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Monitoring olive oils quality and oxidative resistance during storage using an electronic tongue

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ABSTRACT

Monitoring olive oils oxidative stability and quality parameters (free acidity, peroxide values, K_{232} and K_{270} extinction coefficients) is needed to guarantee that, during storage, their levels remain within the legal thresholds enabling their commercialization as high-value extra-virgin olive oils. Physicochemical levels are assessed using time-consuming routine analytical reference techniques. In this work, the feasibility of a novel approach that merges an electronic tongue and chemometric tools, for monitoring extra-virgin olive oils' quality along one year of storage at dark or exposed to light is discussed. The results confirmed that physicochemical parameters varied with the storage lighting conditions and more significantly with time. Also, multiple linear regression models, using sub-sets of 22–28 sensors selected with a meta-heuristic simulated annealing algorithm, allow evaluating the storage time-evolution of olive oils' peroxide values, extinction coefficients and oxidative stabilities with satisfactory accuracy ($R^2 \geq 0.98$ and ≥ 0.96, for leave-one-out and repeated K-fold cross-validation procedures, respectively). The capability of monitoring, in a single electrochemical assay, legal required quality parameters of olive oils, decreases considerable the analysis time and cost, allowing checking the compliance of extra-virgin olive oil quality with labeling. So, the use of electronic tongues for extra-virgin olive oil shelf-life assessment could be envisaged.

1. Introduction

Extra-virgin olive oils (EVOO) are quite appreciated by consumers due to their quality, sensory attributes and health benefits. So, as pointed out in the literature, there still is a commercial need to develop fast, portable and low-cost analytical methods for guaranteeing olive oil commercial category namely to distinguishing EVOO from virgin and lampante olive oils (VOO and LOO, respectively). Olive oils physicochemical parameters have been shown as important markers for quality assessment and olive oil grade discrimination, minimizing the risk of incorrect or abusive olive oils labeling (Garcia, Martins, & Cabrita, 2013; Sinelli, Cerretani, Di Egidio, Bendini, & Casiraghi, 2010). However, besides olive cultivar, edapho-climatic conditions, harvesting and technological procedures, EVOO's physicochemical quality is also greatly influenced by storage conditions, namely time, temperature, type of packing material, exposition to air and/or to light (Abbadi et al., 2014; Ayyad et al., 2015; Ben-Hassine et al., 2013; Bubola, Koprivnjak, Sladonja, & Belobrajić, 2014; Caponio et al., 2013; Cossignani, Luneia, & Damiani, 2007; Fadda et al., 2012; Gómez-Alonso, Mancebo-Campos, Salvador, & Fregapane, 2007; Jabeur, Zribi, Abdelhedi, & Bouaziz, 2015; Pristouri, Badeka, & Kontominas, 2010). Indeed, the levels of olive oils physicochemical quality parameters, such as the ultra-violet light absorption extinction coefficients (K_{232} and K_{270}), free acidity (FA) and peroxide value (PV) may significantly increase during storage

