

Assessment of Table Olives' Organoleptic Defect Intensities Based on the Potentiometric Fingerprint Recorded by an Electronic Tongue

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Abstract Table olives are prone to the appearance of sensory defects that decrease their quality and in some cases result in olives unsuitable for consumption. The evaluation of the type and intensity of the sensory negative attributes of table olives is recommended by the International Olive Council, although not being legally required for commercialization. However, the accomplishment of this task requires the training and implementation of sensory panels according to strict directives, turning out in a time-consuming and expensive procedure that involves a degree of subjectivity. In this work, an electronic tongue is

proposed as a taste sensor device for evaluating the intensity of sensory defects of table olives. The potentiometric signal profiles gathered allowed establishing multiple linear regression models, based on the most informative subsets of signals (from 24 to 29 recorded during the analysis of olive aqueous pastes and brine solutions) selected using a simulated annealing meta-heuristic algorithm. The models enabled the prediction of the median intensities ($R^2 \geq 0.942$ and $RMSE \leq 0.356$, for leave-one-out or repeated K -fold cross-validation procedures) of butyric, musty, putrid, winey-vinegary, and zapateria negative sensations being, in general, the predicted intensities within the range of intensities perceived by the sensory panel. Indeed, based on the predicted mean intensities of the sensory defects, the electrochemical-chemometric approach developed could correctly classify 86.4% of the table olive samples according to their trade category based on a sensory panel evaluation and following the International Olive Council regulations (i.e., extra, 1st choice, 2nd choice, and olives that may not be sold as table olives). So, the satisfactory overall predictions achieved demonstrate that the electronic tongue could be a complementary tool for assessing table olive defects, reducing the effort of trained panelists and minimizing the risk of subjective evaluations.

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Introduction

Table olives are worldwide consumed. During table olive production, the negative attributes related to the technological procedures should be minimized and controlled in order to obtain natural or minimally processed products, retaining their nutritional and healthy properties. These properties are mainly

related to table olives' fatty acid composition (Bianchi 2003) as well as to minor constituents, such as tocopherols and phenolic compounds (Montaño et al. 2005). According to the International Olive Council (IOC 2004), table olives are a complete food from a nutritional point of view, which contain primarily water, fat, carbohydrates, protein, dietary fiber, pectin, biophenols, vitamins, organic acids, and mineral elements. Table olives are usually rich in natural antioxidants (i.e., polyphenols) being their antioxidant capacities and functional effects on human well-being often reported (Boskou et al. 2006; Lanza 2012). Nevertheless, its quality depends on the suitability of the raw material, the processing technology, the nutritional composition, and the presence and intensity of organoleptic negative attributes. Unpleasant sensory sensations are mainly caused by off-odor substances, which appear during table olives' processing steps used in their production, and include defects related to the "abnormal fermentation" (e.g., olfactory negative sensations perceived directly or retronasally, reminiscent of the odor of decomposing organic matter caused by the development of contaminating microorganisms: butyric fermentation, putrid fermentation, and zapateria) or "other defects" (e.g., olfactory sensations: rancid and cooking effect; or olfactory–gustatory sensations: musty, metallic, earthy, soapy, and winey-vinegary) (Lanza 2012; IOC 2011).

Although not legally required, table olives may be classified according to the evaluation of negative sensory attributes, performed by a trained sensory panel (IOC 2011). Taking into account the median intensity of the defect predominantly perceived (DPP) (e.g., perceived with the greatest intensity, in a scale from 1 (not perceived) to 11), the samples shall be classified in four categories (IOC 2011): Extra or Fancy (DPP \leq 3); First, 1st Choice or Select ($3 < \text{DPP} \leq 4.5$); Second, 2nd Choice or Standard ($4.5 < \text{DPP} \leq 7.0$); and olives that may not be sold as table olives (DPP > 7.0). Recently, Lanza and Amoruso (2016) verified that there is a relation between table olives' negative sensations and the gustatory/kinesthetic changes in the entire organoleptic profile of the table olives being samples classified as "Extra or Fancy" with DPP greater than 1.0, quite similar to samples with more intense negative attributes. Based on this finding, Marx et al. (2017a) proposed an additional trade quality category (i.e., extra without defects) that included table olives for which no organoleptic defect could be perceived by a sensory panel (DPP equal to 1, for all negative attributes evaluated). So, either for classification purposes (following the IOC recommendations) or for ensuring the consumers' confidence when purchasing this type of highly appreciated food product, the sensory assessment of the intensity of organoleptic negative attributes is of utmost relevance. However, the scarce number of available trained sensory panels together with the low number of table olive samples that can be evaluated per day, according to the IOC directives, and considering the time–effort and cost involved in the training process of new panelists make the

development of complementary sensory analytical tools a practical need, which success could allow the implementation of such evaluation a legal requirement.

Qualitative and/or quantitative E-tongue-based approaches have been successfully reported for olive oil physico-chemical and positive sensory sensation assessment (Apetrei et al. 2010; Apetrei et al. 2016; Apetrei and Apetrei 2013; Cosio et al. 2007; Dias et al. 2016a; Rodríguez-Méndez et al. 2010; Rodrigues et al. 2016a, b; Santonico et al. 2015; Veloso et al. 2016). Regarding the evaluation of negative sensory attributes, Borràs et al. (2015) proposed partial least squares discriminant classification models, based on mid-infrared spectra, to differentiate extra-virgin olive oils (defect absent) from lower-quality olive oils (defect present). Concerning table olive sensory evaluations, using analytical techniques, few works have been described in the literature. Panagou et al. (2008) successfully applied an electronic nose (E-nose) to differentiate the quality (i.e., acceptable, marginal, and unacceptable) of fermented green table olives based on their volatile fingerprints. Recently, Marx et al. (2017a) applied an E-tongue-chemometric fusion strategy to satisfactorily classify table olives according to the sensory quality level and to differentiate organoleptic negative attributes perceived in table olives. Also, it was found that the potentiometric E-tongue signal profiles varied linearly (sensitivities ranging from -287 mV/decade up to $+197$ mV/decade) with the decimal logarithm of the concentration of aqueous solutions of standard compounds that mimicked butyric, putrid, and zapateria negative sensations (Marx et al. 2017a). However, in this last work, the quantitative potential use of E-tongue for quantifying the defect intensities perceived in table olives was not addressed. Indeed, regardless of the successful qualitative classification of commercial table olives according to their sensory quality levels reported by Marx et al. (2017a), the reported strategy would not allow to infer about the number, type, and median defect intensities that existed in each one of the evaluated samples.

So, in this work and for the first time, the potential application of a potentiometric E-tongue to quantify the median intensity of organoleptic defects perceived by a sensory panel in commercial table olives was evaluated. This electrochemical-based approach was already successfully applied for assessing the median intensities of basic gustatory sensations (i.e., acid, bitter, and salty tastes) perceived in table olives (Marx et al. 2017b). The procedure adopted in the present study also involves the selection of the most informative sub-sets of sensors, achieved using a meta-heuristic simulated annealing (SA) algorithm, and their new application to establish multiple linear regression (MLR) models to determine the intensity of common sensory defects (e.g., butyric, putrid, zapateria, winey-vinegary, and musty), perceived by a trained sensory panel in commercially available Portuguese and Spanish table olives. The performances of the E-tongue-MLR-SA models established were evaluated using the

leave-one-out (LOO) and the repeated *K*-fold cross-validation (CV) procedures, being the last CV variant used to minimize the risk of overoptimistic results attributed to the LOO-CV technique, allowing keeping 25% of data as an internal test dataset. The feasibility of E-tongue-MLR-SA models for quantifying the intensities of organoleptic defects perceived in table olives is evaluated for the first time. Finally, based on the mean intensities of each sensory defect, predicted by the E-tongue-MLR-SA, to each table olive sample a trade category was attributed, following the IOC regulations (IOC 2011), and a comparison was carried out with the trade category attributed based on the sensory panel evaluation (Marx et al. 2017a), allowing to assess the qualitative predictive capability of the proposed electrochemical-chemometric approach.

