al., 2018; Thamkaew et al., 2021). Consequently, to ensure its optimal quality, the evaluation of each specific pre-drying and drying method for each type of herb is required constantly.

However, as happens in many food products, assessment of their quality is rather subjective. For example, the quality of medical dried herbs is mainly defined by the content of bioactive compounds, while the quality of culinary dried herbs is also highly influenced by their colour and fresh-like characteristic aroma (Thamkaew et al., 2021). From an analytical point of view, quality has to be bound to a measurable characteristic or concentration of a (or some) specific compound(s) (Cetó, Gonzalez-Calabuig, Capdevila, Puig-Pujol, & del Valle, 2015). For this reason, over the last years, quality control approaches are shifting from compound oriented to pattern-oriented strategies (Cavanna, Righetti, Elliott, & Suman, 2018; Cetó & Pérez, 2020). That is, rather than focusing on the detection of a few specific components which have been previously identified, targeting all detectable components of the sample. In this direction, electronic tongues (ETs) may offer an interesting alternative (Cetó, Voelcker, & Prieto-Simón, 2016; Ciosek & Wroblewski, 2007; del Valle, 2010; Lu, Hu, Hu, Li, & Tian, 2022).

Based on the aforementioned, herein we explore the usage of an ET as a tool to assist on the identification and the monitoring of herbs' quality. ETs are biomimetic systems based on the combination of multi-sensor devices with some pattern recognition and/or multivariate calibration method applied to the analysis of liquids (Cetó et al., 2016; del Valle, 2010). The proposed approach is based on the use of a simple extraction method prior to the analysis of the samples with an array of voltammetric sensors, which in combination with partial least squares (PLS) regression and principal component analysis (PCA) allowed evaluating herbs' quality. Concretely, quantitative analysis of total phenolic composition of different herbs was first evaluated as an indicator of its preservation, as it has been widely reported that its composition changes significantly during the drying process. Next, voltammetric fingerprints were submitted to PCA to also assess the potential of the ET to qualitatively discriminate the different herbs and its drying.

## 2. Material and methods

### 2.1. Reagents and materials

All reagents used were analytical reagent grade and all solutions were prepared using ultrapure water (18.2 MΩ cm) from a Milli-Q system (Millipore, Billerica, MA, USA).

Potassium hydrogenphosphate, potassium dihydrogenphosphate and potassium chloride, which were used for the preparation of the pH 7.0 phosphate buffer, were purchased from Merck (Darmstadt, Germany). Ethanol 96% (v/v) (Scharlau; Barcelona, Spain) was used for the extractions. For the spectrophotometric measurements, Folin-Ciocalteu's reagent and sodium carbonate obtained from Panreac Química (Barcelona, Spain) as well as gallic acid (Sigma-Aldrich, St. Louis, MO, USA) were used. Lastly, for the preparation of the electrodes, Epotek H77 resin and its corresponding hardener were purchased from Epoxy Technologies (Billerica, MA, USA), graphite powder (particle size <50 μm) was obtained from BDH (BDH Laboratory Supplies, Poole, UK), Prussian blue was obtained from Acros Organics (Geel, Belgium) and nanoparticles of bismuth (III) oxide, copper (II) oxide, tin (IV) oxide and titanium (IV) oxide were obtained from Sigma-Aldrich.

### 2.2. Herb samples

A total of 53 samples from different spices and herbs were considered for this study. Each of the samples was from a different producer and obtained from several local shops in Catalonia, and consequently, produced under standards set by local regulations.

On the one side, a first set of samples was purchased to evaluate the

potential of the ET to quantitatively assess the total phenolic content, regardless of the sample type; hence, widening the number of different herbs & spices considered. On the other hand, a second set of samples was also purchased aimed to assess the potential of the ET to discriminate between different herbs. The aim was to ensure that the discrimination of the different samples was not only due to the total phenolic content, for which reason the number of classes was restricted, but the variability within classes increased.

