Extra virgin olive oil: a comprehensive review of efforts to ensure its authenticity, traceability, and safety

Julián Lozano-Castellón [a,b,1], Anallely López-Yerena [a,1], Inés Domínguez-López [a,b], Aina Siscart-Serra [a], Nathalia Fraga [a], Samantha Sámano [a], Carmen López-Sabater [a,b], Rosa M Lamuela-Raventós [a,b], Anna Vallverdú-Queralt [a,b] *, Maria Pérez [a,c] *

[a] Department of Nutrition, Food Science and Gastronomy XIA Faculty of Pharmacy and Food Sciences, Institute of Nutrition and Food Safety (INSA-UB), University of Barcelona, 08028 Barcelona, Spain

[b] Consorcio CIBER, M.P. Fisiopatología de la Obesidad y la Nutrición (CIBERObn), Instituto de Salud Carlos III (ISCIII), Madrid, Spain.

[c] Laboratory of Organic Chemistry, Faculty of Pharmacy and Food Sciences. University of Barcelona, Spain.

[1] authors contributed equally to the work

***Corresponding Authors:** Department of Nutrition, Food Science and Gastronomy, XIA, INSA, Pharmacy School, Av Joan XXIII s/n, University of Barcelona, Barcelona, Spain, Phone: +34 934024508; M.P: mariaperez@ub.edu ; A.V-Q.: avallverdu@ub.edu

Word count: 15017

Short version of title (running head): EVOO biomarkers of authencity and health

Abstract

The growing demand for extra virgin olive oil (EVOO), appreciated for its unique organoleptic properties and health benefits, has led to various fraudulent practices to maximize profits, including dilution with lower value edible oils. The adulterated oils would be of poor nutritional quality, more readily oxidized, and may contain unhealthy substances formed during processing. Nevertheless, the range of available techniques to detect fraud in EVOO production has been growing. Reliable markers of EVOO adulteration include fatty acids and minor components such as sterols, tocopherols, triterpene alcohols, phenolic compounds, phospholipids, volatile compounds, and pigments. Additionally, increasing consumer interest in high-quality EVOO has led to the development of robust scientific methods for its traceability.

This review focuses on i) the usefulness of certain compounds as markers of EVOO adulteration; ii) the potential health risks of consuming adulterated EVOO; and iii) reliable methods for the geographical traceability of olive oil.

In conclusion, fraudulent production practices need to be detected to preserve the beneficial health effects of EVOO and to avoid the potential risks associated with ingesting substandard oil. In this work, as EVOO certification and regulatory framework limitations have already been extensively reviewed, we focus our attention on biomarkers that guarantee both the authenticity and traceability of oil, and consequently its health properties. When is unavailable to obtain a high resolution mass spectrometry full fingerprint, stigmastadienes and the sterolic profile are proposed as reliable markers.

Keywords: Authenticity, botanical traceability, geographic traceability, biomarkers, food safety, healthy fat.

1. Introduction

Extra virgin olive oil (EVOO), considered to be the highest quality olive oil, is much appreciated for its organoleptic and nutritional properties. Moreover, its consumption has been gaining popularity beyond the Mediterranean area, where it is the main dietary source of fat (De Santis, Cariello, Piccinin, Sabbà, & Moschetta, 2019; Flori et al., 2019). EVOO is mostly composed of triglycerides (TAG) (98%), mainly monounsaturated fatty acids (MUFA) (80%) such as oleic acid (C18:1), which are responsible for its physicochemical properties (Carranco, Farrés-Cebrián, Saurina, & Núñez, 2018; Jimenez-Lopez et al., 2020). In the remaining unsaponifiable fraction (1-2%) stand different compounds which have known health benefits, such as phenolic compounds and triterpenic acids (Cárdeno et al., 2014; Carranco et al., 2018; Criado-Navarro et al., 2021; Gelmini et al., 2016). The nutritional profile of EVOO, particularly its richness in antioxidant and anti-inflammatory phenolic compounds together with its fatty acid (FA) profile, is associated with multiple health benefits, including the prevention of cardiovascular diseases, cancer, neurodegeneration and diabetes (Castro-Barquero et al., 2020; Estruch et al., 2018; García-Gavilán et al., 2018; Guasch-Ferré et al., 2014; Lecerf, 2009; Tresserra-Rimbau et al., 2014).

Although all EVOO is produced following the same basic procedure, its composition and sensory properties vary depending on factors such as the olive variety and ripeness, environmental conditions and the technological methods used in processing, and the storage conditions (Boneza & Niemeyer, 2018; Correa Fuentes, Martín Gordo, Pinzón Reyes, & Cárdenas González, 2017; López-Yerena et al., 2019, 2021, 2020; Polari, Garcí-Aguirre, Olmo-García, Carrasco-Pancorbo, & Wang, 2018). However, to be in EVOO category (instead of virgin olive oil or olive oil), certain criteria must be fulfilled: production should be exclusively by mechanical procedures; free fatty acidity should not be over 0.8%; the oil must not have sensory defects and include fruity attributes (Jimenez-Lopez et al., 2020). EVOO should also have a low peroxide index and specific spectrophotometric values (K_{232}≤2.5, K_{270}≤0.22, ΔK≤0.01), which are indicators not only of quality, but also authenticity (Jimenez-Lopez et al., 2020; Ün & Salim, 2018).

Even though the olive oil sector is highly regulated by the European Union (EU) by Reg. (EEC) 2568/91 as amended (COMMUNITY, 2013b), and the International Olive Council (IOC), which also establishes methods for their analysis, it is known that there are still drawbacks in their analytical methods. (Conte et al., 2020). Authentication of food and the detection of adulteration have been concerns throughout the evolution of food industries (Bansal, Singh, Mangal, Mangal, & Kumar, 2017; Perez, Lopez-Yerena, & Vallverdú-Queralt, 2020; Stadler, Tran, Cavin, Zbinden, & Konings, 2016), and EVOO production is no exception (Carranco et al., 2018; González-Domínguez, Sayago, Morales, & Fernández-Recamales, 2019). Owing to the growing complexity of food production chains, more exhaustive measures and strategies are required to tackle fraudulent attempts by food producers to boost their profits (Creydt & Fischer, 2018).

In this review, we focus on the most recent approaches to detect adulteration of EVOO with lower value oils based on the profiling of FA and minor components such as phenolic compounds, volatile compounds, tocopherols, phospholipids, and pigments, proposing stigmastadienes and sterols as markers for detecting its adulteration. We also look at the potential health risks of consuming adulterated EVOO and describe reliable methods for the geographical traceability of olive oil. Although there are excellent reviews focused on olive oil authentication (Bajoub, Bendini, Fernández-

Gutiérrez, & Carrasco-Pancorbo, 2018) and the weaknesses in the regulatory framework (Conte et al., 2020), our approach draws attention to global traceability organized by biomarkers, not by analytical techniques or type of adulteration and, most importantly, includes a section that highlights the possible risks of ingesting fraudulent EVOO.

2. Fraud, authentication, and traceability of EVOO

2.1 Fraudulent activities in EVOO production

Fraudulent activities in the food industries, a global issue of increasing concern, comprise a wide range of malpractices, including adulteration (dilution, substitution, and unapproved enhancement), mislabeling and smuggling (Stadler et al., 2016). Economic gain is the main reason for committing food fraud, but its significance is not only a question of illegality, as it can lead to adverse health consequences for the consumer (Bansal et al., 2017). An extensive study analyzing the evolution of food fraud over 30 years (1980-2010) found that the type of adulterated food most frequently reported in academic articles is olive oil (Moore, Spink, & Lipp, 2012). As traditional methodologies for identifying such adulterations through the assessment of quality and purity have some limitations (Conte et al., 2020), new analytical techniques need to be developed (Barbieri et al., 2020; Creydt & Fischer, 2018; Damiani, Cavanna, Serani, Dall'Asta, & Suman, 2020; Violino et al., 2021).

The most commonly reported frauds in EVOO production are: (i) mixing EVOO with lower quality oils that are cheaper to produce; (ii) mislabeling, for example, falsely claiming that refined olive oil is EVOO, or concealing the real place of origin; and (iii) price fraud, which consists of reducing the oil price to artificially lower the market value (García Martínez, 2020). Adulteration can be with refined olive oil (Garcia, Martins, & Cabrita, 2013; Torrecilla, Rojo, Dominguez, & Rodríguez, 2010), refined olive−pomace oil (Fragaki, Spyros, Siragakis, Salivaras, & Dais, 2005; Torrecilla et al., 2010), lampante olive oil (Fragaki et al., 2005) and olive pomace oil (Guimet, Ferré, & Boqué, 2005; Yang & Irudayaraj, 2001). Other edible oils from other plants such as hazelnut (Arlorio et al., 2010; Chiavaro, Vittadini, Rodriguez-Estrada, Cerretani, & Bendini, 2008), soybean (da Silveira et al., 2017; Fasciotti & Netto, 2010; Jabeur et al., 2014; Mendes et al., 2015; Milanez et al., 2017; Poiana et al., 2015; Tiryaki & Ayvaz, 2017; Yang & Irudayaraj, 2001), corn (Jabeur et al., 2014), canola (Salivaras & McCurdy, 1992) and sunflower (Jabeur et al., 2014) have also been used.

Different EVOO components have been proposed as possible markers of fraudulent activities, including natural chlorophyll, diacylglycerols (DAG), FA, wax esters, phenolic compounds, and sterols (Garcia et al., 2013; Gómez-Coca, Pérez-Camino, Bendini, Gallina Toschi, & Moreda, 2020).

2.2 Authentication

Consumer demand for authenticity and traceability of food products means that quality certification is of great interest for the food sector. In this context, a range of government legislation and regulations as well as international agency guidelines have been published (Conte et al., 2020; Melucci et al., 2016).

The quality of olive oil is governed by the IOC together with the European Community and the Codex Alimentarius Commission (Stadler et al., 2016), an organization that proposes several analytical methods, including gas chromatography (GC), high performance liquid chromatography (HPLC) or gravimetry, among others. European regulation derives from the IOC and Codex Alimentarius Commission regulations, as well as most of the national regulations, such as Brazilian one. However, there are some countries still lacking internal regulations for EVOO, in the case of USA, the FDA still has no regulation defining this product.

In Europe, to achieve the category of EVOO, the oil must comply with a range of parameters set out in the EU Regulation No. 2568/91 (COMMUNITY, 1991) and consequently modified in the EU Regulation 1348/2013 (COMMUNITY, 2013b), which are assessed by chemical and sensory tests using IOC methods and standards (Jimenez-Lopez et al., 2020). The quality criteria evaluated in olive oil are free acidity, the peroxide index, ultraviolet (UV) absorption (coefficients K232, K270 and ΔK), fatty acid ethyl esters, sensory characteristics (Conte et al., 2020; Jimenez-Lopez et al., 2020). These basic quality parameters, together with composition in fatty acids and sterols as well as profiles in phospholipids, tocopherols, phenolic molecules, volatile compounds etc., can offer information very useful to check authenticity of EVOO and possible applied fraudulent activities to it (Azizian et al., 2015; Mikrou et al., 2020). Some examples are presented in **Table 1** with the analytical techniques and the data processing methods used for their determination.