Materials and Methods

Table Olive Samples

Forty-four commercial table olive samples, from 18 different brands and different batches, were purchased in local supermarkets in Bragança (Portugal) and Zamora (Spain). Olive cultivars and the technological process applied for table olive production have been previously reported (Marx et al. 2017a). Briefly, table olives studied were obtained mostly by natural fermentation (samples 1 to 27, 30, 36 to 40, and 42 to 44 of Table 1), Spanish-style (samples 28, 29, 32, 34, 35, and 41 of Table 1), California-style (sample 33 of Table 1), or a mix of different technological processes (sample 31 of Table 1). Regarding the olive cultivars, the studied samples were mainly from Galega cv (samples 1 to 22, 24, 30, and 40 of Table 1), Cobrançosa cv (samples 41 to 43 of Table 1), Hojiblanca cv (samples 29, 33, and 34 of Table 1), Negrinha de Freixo cv (samples 26 and 44 of Table 1), Manzanilla cv (sample 28 and 36 of Table 1), Gordal (sample 32 of Table 1), Empeltre cv (sample 37 of Table 1), and mixed cultivars (samples 23, 25, 27, 31, 35, 38, and 39 of Table 1).

Table Olive Organoleptic Analysis: Sensory Panel and Sample Preparation

The organoleptic assessment of table olives was performed by trained sensory panel that evaluated and graded each sample (olive and brine solution) according to a pre-defined intensity scale that varied from a minimum level of 1 (defect not perceived) to a maximum level of 11. Among other attributes, the presence and intensity of negative olfactory (e.g., butyric, putrid, zapateria, and rancid sensations) and/or olfactory-gustatory attributes (e.g., musty and winey-vinegary sensations), usually used for commercially classifying table olives according to sensory quality trade levels, were assessed by the sensory panel of the School of Agriculture of the Polytechnic Institute of Bragança

(Portugal), composed by eight skilled tasters, which were trained following the IOC directives (IOC 2011) and as described in a previous work (Marx et al. 2017a). All panelists have voluntarily accepted to be a part of the sensory panel and participate periodically in research sensorial quality studies. The panel evaluation performance was also assessed according to the IOC regulation (IOC 2011). During the training sessions and table olive analysis, the panel leader collected the profile sheets completed by each of the panelists and reviewed the intensities recorded for each of the descriptors (i.e., butyric, musty, putrid, winey-vinegary, and zapateria standard solutions and table olive samples). When an anomaly record was detected, the identified panelists revised the profile sheets and, if necessary, repeated the test. The median intensity of each perceived defect and the respective confidence intervals were calculated according to the method described in the IOC regulations, using the official computer program (IOC 2011), and only considered the negative attributes for which the coefficients of variation of the intensities perceived by the panelists were equal or lower than 20%. The table olive samples were evaluated following the IOC regulations (IOC 2011). Briefly, the number of olives varied according to their size, ensuring always that the bottom of the glass used for the sensory analysis was full when placing the olives side by side in a single layer, being covered with the brine solution, which is poured over the olives until they are completely covered or at least up to three quarters of the height of the olives. For olives with a size-grade greater than 91/100 size-grade, the volume of sample contained in the glass was always less than half the height of the glass (i.e., ~30 mm). For table olives with a size-grade below 91/100, a minimum of three olives were analyzed.

E-Tongue Device

The E-tongue multi-sensor device was previously described (Marx et al. 2017a). Briefly, it included two print-screen potentiometric arrays (9.5 cm of width and 2.5 cm of height) containing each one 20 sensors (3.6 mm of diameter and 0.3 mm of thickness). The composition of the E-tongue sensors included lipid additives (~3%), plasticizer compounds (~65%), and high molecular weight polyvinyl chloride (PVC, ~32%), chosen due to their signal stability over time and repeatability towards the basic standard taste compounds (sweet, acid, bitter, salty, and umami) (Dias et al. 2009). Each sensor was coded with a letter S (for sensor) followed by a code for the sensor array (1: or 2:) and the number of the membrane (1 to 20), corresponding to different combinations of plasticizer and additive compounds.

Table Olive Commercial Samples and Brine Solutions

The 44 commercial table olive samples were electrochemically analyzed (both olives and respective brine solutions) using the E-tongue device. For that, brines were assayed directly

Table 1 Median values of the intensities of each defect perceived by the sensory panel (8 panelists belonging to a trained sensory panel) for the evaluation of the table olive samples and respective brine solutions according to the regulations of the IOC (2011)

Sample	Organoleptic defects and respective intensity levels				
	Abnormal fermentation ^a			Others defects ^b	
	<i>Butyric</i>	<i>Putrid</i>	<i>Zapateria</i>	<i>Musty</i>	<i>Winey-vinegary</i>
1	NP	3 (2.5–3.5)	NP	NP	2.5 (1.5–4.5)
2	NP	NP	NP	NP	4 (2.5–4.5)
3	NP	NP	3 (1.5–3.5)	NP	NP
4	NP	NP	NP	NP	2.5 (1.5–3)
5	NP	7.5 (6–8)	NP	NP	NP
6	NP	3.5 (3–5.5)	NP	NP	4.5 (3–7.5)
7	NP	6.5 (4–8)	NP	NP	4 (2.5–4.5)
8	NP	6 (5.5–7)	NP	NP	3 (1.5–4)
9	NP	6.5 (5–7.5)	NP	NP	3.5 (3–4)
10	NP	4 (3.5–5)	NP	NP	3.5 (1.5–4.5)
11	NP	NP	2.5 (1–3.5)	3 (1.5–4.5)	NP
12	NP	NP	3.5 (3–4)	NP	4.5 (3–5)
13	NP	NP	2.5 (1.5–3)	NP	3 (3–4)
14	NP	NP	NP	NP	4 (2.5–4.5)
15	5.5 (4.5–7)	7.5 (6.5–8)	NP	NP	2.5 (1.5–3)
16	6 (4.5–7)	NP	NP	4.5 (3.5–5.5)	1 (1–3)
17	4.5 (3–5.5)	NP	NP	NP	3 (2.5–4)
18	NP	NP	4.5 (2.5–6)	NP	3 (2.5–3.5)
19	NP	4 (3–5)	NP	NP	3.5 (3–4.5)
20	NP	4 (3–4)	NP	NP	3.5 (2.5–4.5)
21	NP	3.5 (3–4)	NP	5 (4.5–6)	NP
22	NP	NP	NP	NP	4.5 (3–5)
23	NP	NP	3 (2–5)	NP	NP
24	NP	NP	NP	3 (2.5–5)	2.5 (1.5–3.5)
25	NP	NP	3 (1.5–4)	NP	NP
26	NP	NP	NP	NP	2.5 (1.5–3)
27	NP	NP	NP	NP	NP
28	NP	NP	NP	NP	NP
29	NP	NP	NP	NP	NP
30	NP	4 (3.5–4.5)	NP	NP	6 (5–6.5)
31	NP	NP	NP	NP	NP
32	NP	NP	NP	NP	NP
33	NP	NP	NP	NP	NP
34	NP	NP	NP	NP	NP
35	NP	2.5 (1.5–3.5)	NP	NP	4.5 (3–5)
36	NP	NP	2.5 (1.5–3.5)	2.5 (1.5–3.5)	4.5 (3.5–6)
37	NP	NP	NP	4 (3–4)	NP
38	NP	6 (3.5–7)	NP	NP	2.5 (1.5–4.5)
39	NP	NP	NP	NP	2.5 (1.5–6)
40	NP	NP	NP	NP	3 (1.5–3.5)
41	7 (6–8.5)	NP	NP	5 (4–5.5)	NP
42	8 (7–8.5)	NP	NP	5 (4–7)	NP
43	NP	NP	NP	10 (8–10.5)	NP
44	8 (7–8.5)	NP	NP	7 (6.5–8)	NP

The range of intensities perceived by the panel (minimum and maximum intensity levels) is given in brackets *NP* defect not perceived by any member of the trained sensory panel (i.e., negative attribute intensity equal to 1, in a scale-range from 1 to 11 (IOC 2011))

^a Sensory defects included in the abnormal fermentation category (butyric, putrid, and zapateria) (IOC 2011)

^b Sensory defects included with the other defects category (winey-vinegary and musty) (IOC 2011)

without any pre-treatment (using 20 mL of undiluted solution) and olives suffered a pre-treatment in order to transform them into an aqueous paste (using 9 g of olives without stone plus 6 mL of water, which was transformed into an aqueous paste) using the same commercial mineral water usually used during the training sessions of the sensory panel (Marx et al. 2017a).