For the quantitative study, 41 samples were considered as detailed in Table 1. Those included common spices and herbs such as: clove (3), coriander (2), cumin (4), laurel (3), oregano (8), rosemary (4), thyme (4), cardamom (1), cinnamon (1), ajwain (1), anise (1) and black pepper (1). Additionally, some herbs related to traditional medicine (obtained directly from local producers) were also considered: chamomile (1), cotton lavender (1), hypericum (1), sesame (1) and olive tree leaves (4). All considered samples were dried ones.

For the qualitative study, a total of 35 samples of the following herbs/spices were considered (7 samples for each): clove, cumin, oregano, rosemary and thyme. All considered samples were dried ones, except rosemary for which 4 fresh samples were also considered.

For its analysis, a simple and non-expensive extraction method was developed. First, 2 g of each sample were weighted in a 10 mL polypropylene (PP) tube, to which 8 mL of ethanol (96% v/v) were introduced. Next, those were sonicated for 15 min and magnetically stirred for 45 min. The extracting solution was then filtered through a glass microfiber filter (GMFA type, Ø 90 mm from Scharlau, Barcelona, Spain) into a 25 mL volumetric flask, and this extraction process was then repeated twice for each sample. Finally, the filter and funnel were rinsed with ethanol and the flask filled up to 25 mL with this solvent. Ethanol extracts of the samples were then measured; alternatively, those could be stored at −18 °C until its analysis.

### 2.3. Electrochemical measurements

Electrochemical measurements were carried out using a multi-channel PGSTAT 30 Autolab potentiostat (EcoChemie, The Netherlands) controlled with GPES 4.7 version software (EcoChemie). The sensor array was formed by 6 graphite-epoxy composite (GEC) working electrodes and a combined redox electrode (Crison 5261, Barcelona, Spain), consisting of a Ag/AgCl reference electrode and a metallic platinum ring which served as reference and auxiliary electrodes. The array of working electrodes was prepared following the conventional methodology in our laboratories (Cetó, Gutiérrez, Moreno-Barón, Alegret, & del Valle, 2011; Olivé-Monllau, Baeza, Bartolí, & Céspedes, 2009), mixing graphite (18% w/w) with epoxy along with different modifiers (2% w/w) (see Supporting Information for a more detailed description). In this manner, a bare GEC electrode was prepared and the other five GECs were modified using Bi<sub>2</sub>O<sub>3</sub>, CuO, SnO and TiO<sub>2</sub> nanoparticles as well as Prussian blue – one modifier per electrode.

Cyclic voltammograms for each of the electrodes were recorded between −1.3V and 1.3V, with a step potential of 10 mV and a scan rate of 80 mV/s, without applying any pre- or post-electrochemical step to

**Table 1**

Information of the samples used in the quantitative analysis of the total phenolic content of different herbs and spices.

Type	# of samples	FC index (GAE mg/L)
Clove	3	3503–4411
Coriander	2	15–149
Cumin	4	155–282
Laurel	3	213–1173
Olive	4	426–668
Oregano	7	489–1354
Rosemary	4	248–1037
Thyme	5	409–1127
Other	9	8–1715

enrich the signal or clean the electrodes. In this direction, to prevent any undesired fouling effect, the electrodes were cleaned after each measurement by dipping them in a 20% v/v ethanol/water solution under magnetic stirring for 30 s.

#### 2.4. Spectrophotometric measurements – Folin-Ciocalteu index

Absorbance measurements were carried out using a Biochrom Novaspec II visible spectrophotometer (Cambridge, UK). The total phenolic content was assessed employing the Folin-Ciocalteu (FC) method following the standard procedure, and expressed as equivalents of gallic acid (GAE) in mg/L (Cetó, Céspedes, & del Valle, 2012; Commission Regulation EEC, 1990). Briefly, 1300  $\mu$ L of deionized water, 400  $\mu$ L of a 20% sodium carbonate solution and 100  $\mu$ L of the FC reagent were mixed with 200  $\mu$ L of the sample (after appropriate dilution in case of the herbs or the different gallic acid stock solutions) in an Eppendorf tube. Then, allowed to react in the darkness at room temperature for 30 min, at which point the absorbance at 760 nm was measured directly. Next, the calibration curve was built from the plot of absorbance vs. concentration of gallic acid. The FC index for the different herb samples was evaluated by interpolation of the absorbance value, multiplying the resulting value by the respective dilution factor.