Authenticity of food or beverages can be established by an exact match between the characteristics, properties and composition of the product and the description on the label. Although the EU closely monitors the olive oil market, the risk of EVOO adulteration remains high due to the economic gains at stake and the increasingly sophisticated techniques employed (Bansal et al., 2017).

Among the EU regulations established to standardize the production and commercialization of EVOO in Europe (Melucci et al., 2016) (the largest producer, exporter, and consumer of olive oil in the world), Regulation EU No. 1019/02 defines its labelling and packaging. Certified labelling is a way of providing the consumer with assurance of food authenticity. The information on EVOO labels also includes its classification as light, medium or intense, according to its sensory attributes.

2.2.1. Fatty acids and glycerides derivatives

FAs are present in the oil as triacylglycerols. Its main component is oleic acid, a MUFA that accounts for 65-80% of the FA content (ranges obtained from: (Konuskan, Arslan, & Oksuz, 2019; Orsavova, Misurcova, Vavra Ambrozova, Vicha, & Mlcek, 2015; Rotondi, Morrone, Bertazza, & Neri, 2021; Vávra, Hájek, & Kocián, 2021; Wang et al., 2019; Yahay, Heidari, Allameh, & Amani, 2021)). Oleic acid is reported to have beneficial cardiovascular effects and, as MUFA, with its single double bond, it is more stable than polyunsaturated fatty acids (PUFA) (Yubero-Serrano, Lopez-Moreno, Gomez-Delgado, & Lopez-Miranda, 2019). Other FA, such as linoleic are much less abundant in EVOO than in other vegetable oils (Jimenez-Lopez et al., 2020). The FA profile of EVOO thus distinguishes it from other edible oils (such as sunflower and peanut oil), as shown in **Figure 1**, and its analysis can provide useful information for the detection of adulteration. However, other edible oils such as rapeseed oil also contain a high proportion of MUFAs. Several studies have compared the FA profiles of EVOO and different oils commonly employed as adulterants, such as sunflower, corn, peanut, coconut, or rapeseed (Aykas, Karaman, Keser, & Rodriguez-Saona, 2020; Mikrou et al., 2020; Yang, Ferro, Cavaco, & Liang, 2013), using GC coupled with a flame ionization detector (FID) or mass spectrometry (MS), the traditional method to evaluate the lipid fraction of EVOO. This methodology, however, requires a pretreatment of the sample to obtain FA methyl esters. Deodorized olive oil in EVOO was successfully detected by the determination of diacylglycerols and free FAs (Gómez-Coca et al., 2020). A comparative study analyzing TAG by electrospray ionization MS (ESI-MS) and FA by GC-FID concluded that the spectroscopic technique was faster and more efficient than the chromatographic method. Applying this novel methodology, olein lineo 10-heptadecenoic was established as a lipid marker for soybean oil added to EVOO (da Silveira et al., 2017).

In recent years, spectroscopic methods have emerged as useful and efficient techniques for the determination of FA; not requiring a derivatization step, they are less time-consuming than chromatographic methods. Thus, a simple technique able to characterize EVOO and detect adulterations with corn oil was developed by Di Girolamo *et al.* (2015) based on matrix-assisted laser desorption/ionization MS (MALDI-TOF MS) with unsupervised hierarchical clustering (UHC), principal component analysis (PCA) and Pearson's correlation analyses. Authentication models for the detection of high linoleic and high oleic vegetable oils in EVOO were developed based on flow injection analysis-heated electrospray ionization-high resolution MS (FIA-HESI-HRMS) combined with partial least squares (PLS)–discriminant analysis (DA), which provided TAG profiles of olive oil samples (Quintanilla-Casas et al., 2021).

The lipid profile of EVOO was also successfully characterized using an untargeted metabolic approach based on flow injection analysis-magnetic resonance MS (FIA-MRMS) and chemometrics (Nikou, Witt, Stathopoulos, Barsch, & Halabalaki, 2020). Indeed, it provided better projection and prediction models than LC-Orbitrap MS, with the additional advantage that it allows simultaneous monitoring of both lipophilic compounds and polyphenols. Moreover, Fourier transform-near infrared (FT-NIR) spectroscopy coupled to chemometric tools such as PLS analysis was able to detect adulterants (i.e., edible oils high in 16:0 (palm oil), 18:1n-9 (palm olein, hazelnut, canola, or high OLA sunflower and safflower oils), 18:2n-6 (soybean, sunflower or corn oil), and ROO) in EVOO (Azizian, Mossoba, Fardin-Kia, Karunathilaka, & Kramer, 2016; Azizian et al., 2015; Azizian, Wang, Li, & Kramer, 2018). Another vibrational technique, FT-Raman, combined with chemometrics (PCA and PLS) accurately classified EVOO samples according to the harvest year, olive variety, geographical origin, and protected designation of origin (PDO), and also detected adulteration with sunflower or waste cooking oil, due to the different unsaturation degree of the FAs (Li et al., 2018; Sánchez-López et al., 2016).

In summary, analytical techniques such as NIR or Raman spectroscopy, are likely to grow in importance and replace traditional chromatographic methods for the detection of adulterants based on FA determination. Overall, they provide efficient results with the advantage of a simpler sample treatment step compared to chromatography, which reduces the time of analysis.

2.2.2. Phospholipids

Olives and olive oil contain a diversity of phospholipids as minor components (Alves, Cunha, Amaral, Pereira, & Oliveira, 2005). The profile of phospholipids, whose concentration in EVOO is lower than in other vegetable oils (Antonelli et al., 2020), can provide a distinct “fingerprint” for traceability and authenticity studies (Alves, Domingues, & Domingues, 2018; Gallina Toschi, Bendini, Lozano-Sánchez, Segura-Carretero, & Conte, 2013).

Phospholipid analysis using an ionic liquid as a matrix and extraction solvent and MALDI-TOF-MS detected the presence of hazelnut oil in EVOO (still detectable at a 1% contamination level) due to a significant increase in phospholipid signals (Calvano, De Ceglie, D’Accolti, & Zambonin, 2012). In another study based on phospholipids analysis, MALDI-TOF MS coupled to UHC and PCA was used to characterize the oil type and was able to reveal very low levels of corn oil in EVOO (as low as 0.5%) (Di Girolamo et al., 2015).

2.2.3. Tocopherols

Tocopherols (vitamin E compounds) are found in seed oils in four different forms: α-, β, γ- and δ-tocopherols (Ergönül & Köseoğlu, 2014) but in EVOO, δ-tocopherol has not been detected, only α-, β-, and γ-tocopherols have been described, with α-tocopherol representing more than 95% of the total tocopherol content (Beltrán et al., 2010). Tocopherol content and composition depend on several agronomic factors, including the cultivar, fruit ripeness and agroclimatic conditions (Beltrán et al., 2010). Some of the efforts developed to combat EVOO fraud have focused on the tocopherol profile as a potential marker able to detect adulteration with high selectivity, sensitivity, and accuracy.

The concentration of tocopherols in EVOO changes when it is adulterated with other vegetable oils, including lower quality olive oils (Omwange et al., 2021). Depending on the adulterating oil, the concentration can increase, when adulterated with sunflower oil for example (Lia, Castellano, Zammit-Mangion, & Farrugia, 2018), or decrease, when adulterated with olive oil (Merás, Manzano, Rodríguez, & de la Peña, 2018). Accordingly, autofluorescence excitation-emission profiles combined with multi-way classification allowed approximately 15% of olive oil and 3% of olive pomace oil to be detected in EVOO (Merás et al., 2018). Front-face fluorescence spectroscopy coupled with UV-induced fluorescence imaging differentiated between pure EVOO and adulterated oils based on a specific region of excitation emission matrices (between 300 and 600 nm) corresponding to tocopherols, tocotrienols, phenolic compounds, oxidation products, and vitamin E (Omwange et al., 2021). The dilution of Maltese

EVOO with several vegetable oils (corn, soybean, linseed, and sunflower) was identified by fluorescence spectrometry combined with PCA, PLSR and an artificial neural network (Lia et al., 2018). In another study, the authenticity of EVOO was assessed using the α/β tocopherol ratio and the presence of δ-tocopherol to detect the fraudulent addition of oils from other sources (Chen et al., 2011).

Tocopherols have antioxidant properties and can therefore be detected by means of electroanalytic methods (Apetrei & Apetrei, 2014; Tsopelas, Konstantopoulos, & Kakoulidou, 2018). Voltametric fingerprinting of EVOO can reveal changes in the concentration of electroactive compounds such as tocopherols. A voltametric e-tongue successfully detected adulterations of olive oil with less than 10% of sunflower, soybean and corn oils (Apetrei & Apetrei, 2014). In another study, voltametric fingerprinting combined with PLS-DA provided a clear discrimination between olive oils (extra virgin and regular) and olive pomace/ seed oils (Tsopelas et al., 2018). This assumption is supported by the much lower tocopherol content of olive oil compared to seed oils (Kamal-Eldin, 2006).

In summary, the advantage of determining the tocopherol profile is that it requires only minimal sample preparation, such as oil dilution (Chen et al., 2011; Lia et al., 2018), obtaining methanolic extracts (Tsopelas et al., 2018) or directly analyzing the EVOO (Apetrei & Apetrei, 2014; Merás et al., 2018), before subsequent analysis by fluorescence and/or voltammetry with chemometrics. One of the least used approaches for the detection of EVOO adulteration, tocopherol fingerprinting likely deserves wider application.

2.2.4. Phenolic compounds

The main phenolic compounds in EVOO are secoiridoids, which are accompanied by phenolic acids, lignans, flavonoids and phenolic alcohols (Lozano-Castellón et al., 2020). As secoiridoids are characteristic of the Oleacea family (Jensen, 2002), a reduction in their content could indicate a possible fraudulent blend, either adding EVOO from another cultivar (ie: *Arbequina* is less rich in phenolic compounds than *Picual*) or mixing EVOO with different types of oil. Most techniques for EVOO phenolic analysis are based on liquid-liquid extraction and various determination methods, the most common being liquid chromatography coupled to different detectors, such as a diode array detector (International Olive Council, 2017b), MS (Alessandri, Ieri, & Romani, 2014), or MS in tandem (Lozano-Castellón et al., 2021). Other methods are nuclear magnetic resonance (Olmo-Cunillera et al., 2020) or GC-MS (Chiou et al., 2007), which need an extra step for volatilizing the target compounds.