The 9 g of olives (without stone) used for the electrochemical analysis corresponded to the number of olives (which depends on the olive size-grade; e.g., Gordal cv: one table olive covered the glass bottom and had an average weight of 9.4 g without stone; Galega cv, six table olives covered the glass bottom and had an average weight of 9.6 g without stone;

Hojiblanca cv, three olives covered the glass bottom and had an average weight of 9.6 g without stone; Cobrançosa cv: three olives covered the glass bottom and had an average weight of 8.6 g without stone) used during the sensory panel evaluations and that ensured the fulfillment of the IOC requirement (i.e., the bottom of the glass should be full when placing the olives side by side in a single layer).

E-Tongue Assays

All samples were analyzed using the E-tongue at controlled temperature (~ 20 °C) under agitation using a magnetic stirrer (VELP Scientifica). The E-tongue was immersed directly in each sample, and after a 5-min stabilization period, the potentiometric signal profiles of the 40 lipid membranes of the multi-sensor system arrays were acquired. During the signal stabilization period (5 min), the 40 sensor signals were monitored (each 10 s) and so enabled the visualization of the potentiometric signal changes along the time (Marx et al. 2017a). Electrochemical assays were carried out in duplicate for each sample unless the coefficients of variation of the potentiometric signals gathered with each E-tongue sensor were greater than 20% (value set according to the IOC regulations for sensory analysis (IOC 2011)), in which cases a third assay was performed. As proposed by Rodrigues et al. (2016a), to minimize the risk of overoptimistic performance of the multivariate models, for data split (establishment of training and internal validation sets) and modeling purposes, only one electrochemical “average” signal profile per sample (assumed as the aqueous paste or brine solution fingerprints) was used, avoiding that results from duplicate assays of the same table olive sample could be included into both training and validation sets.

Statistical Analysis

Multiple linear regression (MLR) models allow estimating and/or predicting the behavior of a dependent variable (i.e., the intensities of the organoleptic negative attributes perceived by the sensory panel during the evaluation of the table olives) through linear combinations of several independent variables (i.e., potentiometric signal profiles of sub-sets of the 40 E-tongue sensors). The use of this statistical technique requires the fulfillment of some assumptions (e.g., linearity of the experimental data, error independence and their normal distribution, homogeneity of variances, and the absence of multi-collinearity between the independent variables). Nevertheless, MLR models may be used even if some requirements are not fulfilled, which has been shown by their satisfactorily predictive performance when applied for estimating several properties of different food matrices (Dias et al. 2014; Dias et al. 2015; Dias et al. 2016b; Rodrigues et al.

2016a). Moreover, previously it was reported that the data generated from a potentiometric E-tongue was better modeled by MLR models compared to other statistical regression strategies such as partial least squares (PLS) and principal component regression (PCR) (Rodrigues et al. 2016a).

Since the signals of potentiometric sensors with cross-selectivity show high collinearity, the meta-heuristic simulated annealing (SA) variable selection algorithm was used to select subsets of different numbers of sensors to establish and select the best E-tongue-MLR model, using the adjusted coefficient of determination (R^2) as the quality criterion (Cadima et al. 2004; Cadima et al. 2012). The SA is an algorithm for feature selection providing several optimal solutions during the optimization search, but not necessarily the best solution. This algorithm mimics an optimization physical process of heating a material and then slowly decreasing the temperature in each iteration in order to obtain a more stable system, i.e., a structure without defects. During the model optimization process, the SA algorithm randomly selects a new point (group of variables) at each iteration, based on a probability distribution with a scale proportional to the temperature, being the magnitude of the new models' quality criterion (better function value) evaluated against the best current value. The algorithm accepts new points that lower the quality criteria, but also with a certain probability, which is dependent on the temperature, points that raise the quality criterion, allowing the algorithm to avoid being trapped in local minima (Cortez 2014). In this work, the E-tongue-MLR-SA models established were based on subsets of 2 to 42 signals, from a total set of 80 signal profiles recorded for each table olive sample by the E-tongue (40 + 40 signals concerning the simultaneous evaluation of olive paste and brine solution, respectively). The best model was selected by its quantitative prediction performance, being set a minimum adjusted R^2 value of 0.99 for the leave-one-out (LOO) cross-validation (CV) procedure and with the lowest number of sensors but that still allowed obtaining a low root-mean-square error (RMSE). Furthermore, a repeated K -fold-CV procedure was also applied for comparison purposes with the LOO-CV technique results, to ensure that there were no overfitting issues, since satisfactory results were obtained in both cross-validation techniques. It should be remarked that although the number of different independent samples studied is similar to those usually used in the literature regarding table olive analysis (Casale et al. 2010; Cano-Lamadrid et al. 2017; Cortés-Delgado et al. 2016; Galán-Soldevilla et al. 2013; Lanza and Amoroso 2016; Marx et al. 2017a, b; Panagou et al. 2008; Ramírez et al. 2016; Sousa et al. 2011), it does not allow the establishment of a representative external dataset, which justifies use of this cross-validation variant.

So, in this study, 4 folds with 10 repeats were chosen ($K = 4$; repeats = 10), resulting into 40 models evaluated

Table 2 Predictive capability of the E-tongue-MLR-SA models established to quantify the median of 5 negative attributes intensities perceived by a trained sensory panel during the evaluation of 44 commercial table olive samples

Category of organoleptic defect (IOC 2011)	Negative attribute	E-tongue-MLR-SA models ^a				
		No. of signals ^b	Determination coefficient (R^2)		Root-mean-square errors ($RMSE$)	
			LOO-CV	Repeated K -fold-CV ^c	LOO-CV	Repeated K -fold-CV ^c
Abnormal fermentation	Butyric	24 ^d	0998	0975 ± 0019	0.163	0.352 ± 0.098
	Putrid	29 ^e	0995	0972 ± 0018	0.214	0.356 ± 0.127
	Zapateria	24 ^f	0985	0942 ± 0040	0.093	0.218 ± 0.067
Others defects	Musty	28 ^g	0998	0980 ± 0022	0.119	0.218 ± 0.107
	Winey-vinegary	28 ^h	0998	0975 ± 0018	0.150	0.229 ± 0081

LOO-CV leave-one-out cross-validation procedure

^a Multivariate linear regression (MLR) model based on the sub-sets of potentiometric signals, established using the simulated annealing (SA) algorithm, selected among the 80 possible signal profiles obtained with the electronic tongue (E-tongue) during the analysis of olive pastes (40 sensor signals) and brines (40 sensor signals)

^b Number of signals included in the E-tongue-MLR-SA model, selected from the 80 electrochemical signals recorded by E-tongue during analysis of each olive paste (40 signals referring to 40 sensor LE) and respective brine (40 signals related to 40 sensors of the E-tongue)

^c Repeated K -fold-CV: cross-validation procedure with 4 folds, ensuring that at least 25% of the original data are used for internal validation and 10 repetitions

^d E-tongue signals included in the E-tongue-MLR-SA model: 11 recorded signals during the analysis of the brines (S1:8, S1:12, S1:16, S1:20, S2:1, S2:3, S2:4, S2:10, S2:12, S2:13, and S2:15) and the other relating to the response of 13 sensors during the analysis of olive paste (S1:6, S1:9, S1:10, S1:11, S1:13, S1:15, S1:16, S2:2, S2:4, S2:5, S2:8, S2:13, and S2:16)

^e E-tongue signals included in the E-tongue-MLR-SA model: 14 recorded signals during the analysis of the brines (S1:2, S1:6, S1:12, S1:15, S1:16, S1:18, S1:19, S2:1, S2:3, S2:9, S2:12, S2:14, S2:16, and S2:17) and the other relating to the response of 15 sensors during the analysis of olive paste (S1:5, S1:6, S1:7, S1:9, S1:13, S1:15, S1:17, S1:19, S2:1, S2:3, S2:7, S2:13, S2:16, S2:17, and S2:19)