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(Abbadi et al., 2014; Afaneh, Abbadi, Ayyad, Sultan, & Kanan, 2013; Fadda et al., 2012; Jabeur et al., 2015; Mendéz & Falqué, 2007; Rababah, Feng, Yang, Eriefej, & Al-Omoush, 2011; Stefanoudaki, Willians, & Harwood, 2010), those of the oxidative stability (OS) decrease (Stefanoudaki et al., 2010) and so, an olive oil classified as extra-virgin when bottled may suffer degradation during storage resulting in an inferior quality grade when purchase and consumed. In fact, olive oils quality changes are inevitable and start immediately after the olive oil extraction due to lipid oxidation, which may lead to rancidity (Ben-Hassine et al., 2013; Vacca, Del Caro, Poiana, & Piga, 2006) or to hydrolytic degradations causing partial loss of healthy minor constituents (Dabbou et al., 2011). Therefore, new analytical methods aiming to ensure the compliance of olive oil quality with labeling is of utmost relevance for olive oils producers and consumers (Abbadi et al., 2014). This aim could be accomplished by the development of simple, green, user-friendly and low-cost analytical devices that could provide fast assessment and monitoring of the physicochemical quality parameters of olive oils, which could be implemented as complementary or alternative methods to the time-consuming classical analytical reference techniques. For example, this need as motivate the development of simple, expeditious and economic techniques compared to the expensive and time consuming classical chromatographic techniques, like the use of Fourier transform infrared (FTIR) spectroscopy combined with chemometrics as a rapid tool to predict phenol content and antioxidant activity of olive fruits and oils (Machado et al., 2015) or the use of Ion Mobility Spectrometry (IMS) for assessing the stability and quality of single-variety EVOO over storage (Garrido-Delgado et al., 2015). Several sensor approaches, based on the use of electronic tongues, noses and/or eyes (E-tongues, E-noses and E-eyes, respectively), have been reported for olive oils qualitative and/or quantitative electrochemical characterization. These studies have proven the potential of single or fusion methodologies between E-tongues, E-noses and/or E-eyes regarding organoleptic characterization (Apetrei, Gutierez, Rodríguez-Méndez, & de Saja, 2007; Apetrei, Apetrei, Villanueva, de Saja, & Gutierrez-Rosales, 2010; Apetrei, Ghasemi-Varnamkhasti & Apetrei, 2016; Apetrei, Rodríguez-Méndez, Parra, Gutierrez, & de Saja, 2004; Rodríguez-Méndez, Apetrei, & de Saja, 2010; Veloso, Dias, Rodrigues, Pereira, & Peres, 2016), olive oil quality levels discrimination (Apetrei & Apetrei, 2013; Apetrei, Rodríguez-Méndez, & de Saja, 2005; Escuderos, Sánchez, & Jiménez, 2011, 2010; García-González & Aparicio, 2004; Oliveri, Baldo, Daniele, & Forina, 2009), olive oil geographical origin (Apetrei et al., 2010; Cosio, Ballabio, Benedetti, & Gigliotti, 2006; Haddi et al., 2013, 2011; Oliveri et al., 2009) or monovarietal olive oil classification according to olive cultivar (Cimato et al., 2006; Dias, Rodrigues, Veloso, Pereira, & Peres, 2016a; Dias et al., 2014). Moreover, the feasibility of applying electrochemical devices for evaluating polyphenolic contents in olive oils have been successfully reported (Apetrei & Apetrei, 2013; Rodríguez-Méndez, Apetrei, & de Saja, 2008) as well for the capability of E-noses and/or E-tongues to indirectly qualitatively assess the oxidation of EVOO at different storage periods and conditions, allowing differentiating olive oil samples stored under different light conditions and storage time-periods (Cosio, Ballabio, Benedetti, & Gigliotti, 2007). So, the implementation of cost-effective sensor-based devices for monitoring EVOO physicochemical quality along the commercialization line and storage, aiming to verify if their quality indexes still meet the required legal thresholds (FA $< 0.8\%$ oleic acid; PV < 20 mEq O_2/kg; $K_{232} < 2.5$ and $K_{270} < 0.22$ according to the Commission regulation (ECC) nº 2568/91) for being commercialized as high-value EVOO, is still an on-going exciting and challenging research topic. In this work, the feasibility of applying an E-tongue (with nonspecific cross-sensitivity lipid membranes) and multiple linear regression (MLR) models, established using a simulated annealing (SA) variable selection algorithm, to simultaneously (i.e., in a single-run assay) quantify PV, K_{232} and K_{270} levels as well as oxidative stability (OS) of bottled EVOO, during one-year of storage (0, 3, 6, 9 and 12 months) and under different lighting conditions (kept at dark or exposed to light) aiming to simulate usual commercial storage of olive oils, is discussed. So, the work aimed to demonstrate the possibility of applying the E-tongue as a fast and cost-effective novel strategy for quantifying quality parameters and OS values of olive oils in a single-run, expanding the demonstrated capability of the electrochemical device for qualitative and semi-quantitative olive oils assessment, namely for monovarietal EVOO discrimination according to olive cultivar (Dias et al., 2016a, 2014; Peres, Veloso, Pereira, & Dias, 2014) or the classification according to sensory intensity perception of positive organoleptic sensations (Veloso et al., 2016).

2. Materials and methods

2.1. Olive oil samples

Thirty six dark amber glass bottles of blend EVOOs, from the same lot, produced at the north of Portugal (Mirandela region), were studied. The selected lot was an extra virgin olive oil with the Protected Designation of Origin (PDO) qualification "Azeite de Trás-os-Montes" PDO. These olive oils were obtained from olives (mainly from cultivars Cobrançosa and Verdeal Transmontana, with a 10% percentage of olives of cv Madural, according to the producer information) collected at the initial maturation indexes (1–3) and extracted at low temperatures (~22 °C). Four olive oil's different bottles were analyzed immediately after packing, regarding to physicochemical parameters (FA, PV, K_{232} and K_{270} extinction coefficients as well as ΔK values, and OS) as well as electrochemically. The other 32 bottles were stored in a laboratory at ambient temperature (during the one-year of storage the temperature varied in the range of 17 °C to 25 °C) in conditions that tried to mimic real-storage conditions of supermarkets, during a one-year storage period (3, 6, 9 and 12 months; being 8 bottles picked and analyzed at each time-period). Also, two different lighting conditions were studied: 16 bottles were stored at dark, protected from any exposition to daylight or artificial light from a lamp; and, the other 16 bottles were stored in lab open shelves exposed to natural daylight (that entered throw 3 windows but without any direct exposition to sun) and also to artificial lightness from 8 fluorescent lamps (lamps Phillips TL-D36W/840) that remained lit 14h per day, trying to mimic the environmental typical conditions of storage supermarket facilities. Each lamp provided a luminous flux of 3250 lm (according to the manufacturer information), which illuminated a 6×9 m^2 laboratory, corresponding to approximately 482 lux. At each storage period olive oils samples were also evaluated physicochemically and electrochemically. Throughout this work, lighting conditions will be coded as "Dark" and "Light" corresponding to olive oils stored in darkness and olive oil kept in shelf exposed to natural and artificial usual light. Concerning the storage date code T0 is used for fresh olive oil (not stored, analyzed just after being packed) and T3, T6, T9 and T12 for olive oils stored during 3, 6, 9 and 12 months.

2.2. Olive oils physicochemical quality parameters and oxidative stability evaluation

The olive oil's quality parameters assessed were the free acidity (FA), peroxide value (PV) and the specific coefficients of extinction at 232 and 270 nm (K_{232}, K_{270}, and ΔK). All the mentioned quality parameters were determined according to European Union