The squalene and tyrosol concentration allowed pure EVOO to be differentiated from EVOO blended with seed oils, pure EVOO presents higher concentration of both compounds, then when plotting the concentration of tyrosol against the concentration of squalene, the blended samples are located near the origin of the plot (0,0) and it is possible to identify them (Hayakawa et al., 2020). Using an e-tongue, it was possible to discriminate between pure EVOO and EVOO containing sunflower, soybean and corn oils, based on the specific electrical properties of phenolic compounds and their lower concentration in the adulterated samples (Apetrei & Apetrei, 2014). Using a partial least squares regression (PLSR) model generated from the HPLC-UV spectrum of the phenolic extract, monovarietal *Arbequina* EVOO was accurately distinguished from the same variety mixed with *Picual* EVOO, refined olive oil or sunflower oil (Carranco et al., 2018). In a recent study, a PLSR model generated from fluorescence spectra of the phenolic profile was used to discriminate between pure EVOO and EVOO blended with olive oil (Omwange et al., 2021). Finally, EVOO adulterated with refined olive pomace oil was identified based on the phenolic and sterol content, as these compounds are more hydrophilic in the pomace after the first oil extraction than in EVOO (Drira et al., 2020).

The EVOO phenolic profile can be affected by numerous factors, such as the olive cultivar, agronomic techniques (López-Yerena et al., 2019), the extraction procedure (López-Yerena et al., 2021) and storage (Castillo-Luna, Criado-Navarro, Ledesma-Escobar, López-Bascón, & Priego-Capote, 2021). As refined vegetable oils contain much lower content of phenolic compounds (Orozco-Solano, Priego-Capote, & Luque de Castro, 2011), their usage to adulterate EVOO will result in a lower phenolic concentration, but not the addition of a new phenolic compound that could potentially serve as a marker. Hence, this type of adulteration is difficult to assess based on phenolics alone, as their concentration can also be reduced depending on the variety, by an extraction parameter or improper storage, so they should be targeted along with other compounds (Nikou et al., 2020).

2.2.5. Volatile compounds

EVOO is highly appreciated by consumers, mostly for its pleasant aroma and characteristic flavor. Aroma depends on the volatile fraction, which differs according to the olive variety, environmental growing conditions, and technological factors during processing operations (Cecchi, Migliorini, & Mulinacci, 2021). Typical flavors and off-flavor compounds that affect the volatile fraction of EVOO are produced by different mechanisms. Positive odors are mainly generated from the oxidation of linoleic and linolenic acids by enzymes of the lipoxygenase pathway, which are released when the fruit is crushed and have a major impact during malaxation. Conversely, the main defective or off-flavors are due to sugar fermentations, amino acid conversion, enzymatic activities of molds (musty), anaerobic microorganisms (muddy). They are also the result of auto- and photo-oxidation of FA during EVOO storage, which produces linear acids, alcohols, esters and ketones (Cecchi et al., 2021; Clodoveo, Hbaieb, Kotti, Mugnozza, & Gargouri, 2014).

Oxidation is the principal cause of the deterioration of olive oil quality. Fatty acids are the fraction most vulnerable to oxidation, their degradation leading to the production of carbonyl compounds and subsequent development of unpleasant flavors and oxidative rancidity (Gargouri, Zribi, & Bouaziz, 2015; Sanmartin et al., 2018; Silva, Anjos, Cavalcanti, & Celeghini, 2015). Autoxidation can occur even in the absence of light by a free radical mechanism in which the absorption of oxygen results in the formation of hydroperoxides. These labile compounds evolve to produce a complex mixture of volatile compounds such as aldehydes, ketones, hydrocarbons, alcohols, and esters, which negatively affect the flavor of olive oil (Frankel, 2014). In fact, the "extra virgin" or even "virgin" designation is granted only if lipid oxidation products such as hydroperoxides do not exceed a stipulated limit and/or produce rancid flavors (Hrncirik & Fritsche, 2005). Positive odors and/or volatile oxidation compounds can also be used as markers of EVOO adulteration (Zhou, Zhang, Chen, Li, & Han, 2021).

The addition of other edible oils dilutes both the aroma and color of EVOO (Violino et al., 2021). However, to date, the technological determination of volatile organic compounds (VOCs) is not required to authenticate EVOO, even though they form an intrinsic part of the quality of the product and the perceived intensity of positive sensory attributes (Violino et al., 2021). There is clearly a need for reliable and inexpensive methods that can rapidly assess the VOC profile of EVOO on an industrial scale.

Over the last decade, numerous studies have targeted VOCs to identify EVOO adulteration with lower value oils such as corn, soybean, sunflower, high oleic sunflower, olive, soft-refined olive and refined olive oils (Azizian et al., 2015; Damiani et al., 2020; Drira et al., 2021; Ozcan-Sinir, 2020; Van Durme & Vandamme, 2016; Violino et al., 2021; Zhou et al., 2021). The techniques applied include GC-MS, GC-FID, GC–olfactometry-MS (GC-O-MS), GC ion mobility spectrometry (GC-IMS), flash GC electronic nose (FGC E-nose), thermogravimetric-GC/MS (TGA-GC/MS), selected ion flow tube MS (SIFT-MS), FT-NIR and vis–NIR (**Table 2**). Among the strategies to isolate VOCs for analysis by GC-MS are purge and trap extraction (Drira et al., 2021), non-thermal plasma treatments (Van Durme & Vandamme, 2016) and profiling of the headspace composition (Damiani et al., 2020). On the other hand, for analysis by SIFT-MS, a temperature-controlled water bath at 30 °C for 30 min was used to release volatile compounds until an equilibrium was reached at the headspace (Ozcan-Sinir, 2020). The application of PCA, PLS1, SIMCA, PLSR and artificial intelligence to spectral data successfully predicted and determined adulterated samples.

In summary, a strong reduction of pentanal and hexanal and its derivative compounds (which contribute to the green odor notes) has been observed in adulterated EVOO (Violino et al., 2021). The higher amounts of PUFA in oils other than EVOO (e.g., sunflower oil) result in a higher rate of volatile oxidation than observed for some oxidation markers (Van Durme & Vandamme, 2016). Although the composition of the volatile fraction of EVOO is not a legal requirement to guarantee its authenticity, this approach could be introduced as a useful tool to support EVOO quality.

2.2.6. Pigments

The color of EVOO is attributed to lipophilic chlorophyll and carotenoid pigments present in the olive fruit (Uncu & Ozen, 2020). EVOO contains a rich variety of chlorophyll derivatives (chlorophyll a and b, pheophytin a and b, and other minor derivatives) and carotenoids (β-carotene, lutein, violaxanthin, neoxanthin and other xanthophylls) (Lazzerini, Cifelli, & Domenici, 2017; Uncu & Ozen, 2020). The presence of pigments depends on the olive variety, the stage of fruit ripeness, environmental growing conditions, the extraction process and storage conditions (Giuffrida, Salvo, Salvo, La Pera, & Dugo, 2007). Pigment profiling of EVOO has been applied as an indication of quality and/or authenticity. Using this approach, corn, rapeseed, soybean, peanut, sunflower, refined olive oil, olive pomace oil and/or old olive oils have been detected as adulterants in EVOO (Ali et al., 2018; Aroca-Santos, Cancilla, Matute, & Torrecilla, 2015; Aroca-Santos, Cancilla, Pariente, & Torrecilla, 2016; Ferreiro-González et al., 2017; Merás et al., 2018; Milanez et al., 2017; Shi et al., 2019; Tan et al., 2018; Uncu & Ozen, 2019).

A clear reduction in chlorophylls and carotenoids in adulterated EVOO has been evaluated by UV-vis, fluorescence spectroscopy and/or FT-IR + UV–vis, which are quick and reliable methods (Ali et al., 2018; Aroca-Santos et al., 2015; Ferreiro-González et al., 2017; Merás et al., 2018; Milanez et al., 2017; Uncu & Ozen, 2019). In this field, stimulated Brillouin scattering combined with UV–vis-NIR (Shi et al., 2019) and front-face fluorescence and visible spectroscopy (Tan et al., 2018) have also been proposed for the authentication of EVOO and the detection of adulteration with other vegetable oils.

Regarding chemometric analysis, PLS, PLSR, partial least squares-Jack-Knife algorithms (PLS-JK), successive projections algorithm-multiple linear regression, stepwise multiple linear regression (SW-MLR) and/or genetic algorithm-multiple linear regression (Ali et al., 2018; Milanez et al., 2017; Tan et al., 2018) have been used. In addition, artificial neural networks (Aroca-Santos et al., 2015), orthogonal projection to latent structures-DA (Uncu & Ozen, 2019), linear-DA (LDA) (Ferreiro-González et al., 2017), parallel factor analysis and discriminant unfolded PLS (Merás et al., 2018) models have been used to identify and quantify adulteration in EVOO samples.

In summary, EVOO adulteration can be determined by comparing visible absorption spectra. In the visible spectrum of EVOO, certain absorption bands in the range of 430–480 nm and 670 nm stand out owing to the presence of various carotenoid and chlorophyll pigments (Ferreiro-González et al., 2017). Adulteration of EVOO with other seed and/or vegetable oils, can be differentiated spectroscopically by the intensity of carotenoids and chlorophylls absorption peaks, as shown in **Figure 2**.

2.2.7. Sterol, triterpene dialcohol and stigmastadiene composition

Sterols and triterpene dialcohols are among the parameters used to officially establish the purity of olive oil. Their composition permits discrimination between olive and other oils and between olive pomace oils and non-solvent-extracted olive oils such as EVOO (International Olive Council, 2021). The addition of other oils will increase the concentration of those compounds and allow then the discrimination between pure EVOO and adulterated EVOOs (Al-Ismail, Alsaed, Ahmad, & Al-Dabbas, 2010). The official method involves several steps: compound separation from the saponifiable fraction; partial purification by chromatography; and then derivatization and analysis by GC-FID. As this is time- and reagent-consuming, shorter and simpler alternative procedures have been developed (Gorassini, Verardo, & Bortolomeazzi, 2019; Mathison & Holstege, 2013; Valli et al., 2021).

Various studies have focused on the analysis of sterols and triterpene dialcohols to classify different EVOOs and detect their adulteration with other oils, including lower quality olive oil or olive pomace oil. Based on the percentages of campesterol and stigmasterol, adulteration with corn, soybean, sunflower and cotton oils was identified (Al-Ismail et al., 2010), whereas uvaol and erythrodiol revealed the presence of olive pomace oil (Mathison & Holstege, 2013). Δ7-stigmastenol and campesterol proved to be effective markers of EVOO adulteration with sunflower and corn oils, respectively (Jabeur et al., 2014). Another study confirmed this usage of Δ7-stigmastenol as well as campesterol as a marker of adulteration with soybean oil (Youseff, Soubh, & Alassaf, 2014). The combination of several parameters, namely total sterol content, desmethylsterol composition and triterpene dialcohols (erythrodiol and uvaol), was successfully used to identify EVOO adulteration with canola, corn, peanut, safflower, soybean, and sunflower oils, but this strategy failed to detect hazelnut oil (Srigley, Oles, Kia, & Mossoba, 2016). The mean values of each sterol allowed EVOOs to be differentiated according to the olive variety and oleaster cultivar, including hybrids (Manai-Djebali et al., 2021). Sterol concentration could differ significantly from different varieties, in the case of Δ5-avenasterol for example, was reported to be between 2.2 and 15.2 % of all sterolic fraction (Manai-Djebali, Oueslati, Martínez-Cañas, Zarrouk, & Sánchez-Casas, 2018). Finally, a new method that analyses free and esterified sterols and free and esterified triterpenic alcohols was able to detect the adulteration of EVOO with only 2% sunflower oil (Valli et al., 2021).