^f E-tongue signals included in the E-tongue-MLR-SA model: 13 recorded signals during the analysis of the brines (S1:1, S1:3, S1:4, S1:5, S1:8, S1:18, S1:19, S2:1, S2:6, S2:8, S2:10, S2:12, and S2:15) the other relating to the response of 11 sensors during the analysis of olive paste (S1:4, S1:6, S1:7, S1:9, S2:2, S2:4, S2:9, S2:10, S2:14, S2:16, and S2:17)

^g E-tongue signals included in the E-tongue-MLR-SA model: 16 recorded signals during the analysis of the brines (S1:2, S1:3, S1:4, S1:9, S1:10, S1:12, S1:13, S1:20, S2:3, S2:4, S2:9, S2:12, S2:14, S2:15, S2:18, and S2:40), and the other 12 for the electrochemical analysis of olive pastes (S1:1, S1:3, S1:9, S1:12, S1:14, S2:1, S2:8, S2:11, S2:12, S2:13, S2:17, and S2:18)

^h E-tongue signals included in the E-tongue-MLR-SA model: 17 recorded signals during the analysis of the brines (S1:2, S1:3, S1:4, S1:8, S1:9, S1:11, S1:15, S1:17, S1:18, S1:19, S1:20, S2:4, S2:9, S2:11, S2:12, S2:13, and S2:20) and the other 11 for the analysis of olive paste (S1:3, S1:13, S1:17, S1:18, S2:3, S2:4, S2:9, S2:11, S2:12, S2:13, and S2:15)

for each sub-set of sensors previously established by the SA algorithm. In a run, the data is randomly divided into K folds (set equal to 4 in this study), being $K - 1$ folds used for training purposes (to establish the best E-tongue-MLR-SA model) and the data of the remaining fold used for test purposes (internal validation). This process is repeated until all folds were used as test internal validation. In other runs, the procedure was repeated (10 times) but with the formation of other folds that included different samples selected randomly.

The possibility of using the selected E-tongue-MLR-SA models (for both LOO-CV and repeated K -fold-CV procedures) as complementary tools for the quantification of the sensory defects present in the table olive

samples was further checked, as suggested by Roig and Thomas (2003a, b). The checking technique involved the establishment of the 95% intervals of confidence (IC) for the slope and intercept values of the single linear regression (LR) obtained by plotting the defect intensities predicted by the E-tongue-MLR-SA models versus the respective intensities of the DPP by the trained sensory panel. The proposed E-tongue-based approach could be foreseen as a satisfactory tool if the 95% IC contained the theoretic values of “zero” and “one” for the intercept and slope values, respectively (Roig and Thomas 2003a, b).

All statistical analyses were performed using the Subselect (Cadima et al. 2004; Cadima et al. 2012) and MASS

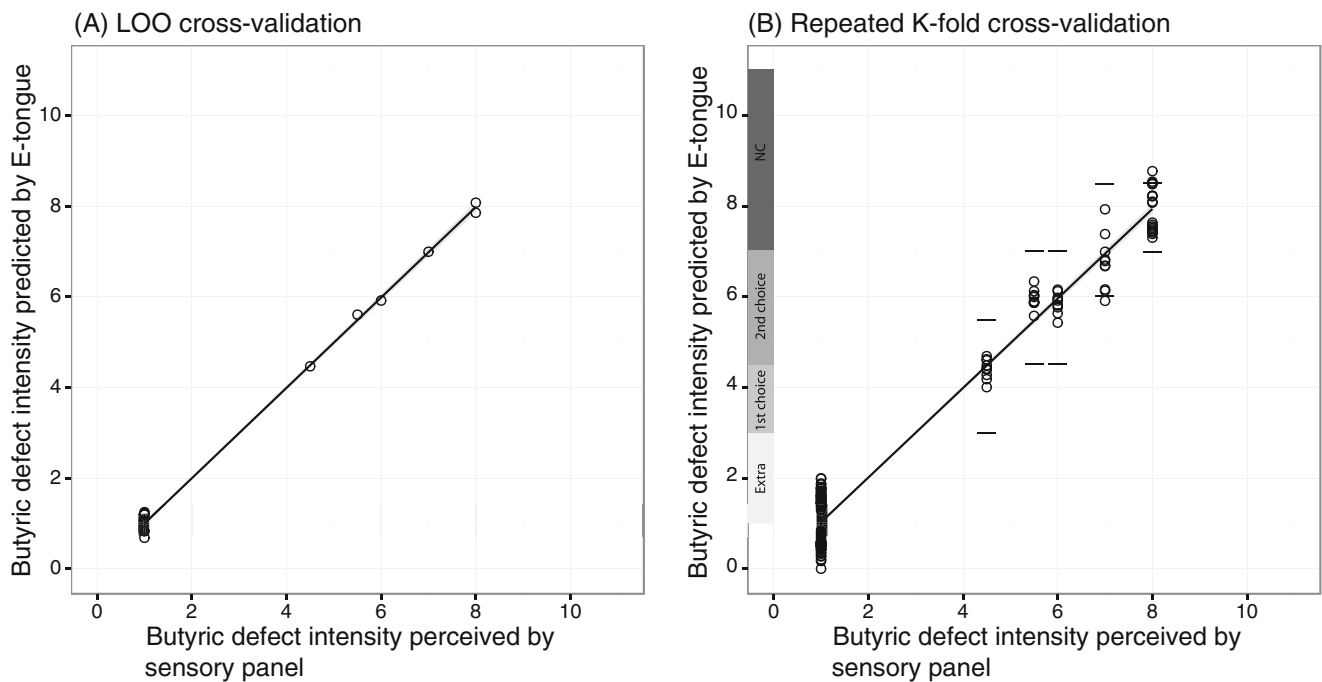


Fig. 1 Quantification of median butyric defect intensities (associated with abnormal fermentation) detected in 44 samples of table olives (olives and respective brine solution) using an E-tongue-MLR-SA

model based on 24 profiles of potentiometric signals. **a** LOO-CV ($R^2 = 0.998$). **b** Repeated K -fold-CV (4 folds \times 10 repeats, $R^2 = 0.975 \pm 0.019$)

(Venables and Ripley 2002) packages of the open-source statistical program R (version 2.15.1), at a 5% significance level.

Results and Discussion

Sensory Defects and Respective Intensities of Table Olives According to the Perception of a Trained Sensory Panel

Each table olive sample and the respective brine solution were evaluated by the sensory panel, trained according to the directives established by the IOC (2011). The median intensities of the negative attributes perceived in each of the 44 independent table olive samples (olive and the respective brine solution) by each of the panelists, regarding olfactory (butyric, putrid, and zapateria) and/or olfactory-gustatory (musty and winey-vinegary) sensations, are shown in Table 1, as well as the interval of the intensities perceived by the sensory panel (lowest and greatest assigned levels).

According to the sensory analysis performed by the trained panelists, for 7 table olive samples no negative attribute was perceived (i.e., intensity equal to 1) and for the other 37 samples 1, 2, or 3 simultaneous organoleptic defects could be perceived during the sensory evaluation of the olives and brine solutions. For these latter samples, in total 5 different negative attributes could be perceived, namely, butyric (6 samples), putrid (14 samples), musty 8 (10 samples), winey-vinegary (26

samples), and zapateria (8 samples). Depending on the intensity of the DPP, the table olives studied could be classified as Extra without defect, Extra, 1st choice, 2nd choice, and olives that cannot be sold as table olives (Marx et al. 2017a).