#### 2.5. Data processing

Multivariate data analysis was done in Matlab R2020b employing its Statistics toolbox by specific routines written by the authors. Correlation between the voltammetric measurements and FC index was evaluated employing partial least squares (PLS) regression. Subsequently, voltammetric responses were also submitted to principal component analysis (PCA) to evaluate samples (dis)similarities and to assess patterns in the data that would allow its discrimination.

Briefly, PCA is an unsupervised pattern recognition method that allows evaluating samples (dis)similarities and assessing initial patterns in the data by projecting the information carried by the original variables onto a smaller number of underlying (“latent”) variables called principal components (PCs) with new coordinates called scores, obtained after data transformation. Then, by plotting the scores on the PCs, one can visualize interrelationships between different variables, and thus detect and interpret sample patterns, groupings, similarities or differences (Cetó, Céspedes, & del Valle, 2013; Jolliffe, 2002). On the other hand, PLS is a supervised regression method, performing a similar linear transformation procedure, decomposing both the response ( $x$ ) and target ( $y$ ) matrices simultaneously to find the latent (or hidden) variables (LVs) in  $x$  that will best predict the latent variables in  $y$  (Cetó et al., 2013; Richards, Bessant, & Saini, 2002). The optimal value of LVs was selected from the plot of the obtained root mean square error against the number of LVs, selecting the value where the curve reached a minimum. Additionally, the residuals and the explained variance plots were also checked to ensure that the proper number of LVs was selected.

### 3. Results and discussion

#### 3.1. Voltammetric responses

The analysis of herbs and spices with the ET required a pre-treatment step so as to be able to carry out the voltammetric measurements (see Fig. S3 for a schematic of the approach). For that purpose, a simple extraction procedure based on sonication and stirring was evaluated, in which ethanol was used as the extracting solvent. Similarly, ethanol was preferred over methanol due to its lower toxicity. The choice of the solvent and the conditions were based on previously reported studies with different herbs (Sarma & del Valle, 2020; Shan et al., 2005), aiming to ensure a high extraction degree. This was important as, on the one side, the concentrations of the different bioactive compounds vary significantly and might be rather low for certain herbs; and on the other

side, samples were to be diluted afterwards to carry out the voltammetric measurements in aqueous buffered media. Thus, upon completion of the extraction, samples were diluted 1:20 with buffer for the voltammetric measurements, and the voltammetric responses for different herbs under this conditions were recorded to evaluate the suitability of the method. That is, to ensure that enough signal was obtained for differently preserved samples, but also that responses for different herbs were also different (Fig. 1).

An extract of the responses obtained for different samples is presented in Fig. 1. As can be seen, a significant difference is obtained in comparison to the blank (Fig. 2A), confirming the extraction of different electroactive compounds from the herbs and spices. Mostly, oxidative signals are obtained, which are related, between others, to the presence of phenolic compounds (responsible of most of the antioxidant properties of the herbs), and that of the volatile essential oils (responsible from their characteristic aroma) (Leja & Czaczkyk, 2016; Shan et al., 2005). Moreover, it should also be noted how the different sensors provide differentiated signals, a situation highly desirable when developing ETs as the aim is to obtain comprehensive information about complex samples by simultaneously detecting a large spectrum of compounds in a short time; information that will later be combined and related to the phenomena under study.