Studies investigating ways of detecting EVOO adulteration have also targeted sterols together with other compounds. Using ^{1}H NMR spectroscopy with lipid signal suppression, Ruiz-Aracama *et al.* (2017) classified EVOOs according to various signals, including those of sterols. A PCA model based on sterol and FA profiles clearly separated EVOO containing cotton and sunflower oils (Kesen, 2019).

Thus, the sterol profile and fatty alcohols have demonstrated to be effective markers for the detection of EVOO adulteration. The official method for their determination has been improved upon, with the development of quicker and more ecological alternatives that require fewer sample preparation steps and consume less reactant to achieve similar results (Mathison & Holstege, 2013; Tena, Wang, Aparicio-Ruiz, García-González, & Aparicio, 2015).

On the other hand, detection of stigmastadienes has been proved to be a highly-sensitive method to notice addition of refined oils to EVOO. Virgin olive oils (VOOs) obtained by cold pressing do not contain enough quantity of these molecules to be measured (less than 0.01 mg/kg^3), and EVOO's are defined by Regulation to contain less than 0.15 mg/kg^3 of stigmastadiene (Uncu & Ozen, 2020). However, during the refining process, the high temperatures to which the oils are exposed, lead to the formation of 3,5-stigmastadiene by the dehydration of sterols, particularly of beta-sitosterol, in measurable amounts ranging from 0.3 to 0.9 mg/kg^3 (Gordon & Firman, 2001). Concretely, the existence of stigmastadienes indicates the use of bleaching clay or high temperature applications executed during the deodorizing of the refining process. Thus, the detection of these steroidal hydrocarbons in VOOs is legally accepted to reveal adulteration of the product by the presence of refined vegetable oils (olive pomace, soybean, sunflower, palm…) (International Olive Council, 2017a;

Schneider, 2016).

Regarding to the stigmastadienes detection, Crews *et al.* (2014) described a test which not only has been shown to be more rapid and easier to apply than other already standardized alternatives, but also occurring to be a method that can detect other sterenes in oils. This method presents a high sensitivity to low levels of stigmastadienes. And even though it does not indicate the exact concentration of refined oil existing, mixtures of less than 5% refined oils can be revealed.

2.2.8. Oil fingerprint applied to the authentication

In recent years, chromatographic and related techniques with spectroscopic detection or coupled to MS, and combined with chemometrics, have proven to have an exceptional capacity to discourse complex food authentication issues by fingerprinting approaches (Medina, Pereira, Silva, Perestrelo, & Câmara, 2019). In this context, flow injection analysis coupled to high-resolution MS (FIA–HRMS), using a fingerprinting strategy and combined with PCA, PLS-DA, and SIMCA was used to distinguish olive oil from other vegetable oils, as well as to perform an evaluation of its quality category (Campmajó, Saurina, & Núñez, 2022). After external validation, remarkable classification accuracies were reached. Moreover, putative identification of the most common ions was performed by HRMS, allowing excellent discrimination of olive oil in front of the other vegetable oil samples using PCA. As mentioned before, similar approach based on TAG profile was satisfactorily used to detect high linoleic and high oleic vegetable oils in EVOO (Quintanilla-Casas et al., 2021).

On the other hand, using chromatographic fingerprints (HPLC coupled to charged aerosol detector and high temperature GC coupled to flame ionization detector) coupled to multivariate techniques was applied to authenticate the geographical origin of EVOO without identifying or quantifying the chemical compounds (Vera, Jiménez-Carvelo, Cuadros-Rodríguez, Ruisánchez, & Callao, 2019). The results were best when fingerprints from the data from the two techniques were combined applying low-level data fusion and PLS-DA was employed as the classification procedure. Similarly, Quintanilla-Casas *et al.* (2020) established fingerprinting as a more efficient approach than profiling sesquiterpene hydrocarbons to classify EVOO according to its origin.

2.2.9. Proposed markers for EVOO adulteration detection

During EVOO adulteration, FA profile and minor compounds profile are changed. Phenolic compounds and volatile compounds decrease, while waxes such as campesterol increase. Hence, monitoring several compounds could be useful to detect adulteration, this is easily achieved recording EVOO fingerprint with HRMS. Nevertheless, this approach requires expensive equipment not always available. Another way of having fingerprints could be monitoring the absorbance or emission spectra in the UV-vis range or using the FT-NIR spectroscopy. However, only active compounds in the NIR or the UV-vis will be detected, hence few information will be obtained compared to the HRMS analysis and it might not be enough for the detection of adulterants.

EVOO is commonly adulterated with refined oils, such as rapeseed or hazelnut, or with solvent extracted oils, such as pomace olive oil. During the deodorization that takes place in the refinement process the oil is heated at 180 ºC or even higher temperatures, this step is present in both chemical and physical refinements (Varona et al., 2021). Thus, analyzing markers of heating, such as stigmastadienes or glycyl sters (GE) (Gordon & Firman, 2001; Kamikata et al., 2019), could be a feasible approach to detect EVOO adulterations with refined oils (Crews et al., 2014). On the other hand, for detecting adulteration with solvent extracted oils, analyzing the sterolic profile could be a useful approach, as the concentration of those compounds will increase (Mathison & Holstege, 2013).

In conclusion, when HRMS is not available, analyzing both stigmastadienes and sterols could be a useful and cheap method to detect EVOO adulteration. As those compounds are analyzed for olive oil categorization (COMMUNITY, 1991), no extra equipment is needed for carrying out those determinations.

2.3 Health issues derived from EVOO fraud.

The health benefits associated with EVOO consumption have been extensively demonstrated, particularly its protective effect against cardiovascular (CV) diseases (Covas et al., 2006; Estruch et al., 2018; Guasch-Ferré et al., 2014). This positive impact has been verified by numerous intervention studies focusing on specific markers of CV disease, which show that EVOO intake can prevent or reduce the inflammatory processes associated with chronic-degenerative conditions such as CV-cerebral diseases and cancer (Casas et al., 2017) and benefits plasma lipid levels and lipid oxidative damage (Covas et al., 2006). EVOO supplementation also leads to an improvement of post-prandial glucose and lipid profiles, an anti-atherosclerosis mechanism that can reduce the risk of developing diabetes (Carnevale et al., 2014). Diabetes prevention could be attributed to the antioxidant property of EVOO (Bullo, Lamuela-Raventos, & Salas-Salvado, 2011), since oxidative stress appears to be involved in β-cell dysfunction and down-regulates insulin secretion. Aside from investigating whether EVOO had any effect on the lipid profile, the Carnevale research group examined whether postprandial glycemic control occurred using an oxidative stress-mediated mechanism, demonstrating that a Mediterranean-type meal supplemented with EVOO is associated with reduced postprandial oxidative stress generated by NOX2 (Violi et al., 2015). Thus, the consumption of EVOO represents a simple but effective nutritional approach to modulating the deleterious effect of different CV risk factors on the vascular system, chiefly oxidative stress, inflammation, postprandial hyperglycemia, and hyperlipidemia (Nocella et al., 2018).

Moreover, EVOO intake is inversely associated with the prevalence of cancer (Psaltopoulou, Kosti, Haidopoulos, Dimopoulos, & Panagiotakos, 2011), as reported in a meta-analysis of nineteen observational studies involving approximately 35,000 individuals and carried out within ten years. More recently, a randomized trial found that women who adhered to a Mediterranean diet supplemented with EVOO had a 62% lower incidence of invasive breast cancer than a control group advised to restrict dietary fats (Toledo et al., 2015). Other health properties of EVOO are related to neuroprotection. Pitt *et al.* (2009) reported that low doses of oleocanthal, one of the main secoiridoids of EVOO, structurally altered amyloid-β oligomers, which play a role in the development of Alzheimer’s disease. Moreover, Batarseh *et al.* (2018) reported that high-oleocanthal EVOO reduced the amyloid-β load and related toxicity in a mouse model of Alzheimer’s disease. Additionally, Li *et al.* (2009) al found that oleocanthal inhibited tau fibrillization. Other phenolic compounds present in EVOO, hydroxytyrosol and oleuropein, also showed neuroprotective effects in the prevention of Parkinson’s disease, as they can interfere in the pathophysiology of the disease by reducing the accumulation of neurotoxins, oxidative stress, and impair autophagy (Achour et al., 2016; Goldstein et al., 2016). Finally, EVOO has shown gut-modulating activity, by increasing the growth of beneficial bacteria and diminishing pathogenic bacteria (Millman et al., 2021).

The health-enhancing properties of olive oil are lost when its composition is altered by industrial practices focused on maximizing profits as is shown in Figure 3. Adulteration with other vegetable oils can result in a product of poor nutritional quality that oxidizes more readily and instead of offering health benefits can even be harmful. Consumption of rancid oil, due to the prooxidant substances present can induce oxidative stress and dysfunction of the vascular endothelium, which plays an essential role in the pathophysiology of several diseases (Carnevale et al., 2014; Casas et al., 2017; Nocella et al., 2018). Additionally, the sustained consumption of *trans* or saturated fats, which include many refined oils (Astrup et al., 2020; Liu & Lu, 2018), is also a health risk. When coupled with a sedentary lifestyle and other bad habits, it increases the likelihood of suffering arteriosclerosis, myocardial infarctions or embolisms (Carnevale et al., 2014; Casas et al., 2016, 2017).

During the refining process of olive oil (the deodorisation step), the content of 3-monochloropropane-1,2-diol (3-MCPD), 2-monochloropropane-1,3-diol esters (2-MCPDE) and GE was found to increase (Hung et al., 2017; Kamikata et al., 2019). As the processing of EVOO does not require the use of high temperatures, it is not expected to contain these substances at quantifiable levels. Therefore, 3-MCPDE, 2-MCPDE and GE have been proposed as complementary indicators of EVOO adulteration (Kamikata et al., 2019; Weesepoel, Alewijn, Wijtten, & Müller-Maatsch, 2021). The mixing of EVOO with refined oils can pose a threat to consumer health if it results in an increase in these compounds, GE and 3-MCPD are classified respectively as “probable human carcinogen” (category 2A) and “possible human carcinogen” (category 2B) by the

International Agency for Research on Cancer (IARC, 2000). The European Food Safety Authority (EFSA) CONTAM Panel established a tolerable daily intake of 0.8 µg/kg body weight per day for 3-MCPD (CONTAM, 2016).

The presence of polycyclic aromatic hydrocarbons, besides being related with EVOO fraud, is also a substance with potentially health risk. These genotoxic and carcinogenic compounds (EFSA, 2008; Cancer, 2006; World Health Organization, 2005) are formed during incomplete combustion or pyrolysis of organic matter, and their presence in oils is attributed to the drying process to which seeds, grains and olive pomace are subjected during processing (Tfouni et al., 2017).