E-Tongue Potential for Quantifying the Intensity of Organoleptic Negative Attributes Perceived in Commercial Table Olives

The predictive capability of the established E-tongue-MLR-SA models (containing between 24 to 29 sensor signals from the 80 recorded during the simultaneous analysis of the aqueous pastes and brine solutions) for each of the five defects perceived by the sensory panel (i.e., butyric, musty, putrid, winey-vinegary, and zapateria negative sensations) in the 44 olive samples was evaluated using LOO-CV and repeated K -fold-CV procedures. The latter CV variant has been implemented with 4 folds and for 10 repetitions, allowing minimizing the risk of overfitting, which could result in overoptimistic results due to the use of unrealistic prediction models. Indeed, in the last procedure, the initial dataset is randomly split into 4 data subsets (4 folds containing each 25% of the data) being each one used at a time for internal validation purposes of the model established with the other 75% of the data. Moreover, the process is repeated 10 times (value set in this work), resulting in the establishment of 40 multivariate linear models that are internally validated 40 times (i.e., 4 folds \times 10 repeats). The predictive performance achieved by the

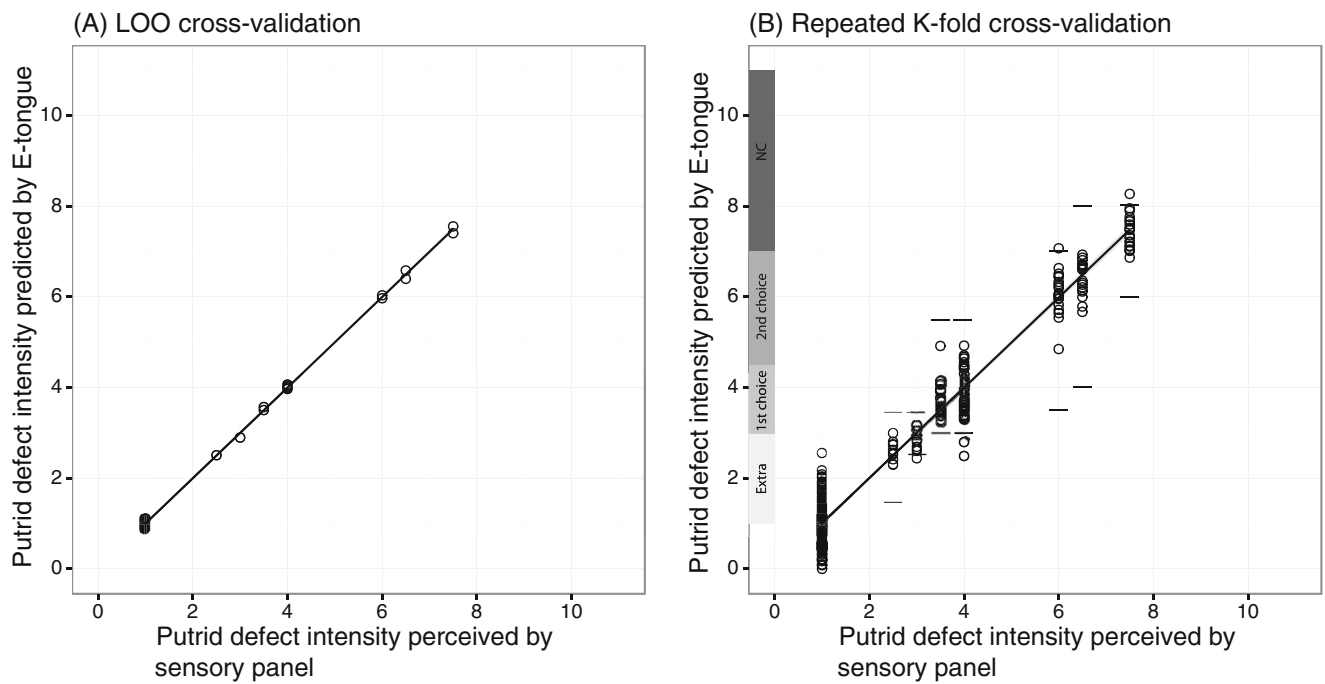


Fig. 2 Quantification of median putrid defect intensities (associated with abnormal fermentation) detected in 44 samples of table olives (olives and respective brine solution) using an E-tongue-MLR-SA model based on 29

profiles of potentiometric signals. **a** LOO-CV ($R^2 = 0.995$). **b** Repeated K-fold-CV (4 folds \times 10 repeats, $R^2 = 0.972 \pm 0.018$)

E-tongue-MLR-SA models established for the quantification of the median intensities of the perceived sensory defects is shown in Table 2. Detailed information (number and type of sensors included in the MLR models) concerning the predictors used for assessing each negative sensory sensation is also given in

Table 2. The overall R^2 and $RMSE$ values obtained for LOO-CV ($0.985 \leq R^2 \leq 0.990$ and $0.093 \leq RMSE \leq 0.214$) and repeated K-fold-CV ($0.942 \pm 0.040 \leq R^2 \leq 0.980 \pm 0.022$ and $0.218 \pm 0.067 \leq RMSE \leq 0.356 \pm 0.127$) procedures pointed out the satisfactory capability of the proposed electrochemical

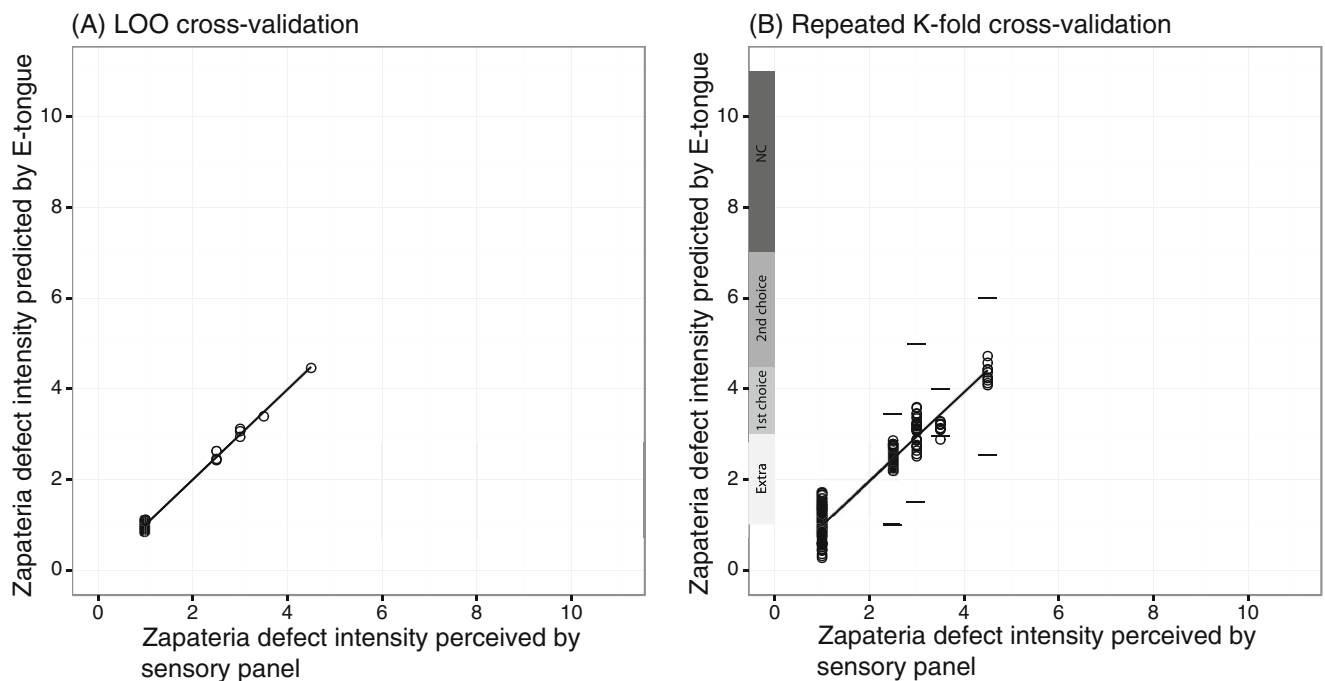


Fig. 3 Quantification of median zapateria defect intensities (associated with abnormal fermentation) detected in 44 samples of table olives (olives and respective brine solution) using an E-tongue-MLR-SA model based

on 24 profiles of potentiometric signals. **a** LOO-CV ($R^2 = 0.985$). **b** Repeated K-fold-CV (4 folds \times 10 repeats, $R^2 = 0.942 \pm 0.040$)