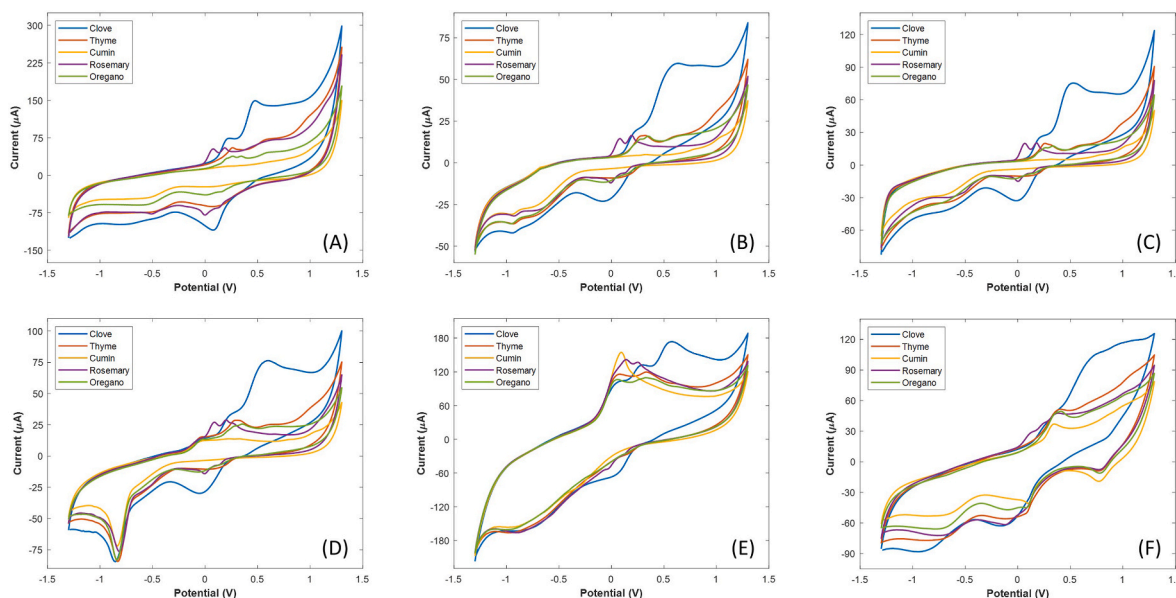
After confirming the suitability of the extraction method and the considered sensor array, the next step was the measurement of a larger set of samples, which were used to build the chemometric models that allow the monitoring of herbs' quality. As already stated, phenolic compounds content varies significantly among the different herbs, but also during the drying process, or with improper storing conditions. Therefore, its monitoring can provide a direct estimation of herbs quality.

#### 3.2. Total phenolic content assessment

The analysis of the total phenolic content, by means of the FC method, confirmed its decay upon samples aging or with improperly storage. More importantly, the voltammetric profiles for the different oregano samples (shown in Fig. 2), were also consistent with this behaviour as evidenced from the smaller peaks obtained for O1 (a very old sample) in comparison to those of O4 (a recently opened sample); opening the room for its usage as an indicator of the herbs and spices freshness and good preservation. However, as it was important to ensure that the performance of the ET was not dependent on the type of herb, a set of samples covering a wider range of typical herbs and spices was selected.

Under the conditions described in Sections 2.3 and 2.4, a total of 41 herbs and spices were measured employing both the ET and the FC method. For the modelling of the data, PLS was chosen as it is a quick, efficient and commonly used multivariate regression method. Moreover, to ensure an unbiased prediction of the model performance, the use of a resampling method was preferred (Molinaro, Simon, & Pfeiffer, 2005). In this manner, the initial set of data was divided between train and test in a fixed ratio (67:33 in this case), and a model was built employing the data of the train subset, which performance was evaluated towards the samples of the test subset. Next, this data division was repeated employing a different arbitrary distribution between both subsets, and a new model was built. This process was repeated a total of 50 times, after which the estimations of the different models for each sample were averaged (separately for the training and testing subsets), what allowed not only to obtain a more unbiased evaluation, but to obtain an estimation of the uncertainties associated with the model.

The performance of the built PLS model, with 4 LVs, is shown in Fig. 3, where the comparison graphs of predicted vs. expected are presented. As can be seen, a good agreement is found between the ET outcome and the reference method. To numerically evaluate this, the regression parameters of the comparison were calculated and the values compared with the theoretical ones (slope and correlation coefficient