Finally,. Arlorio *et al*. (2010) demonstrated that the adulteration of EVOO with hazelnut oil introduces a potential risk for consumers with hazelnut allergies, after analyzing the allergen and protein content of EVOO deliberately spiked with raw cold-pressed hazelnut oil or solvent-extracted hazelnut oil. Sodium dodecyl sulfate polyacrylamide gel electrophoresis (SDS–PAGE) analysis confirmed the presence of hazelnut proteins in solvent-extracted hazelnut oil, which were still detectable at a 1% contamination level in solvent-extracted hazelnut oil spiked EVOO, and therefore showing the potential health risk for sensitized people.

3 Traceability

Product traceability refers to the ability to monitor a product through all the stages of the production chain from its origin to the final destination. This process allows the product to be registered and identified, thereby guaranteeing its quality and protecting the consumer (Melucci et al., 2016). Due to the growing demand for high quality

EVOO, its characteristics and composition need to be specified, including the geographic origin and variety of the olives (Melucci et al., 2016; Violino et al., 2019).

Monovarietal olive oils are distinguished by a recognizable taste associated with a particular cultivar. In contrast, coupage olive oils are produced from a blend of olive varieties, with the main objective of obtaining an exotic and singular aroma and flavor (Campestre, Angelini, Gasbarri, & Angerosa, 2017).

The EU Regulation 2081/92, which was established to protect and promote quality food products, specifies that EVOO must be labeled according to its origin. Accordingly, oils can be registered within different schemes, as shown in **Table 3**: protected designation of origin (PDO), protected geographical indication (PGI) and traditional specialty guaranteed (TSG) (Violino et al., 2019).

To qualify for a PDO, the olive oil must comply with specific requirements regarding the geographic origin, cultivar, organoleptic characteristics, production methods and agronomic practice (Giménez, Pistón, Martín, & Atienza, 2010). Certification and denomination require EVOO traceability to be established, focusing on the region of the olive tree (geographic traceability) and the cultivar (botanical traceability).

3.1 Geographic and botanical traceability

Olive oil from different geographic sources can vary considerably in its components, reflecting variable factors such as the growth environment, climate, soil, and water quality. The Commission Implementing Regulation 2013 EU No. 1335/13 stipulates that the location of the olive harvest must be stated on EVOO labels (COMMUNITY, 2013a), as well as olive oil labels, for example “blend of olive oils (not) of EU origin” or “blend of olive oils of EU origin and not of EU origin” (Melucci et al., 2016).

Different strategies have been developed to characterize EVOO from several origins to detect fraudulent practices. Instead of analyzing one or a group of marker compounds, researchers have focused in analyzing whole spectra of techniques such as infrared or mass spectroscopy. These techniques usually require few or non pretreatments steps, however, chemometrics is needed to process all the data.

Although few studies on NIR and FTIR spectroscopies as a non-invasive techniques for food authentication have been published to date, the results indicate they are robust and reliable methods (Galtier et al., 2007; Garrido-Varo, Sánchez, De la Haba, Torres, & Pérez-Marín, 2017; Tapp, Defernez, & Kemsley, 2003). For example, NIR spectroscopy categorized 125 olive oils into five geographically close designations of origin in France (Galtier et al., 2007). This study demonstrated that the origin of olive oil can be traced by analyzing NIR spectra, which is related to the FA and TAG profiles, without the need for time-consuming physical and chemical procedures to analyze those compounds. In the same line, two models (spinning and static sample presentation) were developed to predict and classify olive oil quality parameters (Garrido-Varo et al., 2017). In another study, FTIR spectroscopy in combination with multivariate analysis was also carried out to distinguish EVOOs from different geographic origin (Tapp et al., 2003).

The traceability of the geographic origin of EVOOs by PCA and DA of the headspace volatile profile as a fingerprint has also been reported (Melucci et al., 2016). In this preliminary study, an FGC E-nose accurately determined the geographic origin of EVOO (100% Italian versus non-100% Italian), and the results indicate it is suitable for the rapid screening of commercial EVOO to verify if the information declared on the label is correct. Similarly, Quintanilla-Casas *et al.* (2020) analyzed EVOO samples from seven countries by headspace solid phase microextraction-GC-MS (HS-SPME-GC-MS) combined with chemometrics. Sesquiterpene hydrocarbons proved to be accurate geographic markers and fingerprinting was established as a more efficient approach than profiling to classify EVOO according to its origin. Further experiments of the same group showed that the sesquiterpene hydrocarbon fingerprints permitted the proper characterization of origin of EVOOs from and not from the European Union (Quintanilla-Casas et al., 2022).

Apart from geographical traceability, botanical traceability is important to assess EVOO trueness. Botanical traceability of EVOO is the ability to identify the olive variety, whose characteristics depend on the type of soil, growing conditions, and climate adaptability (Montealegre, Marina Alegre, & García-Ruiz, 2010). Although PDO verification is useful for EVOO classification and certification, its accomplishment is not always straightforward. Olive cultivars used to be distinguished by morphological and pomological traits, an approach limited by the influence of external uncontrolled factors (Sanz-Cortés et al., 2003). More efficient methods involve compositional analysis and genetic markers (Montealegre et al., 2010). The most relevant compositional markers for establishing the botanical traceability of olive oils are summarized in **Table 4**. As the chemical composition of EVOO is strongly affected by environmental conditions, fruit ripening, and the extraction technology, the botanical origin cannot be identified by a single compositional marker (ie. FA, phenolic compounds, volatile compounds, pigments, etc.), and instead several parameters are analyzed together using chemometric tools.

Aranda *et al.* 2004 classified 4 Spanish cultivars (*Cornicabra*, *Arbequina*, *Hojiblanca* and *Picual*) according to their distinct FA and triglyceride profiles and according to their distinct FA in the 2-position in the triglycerides profiles. For analyzing the FA in the 2-

position several sample preparation steps are needed, while the FA acid profile is quicker to obtain. The differences in the total FA profile permitted a successful discrimination with easier analysis methodology than the FA in the 2-position. Sterol composition together with chemometrics allowed the discrimination between 3 Portuguese cultivars (*Cobrançosa*, *Madural* and *Verdeal*) (Alves et al., 2005). Although sterol composition alone was not enough to distinguish EVOOs from *Manzanilla Cacereña* cultivar from EVOOs from other cultivars (Diaz, Merás, Casas, & Franco, 2005).

Different varieties will present different enzymatic activities, which result in different volatile and phenolic profiles, those have also been used to discriminate between varieties (Gómez-Alonso, Salvador, & Fregapane, 2002; Haddada et al., 2007). In addition, tocopherols, pigments and hydrocarbons are also affected by the olive enzymes, which depend on the cultivar, hence those markers have been used successfully as well as discriminants between different olive cultivars (Baccouri et al., 2007; Bueno, Casas, García, & González, 2005; Giuffrida et al., 2007).

In spite of all that, the environmental and technical factors have such a high impact on EVOO composition (Olmo-Cunillera et al., 2021) that the best approach for assessing EVOO botanical origin are the genetic markers. Although olive oils have a low protein content, peptide separation by capillary electrophoresis has proved to be an effective method for the differentiation of monovarietal olive oils. However, UV detection of the protein profiles, as well as protein isolation and solubilization, needs to be performed first (Monasterio, Fernández, & Silva, 2013; Montealegre et al., 2010). This approach has been further developed by coupling capillary electrophoresis with MS, which constitutes a reliable and rapid method to assess EVOO authenticity and quality (Monasterio et al., 2013; Sánchez-Hernández, Marina, & Crego, 2011).

4 Conclusions

In conclusion, analyzing the whole EVOO fingerprint seems to be the best approach to detect EVOO adulteration, however expensive equipment (HRMS) is needed. Then, both stigmastadienes and sterolic profile are proposed as markers for detecting EVOO blended with refined oils and solvent extracted oils.

Nowadays, international consumer demand for EVOO is growing, and fraudulent practices to maximize profits by reducing oil quality, are also on the rise. These deceitful practices not only reduce the quality of the oil and consequently its beneficial effects on health, but also may pose a significant risk to the ingestion of toxic, carcinogenic or allergic substances. In the fight against fraud, the EU, IOC, and Codex Alimentarius have established several standardized regulations that EVOO producers need to comply with. These endeavors are supported by scientific research focused on developing and improving strategies and novel analytical technologies that can identify possible adulterations, such as mixing EVOO with refined olive oil or other edible oils.

In addition, analytical methods such as FTIR or GC-MS can also authenticate the product by determining either the geographic or botanical origin of the oil, and tracing all the stages in the production chain.

Acknowledgements

This research was funded by CICYT [AGL2016- 75329-R]; it is part of the project [PID2020-114022RB-I00], funded by MCIN/ AEI/10.13039/501100011033; CIBEROBN from the Instituto de Salud Carlos III, ISCIII from the Ministerio de Ciencia, Innovación y Universidades, (AEI/FEDER, UE) and Generalitat de Catalunya [2017SGR 196]. Julián Lozano-Castellón and Inés Domínguez-López thank the Ministry of Science Innovation and Universities for the FPI and FPU contracts, respectively [BES-2017-080017] and [FPU20/02478]. Anna Vallverdú-Queralt thanks the Ministry of Science Innovation and Universities for the Ramon y Cajal contract [RYC-2016-19355].

Conflicts of Interest

Rosa María Lamuela-Raventós declare to receive lecture fees from Cerveceros de España and lecture fees and travel support from Adventia. The other authors declare no conflict of interest.

5 References

(CONTAM), E. P. on C. in the F. C. (2016). Risks for human health related to the presence of 3-and 2-monochloropropanediol (MCPD), and their fatty acid esters, and glycidyl fatty acid esters in food. *Efsa Journal*, *14*(5), e04426.

(EFSA), E. F. S. A. (2008). Polycyclic Aromatic Hydrocarbons in Food-Scientific Opinion of the Panel on Contaminants in the Food Chain. *EFSA Journal*, *6*(8), 724.