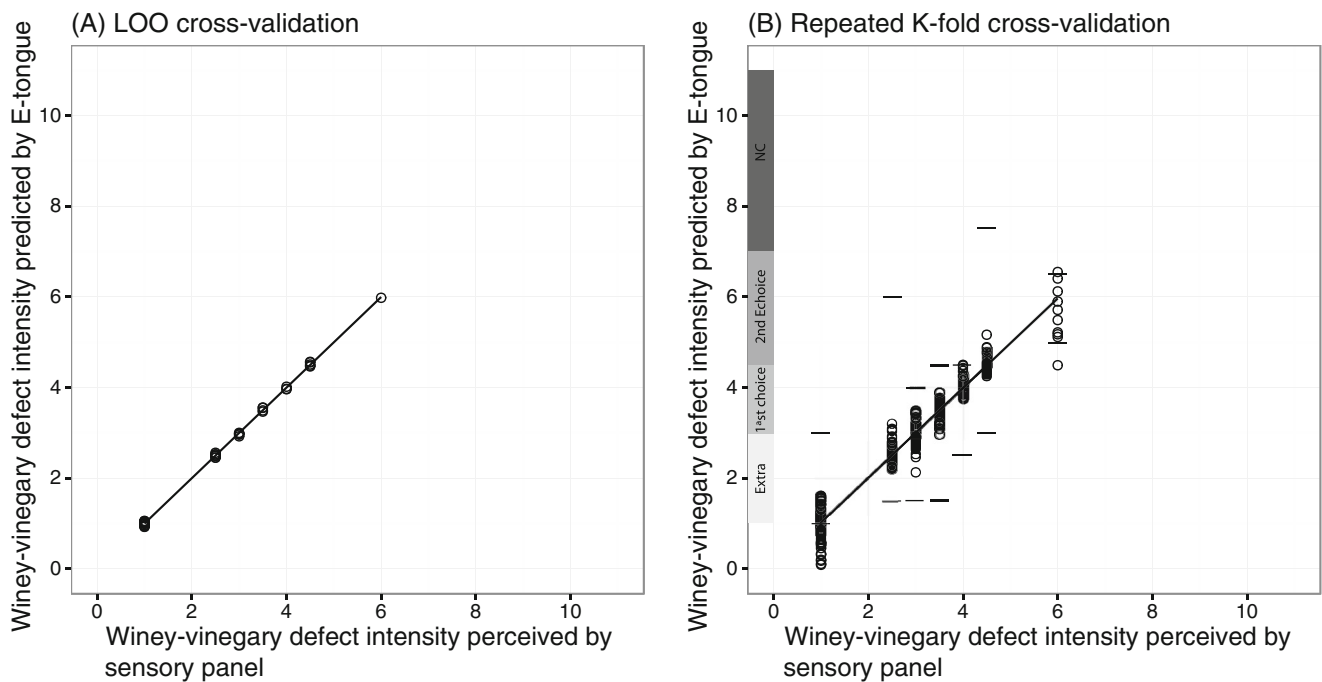


Fig. 4 Quantification of median winery-vinegary defect intensities (associated with other defect category) detected in 44 samples of table olives (olives and respective brine solution) using an E-tongue-MLR-SA

model based on 28 profiles of potentiometric signals. **a** LOO-CV ($R^2 = 0.998$). **b** Repeated K -fold-CV (4 folds \times 10 repeats, $R^2 = 0.975 \pm 0018$)

based approach for quantifying the median intensities of the organoleptic defects perceived in table olives, based on the simultaneous analysis of olive aqueous pastes and brines. The quality of the regression results for both CV variants used in this study (LOO-CV and repeated K -fold-CV) can also be verified from the

visualization of Figs. 1, 2, 3, 4, and 5, for butyric, putrid, zapateria, winery-vinegary, and musty defects, respectively.

As expected, in the case of repeated K -fold-CV (4 folds \times 10 repeats), the predicted median intensities of the 5 perceived sensory defects, obtained with the E-tongue-MLR-SA

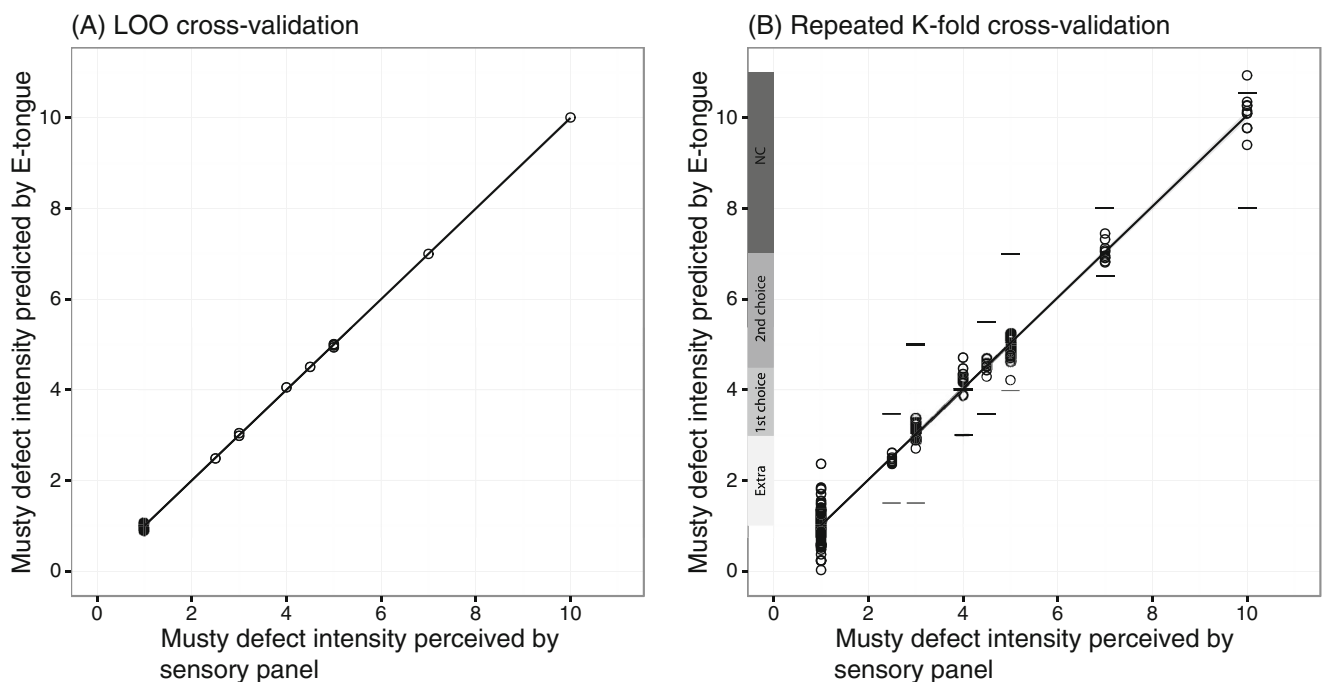


Fig. 5 Quantification of median musty defect intensities (associated with other defect category) detected in 44 samples of table olives (olives and respective brine solution) using an E-tongue-MLR-SA model based on 28

profiles of potentiometric signals. **a** LOO-CV ($R^2 = 0.998$). **b** Repeated K -fold-CV (4 folds \times 10 repeats, $R^2 = 0.980 \pm 0022$)

Table 3 Table olive trade category according to the IOC regulations (IOC 2011): predicted classifications according to the E-tongue-MLR-SA models and classifications based on the sensory panel evaluation (Marx et al. 2017a)