**Fig. 1.** Voltammetric fingerprints obtained for different herbs and spices extracts with the considered sensor array: (A) bare graphite, (B)  $\text{TiO}_2$ , (C)  $\text{SnO}$ , (D)  $\text{Bi}_2\text{O}_3$ , (E)  $\text{CuO}$  and (F) Prussian blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

equal to 1, and intercept equal to 0). Furthermore, the confidence intervals for those were also calculated to assess whether or not the theoretical values were contained within those. However, if the tests of significance for the slope and intercept are carried out independently, erroneous conclusions could be derived as the strong correlation between the estimated slope and intercept of a straight line obtained by least squares would be ignored (Mandel & Linning, 1957). Consequently, the joint confidence intervals for the slope and intercept were calculated, which are presented in Fig. 3C. As can be easily noted from the plot, the ideal point (1,0) is within the ellipsoidal confidence intervals for both subsets, allowing to state that there are no significant differences between the values predicted by the model and the FC method. In this sense, it is important to note the enlarged scale of the Y-axis (corresponding to the slope of the comparison), as otherwise it might lead to the impression that the discrepancies are higher. Similarly, the values of the intercept are also rather low if comparing to the range of values observed (in the order of 1000's). Lastly, it is also important to highlight the similar “area” of both ellipses, evidencing the similar performance obtained for the training and testing subsets.

### 3.3. Qualitative analysis of herbs and spices

The next step was to assess the potential of the ET to qualitatively discriminate the different herbs and the potential changes of those during the drying process. That is, to confirm that the ET did not only provide information about the phenolic composition, but a global fingerprint, from which further information could be obtained. Despite it is true that the recorded signals are dependent on the phenolic content (Fig. 2A), it cannot be neglected that the voltammetric profile of those is also characteristic of the considered spice (Figs. 1 and 2B). Therefore, oppositely to the previous scenario, in order to be able to evaluate the clustering of the data, it was important to increase the number of samples of each class.

In this direction, the discrimination of some of the more common herbs was initially considered, which corresponds to the simplest scenario. Namely, clove, cumin, oregano, rosemary and thyme were considered, for which different samples had been obtained from different producers.

Thus, the first step was to submit the obtained responses for those herbs to PCA. The use of an unsupervised pattern recognition method at

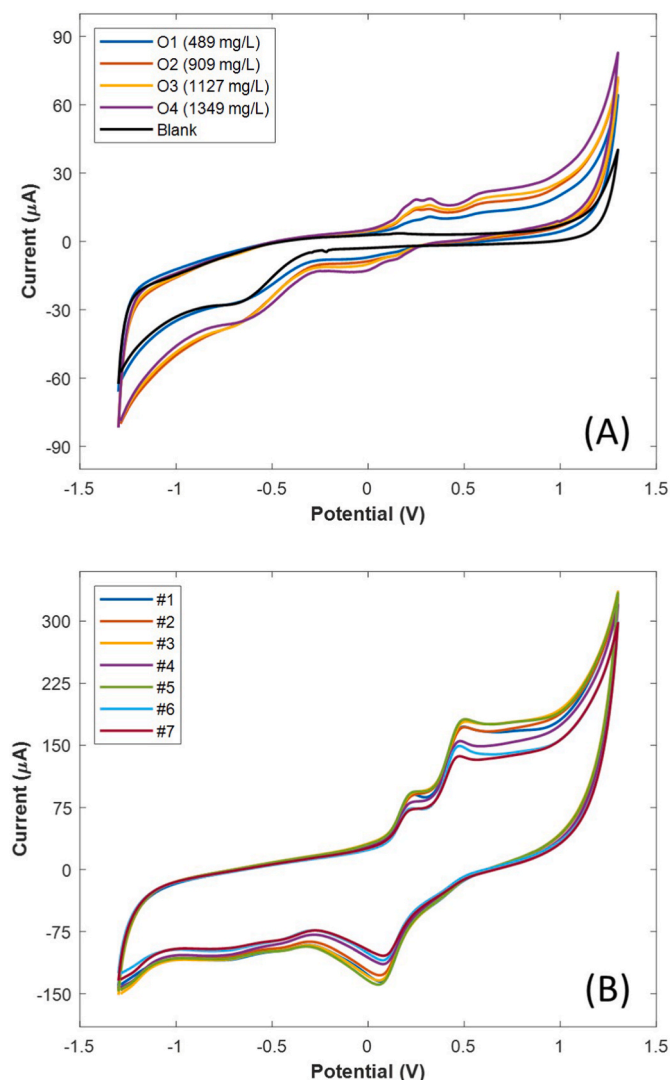
this initial stage, over other supervised methods such as partial least squares discriminant analysis (PLS-DA) or support vector machines (SVMs), was preferred as any clusters that may arise in the scores plot will be based only on its response profile rather than any prior expected similarities (Lavine & Rayens, 2009). This is because PCA provides a better representation of samples (dis)similarities based only on the measurements' variance, but not on any *a priori* class assignment.