Achour, I., Arel-Dubeau, A.-M., Renaud, J., Legrand, M., Attard, E., Germain, M., & Martinoli, M.-G. (2016). Oleuropein Prevents Neuronal Death, Mitigates Mitochondrial Superoxide Production and Modulates Autophagy in a Dopaminergic Cellular Model. *International Journal of Molecular Sciences*, *17*(8), 1293. https://doi.org/10.3390/ijms17081293

Al-Ismail, K. M., Alsaed, A. K., Ahmad, R., & Al-Dabbas, M. (2010). Detection of olive oil adulteration with some plant oils by GLC analysis of sterols using polar column. *Food Chemistry*, *121*(4), 1255–1259.

https://doi.org/10.1016/j.foodchem.2010.01.016

Alessandri, S., Ieri, F., & Romani, A. (2014). Minor Polar Compounds in Extra Virgin Olive Oil: Correlation between HPLC-DAD-MS and the Folin-Ciocalteu Spectrophotometric Method. *Journal of Agricultural and Food Chemistry*, *62*(4), 826–835. https://doi.org/10.1021/jf403104a

Ali, H., Saleem, M., Anser, M. R., Khan, S., Ullah, R., & Bilal, M. (2018). Validation of Fluorescence Spectroscopy to Detect Adulteration of Edible Oil in Extra Virgin Olive Oil (EVOO) by Applying Chemometrics. *Applied Spectroscopy*, *72*(9), 1371–1379. https://doi.org/10.1177/0003702818768485

Alves, A. Q., da Silva, V. A., Góes, A. J. S., Silva, M. S., de Oliveira, G. G., Bastos, I. V. G. A., … Alves, A. J. (2019). The Fatty Acid Composition of Vegetable Oils and Their Potential Use in Wound Care. *Advances in Skin & Wound Care*, *32*(8), 1–8. https://doi.org/10.1097/01.ASW.0000557832.86268.64

Alves, E., Domingues, M., & Domingues, P. (2018). Polar Lipids from Olives and Olive Oil: A Review on Their Identification, Significance and Potential Biotechnological Applications. *Foods*, *7*(7), 109. https://doi.org/10.3390/foods7070109

Alves, M. R., Cunha, S. C., Amaral, J. S., Pereira, J. A., & Oliveira, M. B. (2005). Classification of PDO olive oils on the basis of their sterol composition by multivariate analysis. *Analytica Chimica Acta*, *549*(1–2), 166–178. https://doi.org/10.1016/j.aca.2005.06.033

Antonelli, M., Benedetti, B., Cavaliere, C., Cerrato, A., Montone, C. M., Piovesana, S., … Capriotti, A. L. (2020). Phospholipidome of extra virgin olive oil: Development of a solid phase extraction protocol followed by liquid chromatography–high resolution mass spectrometry for its software-assisted identification. *Food*

Chemistry, *310*, 125860.

Apetrei, I. M., & Apetrei, C. (2014). Detection of virgin olive oil adulteration using a voltammetric e-tongue. *Computers and Electronics in Agriculture*, *108*, 148–154.

Aranda, F., Gómez-Alonso, S., Rivera Del Álamo, R. M., Salvador, M. D., & Fregapane, G. (2004). Triglyceride, total and 2-position fatty acid composition of Cornicabra virgin olive oil: Comparison with other Spanish cultivars. *Food Chemistry*, *86*(4), 485–492. https://doi.org/10.1016/j.foodchem.2003.09.021

Arlorio, M., Coisson, J. D., Bordiga, M., Travaglia, F., Garino, C., Zuidmeer, L., … Martelli, A. (2010). Olive oil adulterated with hazelnut oils: simulation to identify possible risks to allergic consumers. *Food Additives and Contaminants*, *27*(1), 11–18.

Aroca-Santos, R., Cancilla, J. C., Matute, G., & Torrecilla, J. S. (2015). Identifying and quantifying adulterants in extra virgin olive oil of the picual varietal by absorption spectroscopy and nonlinear modeling. *Journal of Agricultural and Food Chemistry*, *63*(23), 5646–5652.

Aroca-Santos, R., Cancilla, J. C., Pariente, E. S., & Torrecilla, J. S. (2016). Neural networks applied to characterize blends containing refined and extra virgin olive oils. *Talanta*, *161*, 304–308.

Astrup, A., Magkos, F., Bier, D. M., Brenna, J. T., de Oliveira Otto, M. C., Hill, J. O., … Krauss, R. M. (2020). Saturated Fats and Health: A Reassessment and Proposal for Food-Based Recommendations. *Journal of the American College of Cardiology*, *76*(7), 844–857. https://doi.org/10.1016/j.jacc.2020.05.077

Aykas, D. P., Karaman, A. D., Keser, B., & Rodriguez-Saona, L. (2020). Non-targeted authentication approach for extra virgin olive oil. *Foods*, *9*(2), 1–17.

https://doi.org/10.3390/foods9020221

Azizian, H., Mossoba, M. M., Fardin-Kia, A. R., Karunathilaka, S. R., & Kramer, J. K. G. (2016). Developing FT-NIR and PLS1 Methodology for Predicting Adulteration in Representative Varieties/Blends of Extra Virgin Olive Oils. *Lipids*, *51*(11), 1309–1321. https://doi.org/10.1007/s11745-016-4195-0

Azizian, H., Mossoba, M. M., Fardin-Kia, A. R., Delmonte, P., Karunathilaka, S. R., & Kramer, J. K. G. (2015). Novel, rapid identification, and quantification of adulterants in extra virgin olive oil using near-infrared spectroscopy and chemometrics. *Lipids*, *50*(7), 705–718.

Azizian, H., Wang, S. C., Li, X., & Kramer, J. K. G. (2018). Improvement of the Fourier Transform Near Infrared Method to Evaluate Extra Virgin Olive Oils by Analyzing 1,2-Diacylglycerols and 1,3-Diacylglycerols and Adding Unesterified Fatty Acids. *Lipids*, *53*(11–12), 1097–1112. https://doi.org/10.1002/lipd.12113

Baccouri, O., Cerretani, L., Bendini, A., Caboni, M. F., Zarrouk, M., Pirrone, L., & Miled, D. D. Ben. (2007). Preliminary chemical characterization of Tunisian monovarietal virgin olive oils and comparison with Sicilian ones. *European Journal of Lipid Science and Technology*, *109*(12), 1208–1217. https://doi.org/10.1002/ejlt.200700132

Bajoub, A., Bendini, A., Fernández-Gutiérrez, A., & Carrasco-Pancorbo, A. (2018). Olive oil authentication: A comparative analysis of regulatory frameworks with especial emphasis on quality and authenticity indices, and recent analytical techniques developed for their assessment. A review. *Critical Reviews in Food Science and Nutrition*, *58*(5), 832–857. https://doi.org/10.1080/10408398.2016.1225666

Bansal, S., Singh, A., Mangal, M., Mangal, A. K., & Kumar, S. (2017). Food adulteration:

Sources, health risks, and detection methods. *Critical Reviews in Food Science and Nutrition*, *57*(6), 1174–1189.

Barbieri, S., Cevoli, C., Bendini, A., Quintanilla-Casas, B., García-González, D. L., & Gallina Toschi, T. (2020). Flash Gas Chromatography in Tandem with Chemometrics: A Rapid Screening Tool for Quality Grades of Virgin Olive Oils. *Foods*, *9*(7), 862.

Batarseh, Y. S., & Kaddoumi, A. (2018). Oleocanthal-rich extra-virgin olive oil enhances donepezil effect by reducing amyloid-β load and related toxicity in a mouse model of Alzheimer's disease. *The Journal of Nutritional Biochemistry*, *55*, 113–123. https://doi.org/10.1016/j.jnutbio.2017.12.006

Beltrán, G., Jiménez, A., del Rio, C., Sánchez, S., Martínez, L., Uceda, M., & Aguilera, M. P. (2010). Variability of vitamin E in virgin olive oil by agronomical and genetic factors. *Journal of Food Composition and Analysis*, *23*(6), 633–639.

Boneza, M. M., & Niemeyer, E. D. (2018). Cultivar affects the phenolic composition and antioxidant properties of commercially available lemon balm (Melissa officinalis L.) varieties. *Industrial Crops and Products*, *112*, 783–789. https://doi.org/10.1016/j.indcrop.2018.01.003

Bueno, E. O., Casas, J. S., García, A. M., & González, L. G. (2005). Discriminating power of the hydrocarbon content from virgin olive oil of Extremadura cultivars. *JAOCS, Journal of the American Oil Chemists' Society*, *82*(1), 1–6. https://doi.org/10.1007/s11746-005-1034-0

Bullo, M., Lamuela-Raventos, R., & Salas-Salvado, J. (2011). Mediterranean Diet and Oxidation: Nuts and Olive Oil as Important Sources of Fat and Antioxidants. *Current Topics in Medicinal Chemistry*, *11*(14), 1797–1810.

https://doi.org/10.2174/156802611796235062

Calvano, C. D., De Ceglie, C., D’Accolti, L., & Zambonin, C. G. (2012). MALDI-TOF mass spectrometry detection of extra-virgin olive oil adulteration with hazelnut oil by analysis of phospholipids using an ionic liquid as matrix and extraction solvent. *Food Chemistry*, *134*(2), 1192–1198.

Campestre, C., Angelini, G., Gasbarri, C., & Angerosa, F. (2017). The compounds responsible for the sensory profile in monovarietal virgin olive oils. *Molecules*, *22*(11), 1833.

Campmajó, G., Saurina, J., & Núñez, O. (2022). FIA–HRMS fingerprinting subjected to chemometrics as a valuable tool to address food classification and authentication: Application to red wine, paprika, and vegetable oil samples. *Food Chemistry*, *373*, 131491. https://doi.org/10.1016/j.foodchem.2021.131491

Cancer, I. A. for R. on. (2006). Monographs on the evaluation of carcinogenic risks to humans. *Http://Monographs. Iarc. Fr/ENG/Classification/Index. Php*.

Cárdeno, A., Magnusson, M. K., Strid, H., de La Lastra, C. A., Sánchez-Hidalgo, M., & Öhman, L. (2014). The unsaponifiable fraction of extra virgin olive oil promotes apoptosis and attenuates activation and homing properties of T cells from patients with inflammatory bowel disease. *Food Chemistry*, *161*, 353–360.

Carnevale, R., Pignatelli, P., Nocella, C., Loffredo, L., Pastori, D., Vicario, T., … Violi, F. (2014). Extra virgin olive oil blunt post-prandial oxidative stress via NOX2 down-regulation. *Atherosclerosis*, *235*(2), 649–658.

Carranco, N., Farrés-Cebrián, M., Saurina, J., & Núñez, O. (2018). Authentication and quantitation of fraud in extra virgin olive oils based on HPLC-UV fingerprinting and multivariate calibration. *Foods*, *7*(4), 44. https://doi.org/10.3390/foods7040044

Casas, R., Sacanella, E., Urpí-Sardà, M., Corella, D., Castaner, O., Lamuela-Raventos, R.-M., … Estruch, R. (2016). Long-term immunomodulatory effects of a mediterranean diet in adults at high risk of cardiovascular disease in the PREvención con DIeta MEDiterránea (PREDIMED) randomized controlled trial. *The Journal of Nutrition*, *146*(9), 1684–1693.

Casas, R., Urpi-Sardà, M., Sacanella, E., Arranz, S., Corella, D., Castañer, O., … Estruch, R. (2017). Anti-Inflammatory Effects of the Mediterranean Diet in the Early and Late Stages of Atheroma Plaque Development. *Mediators of Inflammation*, *2017*, 1–13. https://doi.org/10.1155/2017/3674390

Castillo-Luna, A., Criado-Navarro, I., Ledesma-Escobar, C. A., López-Bascón, M. A., & Priego-Capote, F. (2021). The decrease in the health benefits of extra virgin olive oil during storage is conditioned by the initial phenolic profile. *Food Chemistry*, *336*, 127730. https://doi.org/10.1016/j.foodchem.2020.127730

Castro-Barquero, S., Tresserra-Rimbau, A., Vitelli-Storelli, F., Doménech, M., Salas-Salvadó, J., Martín-Sánchez, V., … Fitó, M. (2020). Dietary polyphenol intake is associated with HDL-cholesterol and a better profile of other components of the metabolic syndrome: A PREDIMED-plus sub-study. *Nutrients*, *12*(3), 689.