Sample number	E-tongue-MLR-SA model performance (repeated K-fold-CV prediction results)					Model predicted trade category ^b	Table olive trade category according to sensory panel analysis ^c
	Estimated sensory defect mean intensities (minimum and maximum values in brackets) ^a						
	<i>Butyric</i> (24 signals E-tongue)	<i>Putrid</i> (29 signals E-tongue)	<i>Zapateria</i> (24 signals E-tongue)	<i>Musty</i> (28 signals E-tongue)	<i>Winey-vinegary</i> (28 signals E-tongue)		
1	1.17 (0.86–1.45)	2.80 (2.44–3.17)	1.15 (0.97–1.26)	0.95 (0.52–1.25)	2.28 (1.88–2.55)	Extra	Extra
2	0.73 (0.56–0.84)	1.14 (0.67–1.74)	0.98 (0.83–1.22)	1.07 (0.90–1.30)	4.07 (3.82–4.32)	1st choice	1st choice
3	1.19 (0.90–1.44)	0.83 (0.48–1.19)	3.10 (2.74–3.23)	0.92 (0.84–1.14)	0.65 (0.52–0.74)	1st choice	Extra
4	1.56 (1.23–1.88)	1.04 (0.72–1.53)	0.83 (0.57–1.34)	0.92 (0.58–1.84)	2.62 (2.22–2.93)	Extra	Extra
5	0.85 (0.71–1.07)	7.36 (7.02–8.27)	0.88 (0.76–1.08)	1.12 (1.03–1.29)	0.92 (0.78–1.13)	NC	NC
6	1.10 (0.74–1.60)	3.53 (3.23–3.96)	1.02 (0.71–1.66)	0.83 (0.68–0.99)	4.61 (4.51–4.78)	1st choice	1st choice
7	0.85 (0.58–1.22)	6.71 (6.60–6.93)	0.93 (0.69–1.07)	1.00 (0.75–1.15)	3.89 (3.80–3.99)	2nd choice	2nd choice
8	1.62 (1.43–1.81)	6.24 (4.85–7.07)	1.23 (1.05–1.35)	1.21 (1.01–1.38)	3.14 (2.98–3.60)	2nd choice	2nd choice
9	0.92 (0.61–1.18)	6.17 (5.67–6.86)	1.18 (1.02–1.42)	1.26 (0.82–2.37)	3.48 (3.29–3.71)	2nd choice	2nd choice
10	0.87 (0.00–1.44)	3.70 (3.30–4.42)	0.98 (0.70–1.20)	0.97 (0.54–1.70)	3.95 (3.44–4.44)	1st choice	1st choice
11	1.08 (0.75–1.32)	1.25 (0.98–1.90)	2.33 (2.20–2.48)	2.96 (2.70–3.10)	1.14 (0.77–1.46)	Extra	Extra
12	0.95 (0.56–1.60)	0.64 (0.08–1.20)	3.16 (2.88–3.28)	0.80 (0.02–1.40)	4.42 (3.80–5.02)	1st choice	1st choice
13	1.33 (0.92–1.77)	1.02 (0.43–2.01)	2.40 (2.28–2.49)	0.79 (0.22–1.07)	2.85 (2.74–2.98)	Extra	Extra
14	0.85 (0.43–1.41)	0.96 (0.45–1.54)	0.95 (0.55–1.09)	1.18 (0.72–1.35)	4.38 (3.92–4.77)	1st choice	1st choice
15	5.96 (5.58–6.34)	7.51 (6.87–7.95)	1.00 (0.91–1.11)	1.00 (0.47–1.38)	2.50 (2.24–2.63)	NC	NC
16	5.86 (5.43–6.16)	1.05 (0.09–1.58)	0.88 (0.78–1.00)	4.52 (4.29–4.70)	0.96 (0.29–1.19)	2nd choice	2nd choice
17	4.41 (4.01–4.69)	0.93 (0.56–1.35)	1.07 (0.77–1.37)	1.02 (0.88–1.25)	2.88 (2.73–2.94)	1st choice	1st choice
18	1.08 (0.50–1.47)	0.87 (0.42–1.19)	4.34 (4.08–4.72)	0.85 (0.74–1.04)	2.98 (2.53–3.12)	1st choice	1st choice
19	1.60 (0.94–2.00)	3.87 (3.55–4.31)	0.74 (0.44–1.01)	1.10 (0.58–1.40)	3.00 (2.38–3.39)	1st choice	1st choice
20	1.01 (0.65–1.32)	4.35 (3.56–4.71)	1.06 (0.84–1.34)	1.09 (0.90–1.35)	3.84 (3.42–4.49)	1st choice	1st choice
21	1.14 (0.96–1.39)	3.96 (3.52–4.92)	1.16 (0.80–1.46)	4.80 (4.22–4.99)	0.88 (0.71–1.06)	2nd choice	2nd choice
22	0.87 (0.51–1.12)	0.94 (0.50–1.78)	0.94 (0.81–1.07)	1.12 (1.00–1.29)	4.33 (3.90–4.77)	1st choice	1st choice
23	1.23 (0.65–1.45)	0.97 (0.56–1.46)	2.76 (2.51–3.10)	1.24 (1.04–1.38)	0.95 (0.68–1.17)	Extra	Extra
24	0.69 (0.21–1.10)	1.21 (0.86–1.55)	1.04 (0.78–1.27)	3.18 (3.00–3.37)	2.29 (1.72–1.12)	Extra	Extra
25	1.17 (0.91–1.44)	1.30 (0.74–1.66)	3.36 (3.17–3.60)	0.94 (0.72–1.03)	0.92 (0.72–1.12)	1st choice	Extra
26	0.63 (0.27–0.89)	0.75 (0.21–1.11)	0.94 (0.46–1.70)	0.94 (0.83–1.12)	2.46 (2.22–2.59)	1st choice	Extra
27	1.27 (1.11–1.49)	1.24 (0.98–1.47)	0.96 (0.77–1.20)	0.94 (0.70–1.23)	1.15 (0.94–1.29)	Extra	Extra
28	0.85 (0.67–1.32)	1.40 (1.13–1.93)	1.05 (0.95–1.23)	0.78 (0.61–0.93)	1.21 (0.84–1.32)	Extra	Extra
29	1.03 (0.74–1.77)	1.35 (0.82–2.17)	1.03 (0.74–1.46)	1.14 (0.37–1.83)	0.99 (0.45–1.44)	Extra	Extra
30	1.05 (0.17–1.65)	3.76 (2.49–4.92)	0.95 (0.36–1.71)	0.90 (0.24–1.80)	6.48 (5.12–6.94)	2nd choice	2nd choice
31	0.99 (0.77–1.23)	0.66 (0.27–0.85)	1.19 (0.59–1.51)	1.12 (0.96–1.47)	1.00 (0.72–1.16)	Extra	Extra
32	0.54 (0.41–0.69)	0.92 (0.64–1.36)	1.03 (0.92–1.14)	1.00 (0.86–1.40)	1.07 (0.96–1.23)	Extra	Extra
33	0.76 (0.29–1.24)	1.22 (0.46–1.85)	0.83 (0.73–1.02)	1.09 (0.74–1.50)	1.01 (0.58–1.30)	Extra	Extra
34	0.75 (0.33–1.24)	0.86 (0.51–1.16)	0.96 (0.86–1.08)	1.17 (1.04–1.49)	1.22 (0.95–1.69)	Extra	Extra
35	1.01 (0.79–1.24)	2.60 (2.30–2.99)	1.03 (0.87–1.18)	1.12 (1.06–1.17)	4.24 (4.05–4.44)	Extra	Extra
36	1.18 (1.05–1.54)	1.19 (0.42–2.04)	2.74 (2.60–2.86)	2.46 (2.36–2.61)	4.87 (4.68–5.04)	1st choice	1st choice
37	1.30 (0.91–1.64)	0.85 (0.49–1.58)	1.06 (0.89–1.23)	4.28 (3.86–4.71)	1.10 (0.91–1.33)	1st choice	1st choice
38	0.76 (0.50–1.07)	5.94 (5.54–6.20)	0.68 (0.51–0.97)	0.96 (0.79–1.09)	2.49 (2.41–2.59)	2nd choice	2nd choice
39	1.04 (0.63–1.32)	1.12 (0.75–2.56)	0.94 (0.75–1.19)	0.83 (0.74–0.94)	2.40 (2.11–2.66)	Extra	Extra
40	1.04 (0.66–1.44)	1.04 (0.17–1.37)	0.95 (0.74–1.16)	0.99 (0.80–1.24)	3.32 (2.95–3.88)	1st choice	Extra

Table 3 (continued)

Sample number	E-tongue-MLR-SA model performance (repeated K-fold-CV prediction results)					Model predicted trade category ^b	Table olive trade category according to sensory panel analysis ^c
	Estimated sensory defect mean intensities (minimum and maximum values in brackets) ^a						
	<i>Butyric</i> (24 signals E-tongue)	<i>Putrid</i> (29 signals E-tongue)	<i>Zapateria</i> (24 signals E-tongue)	<i>Musty</i> (28 signals E-tongue)	<i>Winey-vinegary</i> (28 signals E-tongue)		
41	6.75 (5.91–7.93)	1.02 (0.56–1.73)	0.74 (0.27–1.15)	4.90 (4.62–5.20)	0.77 (0.18–1.00)	2nd choice	NC
42	7.48 (7.31–7.64)	0.80 (0.19–1.46)	1.23 (1.02–1.58)	5.04 (4.77–5.24)	0.86 (0.57–1.26)	NC	NC
43	0.92 (0.44–1.30)	0.66 (0.00–1.42)	1.06 (0.60–1.51)	10.11 (9.40–10.94)	0.85 (0.29–1.67)	NC	NC
44	8.37 (8.08–8.78)	0.82 (0.82–1.12)	0.82 (0.60–0.93)	7.04 (6.81–7.45)	1.00 (0.73–1.33)	NC	NC