The obtained scores plot is shown in Fig. 4. The cumulative variance with only the first two principal components (PCs) was ca. 69.4%, meaning that a large portion of the variance of the initial data is now summarized with only those two variables. Secondly, certain clusters can be clearly seen as a result of the analysis. For the sake of clarity, different symbols have been employed for the different samples, but it has to be remarked that those are only used to facilitate the interpretability of the plot and that do not correspond to any classification. Furthermore, in the same line, the confidence limits for the different clusters were also calculated using the *t* distribution and plotted (Cetó et al., 2016; Wagner & Castner, 2001). Lastly, the Silhouette clustering metric was also calculated (Kaufman & Rousseeuw, 1990), which ranges from  $-1$  to  $+1$ , and provides a measure of how good the clustering is by comparing the intra-cluster distances (cohesion) with respect to the intra-cluster distances (separation). In this case, the calculated value was 0.905, which is indicative of a good clustering.

Despite PCA is not performing any classification itself, it cannot be denied that the ET is capable to discriminate between the different herbs. While PC1 may seem to still be related with the higher phenolic content, it is clear that it is not the only factor contributing to the discrimination, as otherwise there would be an overlapping between different herbs as the FC ranges observed for certain herbs and spices were overlapped (Table 1). Indeed, this discrimination arises from the different voltammetric profiles obtained for each herb/spice (Fig. 1), as the actual phenolic compounds and essential oils present are different for each plant.

Interestingly, another aspect to be highlighted is the higher dispersion for the rosemary cluster, which was attributed to the fact that both fresh and dried (including new and old) samples had been considered, while all the other herbs and spices samples were dried ones. Indeed, a deeper look at the samples from that cluster revealed that the 4 closer central ones corresponded to the freshly collected. Thus evidencing the huge potential of the ET to be used as a tool for the evaluation and





**Fig. 2.** Voltammetric fingerprints obtained for (A) four different oregano samples with increasing phenolic content employing the SnO<sub>2</sub> sensor, and (B) the seven clove samples considered in this study with the bare graphite sensor.

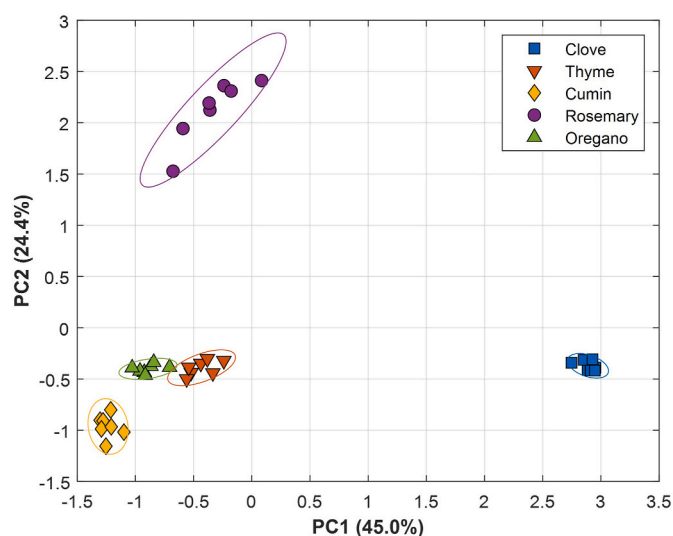
monitoring of the quality and drying process of different herbs and spices.

In this direction, upon establishing the basis of ETs usage on this field, further studies will focus on the actual evaluation of samples submitted to different drying processes under well-controlled conditions, rather than relying on commercial samples.