Cecchi, L., Migliorini, M., & Mulinacci, N. (2021). Virgin olive oil volatile compounds: composition, sensory characteristics, analytical approaches, quality control, and authentication. *Journal of Agricultural and Food Chemistry*, *69*(7), 2013–2040.

Chen, C.-G., Wang, P., Zhang, Z.-Q., Ye, Y.-B., Zhuo, S.-Y., Zhou, Q., … Zhang, B. (2020). Effects of plant oils with different fatty acid composition on cardiovascular risk factors in moderately hypercholesteremic Chinese adults: a randomized, double-blinded, parallel-designed trial. *Food & Function*, *11*(8), 7164–7174.

https://doi.org/10.1039/D0FO00875C

Chen, H., Angiuli, M., Ferrari, C., Tombari, E., Salvetti, G., & Bramanti, E. (2011). Tocopherol speciation as first screening for the assessment of extra virgin olive oil quality by reversed-phase high-performance liquid chromatography/fluorescence detector. *Food Chemistry*, *125*(4), 1423–1429.

Chiavaro, E., Vittadini, E., Rodriguez-Estrada, M. T., Cerretani, L., & Bendini, A. (2008). Differential scanning calorimeter application to the detectionof refined hazelnut oil in extra virgin olive oil. *Food Chemistry*, *110*(1), 248–256.

Chiou, A., Salta, F. N., Kalogeropoulos, N., Mylona, A., Ntalla, I., & Andrikopoulos, N. K. (2007). Retention and Distribution of Polyphenols after Pan-Frying of French Fries in Oils Enriched with Olive Leaf Extract. *Journal of Food Science*, *72*(8), S574–S584. https://doi.org/10.1111/j.1750-3841.2007.00493.x

Clodoveo, M. L., Hbaieb, R. H., Kotti, F., Mugnozza, G. S., & Gargouri, M. (2014). Mechanical strategies to increase nutritional and sensory quality of virgin olive oil by modulating the endogenous enzyme activities. *Comprehensive Reviews in Food Science and Food Safety*, *13*(2), 135–154.

COMMUNITY, E. (1991). European Commission Regulation 2568/91 on the characteristics of olive oil and olive residue oil and on the relevant methods of analysis, and subsequent amendments. Official Journal of European Community.

COMMUNITY, E. (2006). COUNCIL REGULATION (EC) No 510/2006 of 20 March 2006 on the protection of geographical indications and designations of origin for agricultural products and foodstuffs. Official Journal of the European Union. Retrieved from https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32006R0510&from=EN

COMMUNITY, E. (2013a). COMMISSION IMPLEMENTING REGULATION (EU) No 1335/2013 of 13 December 2013 amending Implementing Regulation (EU) No 29/2012 on marketing standards for olive oil. Official Journal of the European Union. Retrieved from https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32013R1335&from=ES

COMMUNITY, E. (2013b). COMMISSION IMPLEMENTING REGULATION (EU) No 1348/2013 of 16 December 2013 amending Regulation (EEC) No 2568/91 on the characteristics of olive oil and olive-residue oil and on the relevant methods of analysis. Official Journal of the European Union. Retrieved from https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32013R1348&from=ES

Conte, L., Bendini, A., Valli, E., Lucci, P., Moret, S., Maquet, A., … Gallina Toschi, T. (2020). Olive oil quality and authenticity: A review of current EU legislation, standards, relevant methods of analyses, their drawbacks and recommendations for the future. *Trends in Food Science & Technology*, *105*, 483–493. https://doi.org/10.1016/j.tifs.2019.02.025

Correa Fuentes, J. D. P., Martín Gordo, D., Pinzón Reyes, S. P., & Cárdenas González, O. E. (2017). Efecto del estado de maduración sobre el contenido de polifenoles totales en frutos de Solanum marginatum. *I3+*, *3*(2), 86–97. https://doi.org/10.24267/23462329.220

Covas, M.-I., Nyyssönen, K., Poulsen, H. E., Kaikkonen, J., Zunft, H.-J. F., Kiesewetter, H., … Marrugat, J. (2006). The Effect of Polyphenols in Olive Oil on Heart Disease Risk Factors. *Annals of Internal Medicine*, *145*(5), 333. https://doi.org/10.7326/0003-4819-145-5-200609050-00006

Crews, C., Pye, C., & Macarthur, R. (2014). An improved rapid stigmastadiene test to detect addition of refined oil to extra virgin olive oil. *Food Research International*, *60*, 117–122. https://doi.org/10.1016/j.foodres.2013.11.023

Creydt, M., & Fischer, M. (2018). Omics approaches for food authentication. *Electrophoresis*, *39*(13), 1569–1581. https://doi.org/10.1002/elps.201800004

Criado-Navarro, I., Ledesma-Escobar, C. A., Olmo-Peinado, J. M., Parrado-Martínez, M. J., Vílchez-García, P. J., Espejo-Calvo, J. A., & Priego-Capote, F. (2021). Influence of fruit destoning on bioactive compounds of virgin olive oil. *LWT*, *145*, 111354. https://doi.org/10.1016/j.lwt.2021.111354

da Silveira, R., Vágula, J. M., de Lima Figueiredo, I., Claus, T., Galuch, M. B., Junior, O. O. S., & Visentainer, J. V. (2017). Rapid methodology via mass spectrometry to quantify addition of soybean oil in extra virgin olive oil: a comparison with traditional methods adopted by food industry to identify fraud. *Food Research International*, *102*, 43–50.

Damiani, T., Cavanna, D., Serani, A., Dall’Asta, C., & Suman, M. (2020). GC-IMS and FGC-Enose fingerprint as screening tools for revealing extra virgin olive oil blending with soft-refined olive oils: A feasibility study. *Microchemical Journal*, *159*, 105374.

De Santis, S., Cariello, M., Piccinin, E., Sabbà, C., & Moschetta, A. (2019). Extra virgin olive oil: lesson from nutrigenomics. *Nutrients*, *11*(9), 2085.

Di Girolamo, F., Masotti, A., Lante, I., Scapaticci, M., Calvano, C. D., Zambonin, C., … Putignani, L. (2015). A simple and effective mass spectrometric approach to identify the adulteration of the mediterranean diet component extra-virgin olive oil with corn oil. *International Journal of Molecular Sciences*, *16*(9), 20896–20912.

Diaz, T. G., Merás, I. D., Casas, J. S., & Franco, M. F. A. (2005). Characterization of virgin olive oils according to its triglycerides and sterols composition by chemometric methods. *Food Control*, *16*(4), 339–347. https://doi.org/10.1016/j.foodcont.2004.03.014

Drira, M., Guclu, G., Portolés, T., Jabeur, H., Kelebek, H., Selli, S., & Bouaziz, M. (2021). Safe and Fast Fingerprint Aroma Detection in Adulterated Extra Virgin Olive Oil Using Gas Chromatography–Olfactometry-Mass Spectrometry Combined with Chemometrics. *Food Analytical Methods*, 1–15.

Drira, M., Kelebek, H., Guclu, G., Jabeur, H., Selli, S., & Bouaziz, M. (2020). Targeted analysis for detection the adulteration in extra virgin olive oil's using LC-DAD/ESI–MS/MS and combined with chemometrics tools. *European Food Research and Technology*, *246*(8), 1661–1677. https://doi.org/10.1007/s00217-020-03522-y

Ergönül, P. G., & Köseoğlu, O. (2014). Changes in α-, β-, γ-and δ-tocopherol contents of mostly consumed vegetable oils during refining process. *CyTA-Journal of Food*, *12*(2), 199–202.

Estruch, R., Ros, E., Salas-Salvadó, J., Covas, M.-I., Corella, D., Arós, F., … Martínez-González, M. A. (2018). Primary Prevention of Cardiovascular Disease with a Mediterranean Diet Supplemented with Extra-Virgin Olive Oil or Nuts. *New England Journal of Medicine*, *378*(25), e34. https://doi.org/10.1056/NEJMoa1800389

Fasciotti, M., & Netto, A. D. P. (2010). Optimization and application of methods of triacylglycerol evaluation for characterization of olive oil adulteration by soybean oil with HPLC–APCI-MS–MS. *Talanta*, *81*(3), 1116–1125.

Feizabadi, A., Noormohammadi, G., & Fatehi, F. (2021). Changes in Growth, Physiology, and Fatty Acid Profile of Rapeseed Cultivars Treated with Vermicompost Under

Drought Stress. *Journal of Soil Science and Plant Nutrition*, *21*(1), 200–208. https://doi.org/10.1007/s42729-020-00353-4

Ferreiro-González, M., Barbero, G. F., Álvarez, J. A., Ruiz, A., Palma, M., & Ayuso, J. (2017). Authentication of virgin olive oil by a novel curve resolution approach combined with visible spectroscopy. *Food Chemistry*, *220*, 331–336.

Fiorini, D., Boarelli, M. C., Conti, P., Alfei, B., Caprioli, G., Ricciutelli, M., … Pacetti, D. (2018). Chemical and sensory differences between high price and low price extra virgin olive oils. *Food Research International*, *105*(November 2017), 65–75. https://doi.org/10.1016/j.foodres.2017.11.005

Flori, L., Donnini, S., Calderone, V., Zinnai, A., Taglieri, I., Venturi, F., & Testai, L. (2019). The nutraceutical value of olive oil and its bioactive constituents on the cardiovascular system. focusing on main strategies to slow down its quality decay during production and storage. *Nutrients*, *11*(9), 1962.

Fragaki, G., Spyros, A., Siragakis, G., Salivaras, E., & Dais, P. (2005). Detection of extra virgin olive oil adulteration with lampante olive oil and refined olive oil using nuclear magnetic resonance spectroscopy and multivariate statistical analysis. *Journal of Agricultural and Food Chemistry*, *53*(8), 2810–2816.

Frankel, E. N. (2014). *Lipid oxidation* (2nd ed.). Elsevier.

Gallina Toschi, T., Bendini, A., Lozano-Sánchez, J., Segura-Carretero, A., & Conte, L. (2013). Misdescription of edible oils: Flowcharts of analytical choices in a forensic view. *European Journal of Lipid Science and Technology*, *115*(11), 1205–1223.

Galtier, O., Dupuy, N., Le Dréau, Y., Ollivier, D., Pinatel, C., Kister, J., & Artaud, J. (2007). Geographic origins and compositions of virgin olive oils determinated by chemometric analysis of NIR spectra. *Analytica Chimica Acta*, *595*(1–2), 136–144.

García-Gavilán, J. F., Bulló, M., Canudas, S., Martínez-González, M. A., Estruch, R., Giardina, S., … Salas-Salvadó, J. (2018). Extra virgin olive oil consumption reduces the risk of osteoporotic fractures in the PREDIMED trial. *Clinical Nutrition*, *37*(1), 329–335. https://doi.org/10.1016/j.clnu.2016.12.030

García Martínez, E. M. (2020). Detección de fraudes en aceites vegetales mediante identificación y cuantificación de ácidos grasos.