Mean intensities of the sensory defects predicted by the E-tongue-MLR-SA models and respective minimum and maximum values (in brackets)

^a E-tongue-MLR-SA models: multivariate linear regression models described in Table 2. Each sample was included 10 times into the validation dataset (composed at least by 25% of the data), according to the repeated K-fold-CV procedure used. Mean, minimum, and maximum values reported correspond to the predicted intensities of sensory attribute when that particular sample was included in the internal validation dataset (not used for training purposes)

^b Table olive trade category classification according to the predicted mean intensities of the defect predominant perceived (DPP), predicted by the E-tongue-MLR-SA model: extra, DPP ≤ 3 ; 1st choice, 3 < DPP ≤ 4.5 ; 2nd choice, 4.5 < DPP ≤ 7 ; NC: olives that cannot be sold as table olives, DPP > 7 (IOC 2011). Misclassified samples are identified in bold

^c Table olive trade category classification according to median intensity of the defect predominant perceived (DPP), assessed by the sensory panel (Marx et al. 2017a): extra, DPP ≤ 3 ; 1st choice, 3 < DPP ≤ 4.5 ; 2nd choice, 4.5 < DPP ≤ 7 ; NC: olives that cannot be sold as table olives, DPP > 7 (IOC 2011)

models, show a higher variability compared to the overoptimistic LOO-CV technique. Nevertheless, the variability observed for each organoleptic negative attribute (symbol “○” in Figs. 1b, 2b, 3b, 4b, and 5b) is of the same order of magnitude or even lower in comparison with the variability of the individual intensity evaluation performed by the 8 trained panelists during the sensory analysis of each table olive sample (olives and respective brine solutions), the range of intensities of which is given in Table 1 and plotted in Figs. 1b, 2b, 3b, 4b, and 5b (horizontal lines, symbol “—,” indicating the minimum and maximum intensities perceived by the sensory panel). This finding strengthens the proposal that the E-tongue-MLR-SA technique may indeed mimic the overall performance of the sensory panel. Furthermore, it is interesting to verify that the proposed electrochemical quantitative tool, despite the variability obtained in the predictions of the intensity of each defect, could be also used to correctly classify the majority of the commercial table olives according to the sensory quality trade category recommended by the IOC (2011): extra, 1st choice, 2nd choice, or olives that cannot be sold as table olives (NC). In fact, based on sensory defects’ predicted mean for each sample of table olives studied (Table 3), a trade category classification according to the E-tongue-MLR-SA models could be attributed. The results shown in Table 3 allowed determining the classification prediction ability of the proposed electrochemical-chemometric approach by comparing the trade categories determined by the model and those attributed based on the sensory panel evaluation (Marx et al. 2017a), which are assumed as the true classifications. The results pointed out that only 6 table olive samples were misclassified resulting into overall sensitivity of 86.4% and a specificity of 88.2%. In more detail, table olive samples were correctly classified into extra, 1st choice, 2nd choice, or NC categories with a successful rate of 78, 92, 100, and 83%, respectively, confirming the potential qualitative capability of the proposed methodology. It should be remarked that the reported results refer to the predictive performance of the model assessed from the internal validation datasets established according to the repeated K-fold-CV procedure and the overall predictive sensitivity is similar to that reported by Marx et al. (2017a) when E-tongue-LDA-SA classification models were used (86% \pm 9%).

Finally, as suggested by Roig and Thomas (2003a, b), the potential of applying the E-tongue-MLR-SA models as a complementary practical tool was evaluated for assessing table olives’ negative organoleptic attributes, aiming to reduce the number of samples that must be evaluated by trained sensory panels, which is assumed as the reference sensory method (IOC 2011). For that, it was evaluated if the slope and intercept values of the single linear regression model established between the intensities predicted by the E-tongue-MLR-SA and the median intensities perceived by the sensory panel were statistically equal to 1 and 0 (as expected theoretically

Table 4 Parameters of the single linear regression established between the values of intensities provided for defects (LOO-CV and repeated *K*-fold-CV) by E-tongue-MLR-AS model and the respective values of median intensity of perceived defects by the panel of tasters for the 5 negative attributes detected in table olive samples (3 and 2 that fall under the category of other defects) (IOC 2011): coefficient of determination (R^2); slopes, intercept values, and respective confidence intervals (CI) at 95%

Regression line parameters	Abnormal fermentation defects			Others defects				
	Butyric (24 signals E-tongue)		Putrid (29 signals E-tongue)	Zapateria (24 signals E-tongue)		Winey-vinegary (28 signals E-tongue)	Musty (28 signals E-tongue)	
	LOO-CV	Repeated <i>K</i> -fold-CV ^c	LOO-CV	Repeated <i>K</i> -fold-CV ^c	LOO-CV	Repeated <i>K</i> -fold-CV ^c	LOO-CV	Repeated <i>K</i> -fold-CV ^c
R^2	0.998	0.965	0.995	0.961	0.985	0.994	0.998	0.982
Slope	0.990	0.989	1.002	0.998	0.976	0.999	1.000	1.006
Slope CI ^a	[0.954; 1.026]	[0.971; 1.006]	[0.970; 1.034]	[0.979; 1.017]	[0.922; 1.029]	[0.991; 1.008]	[0.981; 1.019]	[0.993; 1.020]
Intercept	0.020	0.031	-0.003	-0.001	0.021	0.002	0.004	-0.0003
Intercept CI ^b	[-0.074; 0.114]	[-0.015; 0.077]	[-0.100; 0.095]	[-0.058; 0.056]	[-0.065; 0.107]	[-0.022; 0.026]	[-0.047; 0.055]	[-0.034; 0.034]

LOO-CV leave-one-out cross-validation

^a 95% slope confidence interval

^b 95% intercept confidence interval

^c Repeated *K*-fold-CV (4 folds × 10 repeats)

for a perfect linear fit), respectively. Table 4 shows the parameters of the single linear regressions established as well as the determination coefficients (R^2), and the slope and intercept values as well as the respective 95% confidence intervals, for both LOO-CV and repeated *K*-fold-CV techniques, for each of the 5 table olives' sensory defects. The results demonstrate that, at the 5% significance level, the slope and intercept values were statistically equal to the expected theoretic values, since the confidence intervals contain the values 1 and 0, respectively. These results confirmed the potential of the proposed E-tongue-MLR-SA technique for sensory assessment of the intensities of organoleptic negative attributes usually found in table olives, and so, allowing reducing the total number of samples that must be subjected to an organoleptic analysis by a trained sensory panel.

Conclusions

The study carried out showed that the fusion of an electronic tongue with linear multivariate regression models enabled to establish a powerful electrochemical tool that can be used as an effective taste sensor device for quantitatively assessing the intensities of negative sensory sensations perceived by trained sensory panels in table olives. Indeed, the electronic tongue was able to record a representative fingerprint of table olives' most common organoleptic negative sensations (intensities of butyric, musty, putrid, winey-vinegary, and/or zapateria defects). Also, it was shown that the proposed quantitative electrochemical-chemometric tool could be satisfactorily used to classify table olive samples according to their trade categories, with a successful classification rate similar to that achieved when supervised classification models were applied. The overall satisfactory performance achieved associated to the simplicity of the proposed procedure, and its low cost and low analysis time, which enables a daily minimum analysis of 40 table olive samples per device (olive aqueous pastes plus brine solutions), may hopefully result in its practical use for table olive sensory analysis, contributing to the legal implementation of the International Olive Council recommendations regarding the commercial classification of table olives according to sensory analysis.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

Ethical Approval This article does not contain any studies with human participants or animals performed by any of the authors.

Informed Consent Not applicable.

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