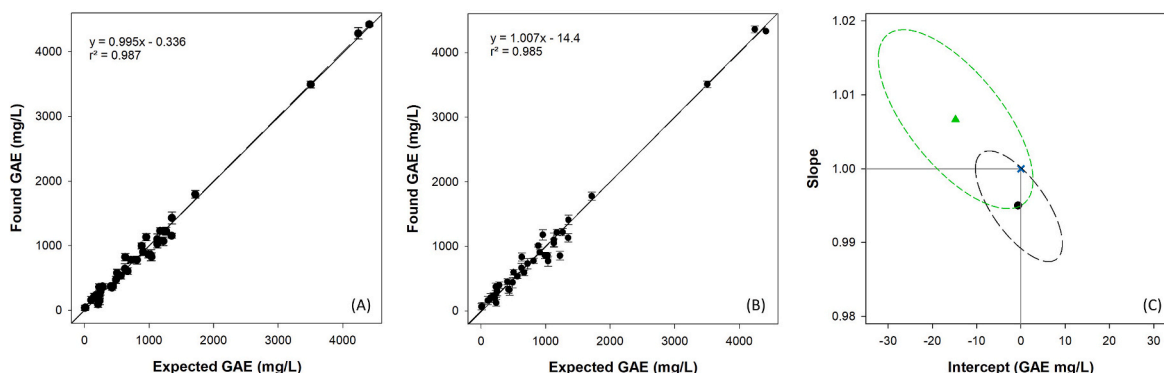
As already introduced, the main difference of the current approach in comparison to traditional ones is not being compound-oriented, what allows extracting further information of the samples by simply changing the set of samples and/or the chemometric tool applied; what provides a significant advantage. This is possible as rather than focusing on the detection of a few specific components which have previously been identified as responsible for a certain characteristic, the aim is to target all detectable components of the sample. That being said, it has to be reckoned that depending on the type of sensors employed, different analytes will be detected and there might be scenarios where voltammetric sensors might not be as suitable.

#### 4. Conclusions

The potential of a voltammetric ET as a suitable tool for the analysis



**Fig. 4.** 2D score plot obtained after PCA of the voltammetric data corresponding to different herbs and spices. Ellipses plotted correspond to 95% confidence limits for each of the groups.



**Fig. 3.** Performance of the optimized PLS model. Comparison graphs of obtained vs. expected GAE for the (A) training and (B) testing subsets. Results provided correspond to the average of the values obtained for each sample after 50 repeated calculations done with random division of samples for train/test subsets each time. Uncertainties calculated at the 95% confidence level. The dashed line corresponds to the theoretical diagonal line ( $y = x$ ). (C) Joint confidence intervals for the (●, black long dashed line) the training and (▲, green short dashed line) testing subsets, calculated at the 95% confidence level. The ideal point (1,0) is also plotted (x). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

of different herbs and spices has been presented herein. The sensor array constituting the ET was based on the use of different electrode modifiers, which allowed to generate different fingerprints for each of the samples. Prior to its analysis, herbs and spices samples were submitted to a simple extraction procedure achieved by sonication followed by magnetic stirring, without requiring any further pre- or post-treatment. A quantitative model for the prediction of total phenolic content in the different samples was built using PLS as the regression model, which can be useful to monitor the change on the phenolic concentration during the drying of the different herbs and spices. Moreover, not only information about the total phenolic content can be obtained, but it has been proven that further qualitative information can be obtained if those same fingerprints are submitted to PCA.

Overall, the main benefits of the herein presented approach include its simplicity, versatility, low-cost and rapid measurement; all of them features necessary to achieve the engagement of the stakeholders. Moreover, the same approach can also be adapted for the discrimination of other solid food samples given the great relevance of phenolic compounds in plant products (e.g., vegetables, fruits or cereals).

### CRedit authorship contribution statement

**Xavier Cetó:** Conceptualization, Methodology, Software, Writing – original draft. **Munmi Sarma:** Investigation, Methodology. **Manel del Valle:** Conceptualization, Funding acquisition, Writing – review & editing.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.lwt.2023.115578>.

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