Garcia, R., Martins, N., & Cabrita, M. J. (2013). Putative markers of adulteration of extra virgin olive oil with refined olive oil: Prospects and limitations. *Food Research International*, *54*(2), 2039–2044.

Gargouri, B., Zribi, A., & Bouaziz, M. (2015). Effect of containers on the quality of Chemlali olive oil during storage. *Journal of Food Science and Technology*, *52*(4), 1948–1959. https://doi.org/10.1007/s13197-014-1273-2

Garrido-Varo, A., Sánchez, M.-T., De la Haba, M.-J., Torres, I., & Pérez-Marín, D. (2017). Fast, low-cost and non-destructive physico-chemical analysis of virgin olive oils using near-infrared reflectance spectroscopy. *Sensors*, *17*(11), 2642.

Gelmini, F., Ruscica, M., MacChi, C., Bianchi, V., Maffei Facino, R., Beretta, G., & Magni, P. (2016). Unsaponifiable Fraction of Unripe Fruits of Olea europaea: An Interesting Source of Anti-inflammatory Constituents. *Planta Medica*, *82*(3), 273–278. https://doi.org/10.1055/s-0035-1558155

Giménez, M. J., Pistón, F., Martín, A., & Atienza, S. G. (2010). Application of real-time PCR on the development of molecular markers and to evaluate critical aspects for olive oil authentication. *Food Chemistry*, *118*(2), 482–487.

Giuffrida, D., Salvo, F., Salvo, A., La Pera, L., & Dugo, G. (2007). Pigments composition in monovarietal virgin olive oils from various sicilian olive varieties. *Food*

Chemistry, *101*(2), 833–837.

Goldstein, D. S., Jinsmaa, Y., Sullivan, P., Holmes, C., Kopin, I. J., & Sharabi, Y. (2016). 3,4-Dihydroxyphenylethanol (Hydroxytyrosol) Mitigates the Increase in Spontaneous Oxidation of Dopamine During Monoamine Oxidase Inhibition in PC12 Cells. *Neurochemical Research*, *41*(9), 2173–2178. https://doi.org/10.1007/s11064-016-1959-0

Gómez-Alonso, S., Salvador, M. D., & Fregapane, G. (2002). Phenolic compounds profile of Cornicabra virgin olive oil. *Journal of Agricultural and Food Chemistry*, *50*(23), 6812–6817. https://doi.org/10.1021/jf0205211

Gómez-Coca, R. B., Pérez-Camino, M. de C., Bendini, A., Gallina Toschi, T., & Moreda, W. (2020). Olive oil mixtures. Part two: Detection of soft deodorized oil in extra virgin olive oil through diacylglycerol determination. Relationship with free acidity. *Food Chemistry*, *330*, 127226. https://doi.org/10.1016/j.foodchem.2020.127226

González-Domínguez, R., Sayago, A., Morales, M. T., & Fernández-Recamales, Á. (2019). Assessment of virgin olive oil adulteration by a rapid luminescent method. *Foods*, *8*(8), 287.

Gorassini, A., Verardo, G., & Bortolomeazzi, R. (2019). Polymeric reversed phase and small particle size silica gel solid phase extractions for rapid analysis of sterols and triterpene dialcohols in olive oils by GC-FID. *Food Chemistry*, *283*, 177–182. https://doi.org/10.1016/j.foodchem.2018.12.120

Gordon, M. H., & Firman, C. (2001). Effects of heating and bleaching on formation of stigmastadienes in olive oil. *Journal of the Science of Food and Agriculture*, *81*(15), 1530–1532. https://doi.org/10.1002/jsfa.986

Guasch-Ferré, M., Hu, F. B., Martínez-González, M. A., Fitó, M., Bulló, M., Estruch, R., … Salas-Salvadó, J. (2014). Olive oil intake and risk of cardiovascular disease and mortality in the PREDIMED Study. *BMC Medicine*, *12*(1), 78. https://doi.org/10.1186/1741-7015-12-78

Guimet, F., Ferré, J., & Boqué, R. (2005). Rapid detection of olive–pomace oil adulteration in extra virgin olive oils from the protected denomination of origin “Siurana” using excitation–emission fluorescence spectroscopy and three-way methods of analysis. *Analytica Chimica Acta*, *544*(1–2), 143–152.

Haddada, F. M., Manai, H., Daoud, D., Fernandez, X., Lizzani-Cuvelier, L., & Zarrouk, M. (2007). Profiles of volatile compounds from some monovarietal Tunisian virgin olive oils. Comparison with French PDO. *Food Chemistry*, *103*(2), 467–476. https://doi.org/10.1016/j.foodchem.2006.08.023

Hayakawa, T., Yanagawa, M., Yamamoto, A., Aizawa, S., Taga, A., Mochizuki, N., … Kodama, S. (2020). A Simple Screening Method for Extra Virgin Olive Oil Adulteration by Determining Squalene and Tyrosol. *Journal of Oleo Science*, *69*(7), 677–684. https://doi.org/10.5650/jos.ess20033

Holey, S. A., Sekhar, K. P. C., Mishra, S. S., Kanjilal, S., & Nayak, R. R. (2021). Effect of oil unsaturation and wax composition on stability, properties and food applicability of oleogels. *Journal of the American Oil Chemists’ Society*. https://doi.org/10.1002/aocs.12536

Hrncirik, K., & Fritsche, S. (2005). Relation between the endogenous antioxidant system and the quality of extra virgin olive oil under accelerated storage conditions. *Journal of Agricultural and Food Chemistry*, *53*(6), 2103–2110.

Hung, W.-C., Peng, G.-J., Tsai, W.-J., Chang, M.-H., Liao, C.-D., Tseng, S.-H., … Cheng, H.-

F. (2017). Identification of 3-MCPD esters to verify the adulteration of extra virgin olive oil. *Food Additives & Contaminants: Part B*, *10*(3), 233–239.

IARC Working Group on the Evaluation of Carcinogenic Risks to Humans. (2000). IARC monographs on the evaluation of carcinogenic risk of chemicals to humans, 469–486.

International Olive Council. (2017a). Method for the Determination of Stigmastadienes in Vegetable Oils, *20*(11), 1–12. Retrieved from https://www.internationaloliveoil.org/wp-content/uploads/2019/11/Method-COI-T.20-Doc.-No-11-Rev.-3-2017-.pdf

International Olive Council. (2017b). Method of Analysis Determination of Biophenols in Olive Oils By Hplc, *20*(29), 1–8. Retrieved from http://www.internationaloliveoil.org/

International Olive Council. (2021). Trade standard applying to olive oils and olive pomace oils. Madrid. Retrieved from https://www.internationaloliveoil.org/wp-content/uploads/2021/07/COI-T15-NC3-REV-16-2021-_ENG.pdf

Jabeur, H., Zribi, A., Makni, J., Rebai, A., Abdelhedi, R., & Bouaziz, M. (2014). Detection of Chemlali extra-virgin olive oil adulteration mixed with soybean oil, corn oil, and sunflower oil by using GC and HPLC. *Journal of Agricultural and Food Chemistry*, *62*(21), 4893–4904.

Jensen, S. (2002). Chemotaxonomy of the Oleaceae: iridoids as taxonomic markers. *Phytochemistry*, *60*(3), 213–231. https://doi.org/10.1016/S0031-9422(02)00102-4

Jimenez-Lopez, C., Carpena, M., Lourenço-Lopes, C., Gallardo-Gomez, M., Lorenzo, J. M., Barba, F. J., … Simal-Gandara, J. (2020). Bioactive compounds and quality of extra virgin olive oil. *Foods*, *9*(8), 1014.

Kamal-Eldin, A. (2006). Effect of fatty acids and tocopherols on the oxidative stability of vegetable oils. *European Journal of Lipid Science and Technology*, *108*(12), 1051–1061. https://doi.org/10.1002/ejlt.200600090

Kamikata, K., Vicente, E., Arisseto-Bragotto, A. P., de Oliveira Miguel, A. M. R., Milani, R. F., & Tfouni, S. A. V. (2019). Occurrence of 3-MCPD, 2-MCPD and glycidyl esters in extra virgin olive oils, olive oils and oil blends and correlation with identity and quality parameters. *Food Control*, *95*, 135–141.

Kesen, S. (2019). Monitoring Fatty Acid and Sterol Profile of Nizip Yaglik Olive Oil Adulterated by Cotton and Sunflower Oil. *Journal of Oleo Science*, *68*(9), 817–826. https://doi.org/10.5650/jos.ess19130

Konuskan, D. B., Arslan, M., & Oksuz, A. (2019). Physicochemical properties of cold pressed sunflower, peanut, rapeseed, mustard and olive oils grown in the Eastern Mediterranean region. *Saudi Journal of Biological Sciences*, *26*(2), 340–344. https://doi.org/10.1016/j.sjbs.2018.04.005

Król, K., Gantner, M., & Piotrowska, A. (2021). The Quality Characteristic and Fatty Acid Profile of Cold-Pressed Hazelnut Oils during Nine Months of Storage. *Agronomy*, *11*(10), 2045. https://doi.org/10.3390/agronomy11102045

Lazzerini, C., Cifelli, M., & Domenici, V. (2017). Pigments in extra virgin olive oils produced in different mediterranean countries in 2014: Near UV-vis spectroscopy versus HPLC-DAD. *LWT*, *84*, 586–594.

Lecerf, J.-M. (2009). Fatty acids and cardiovascular disease. *Nutrition Reviews*, *67*(5), 273–283. https://doi.org/10.1111/j.1753-4887.2009.00194.x

Li, W., Sperry, J. B., Crowe, A., Trojanowski, J. Q., Smith III, A. B., & Lee, V. M.-Y. (2009). Inhibition of tau fibrillization by oleocanthal via reaction with the amino groups of tau. *Journal of Neurochemistry*, *110*(4), 1339–1351. https://doi.org/10.1111/j.1471-4159.2009.06224.x

Li, Y., Fang, T., Zhu, S., Huang, F., Chen, Z., & Wang, Y. (2018). Detection of olive oil adulteration with waste cooking oil via Raman spectroscopy combined with iPLS and SiPLS. *Spectrochimica Acta - Part A: Molecular and Biomolecular Spectroscopy*, *189*, 37–43. https://doi.org/10.1016/j.saa.2017.06.049

Lia, F., Castellano, A. M., Zammit-Mangion, M., & Farrugia, C. (2018). Application of fluorescence spectroscopy and chemometric models for the detection of vegetable oil adulterants in Maltese virgin olive oils. *Journal of Food Science and Technology*, *55*(6), 2143–2151.

Liu, W., & Lu, G. H. (2018). Cis-trans isomerization of unsaturated fatty acids in edible oils to prepare trans fat. *Grasas y Aceites*, *69*(3), 268. https://doi.org/10.3989/gya.0225181

López-Yerena, A., Lozano-Castellón, J., Olmo-Cunillera, A., Tresserra-Rimbau, A., Quifer-Rada, P., Jiménez, B., … Vallverdú-Queralt, A. (2019). Effects of organic and conventional growing systems on the phenolic profile of extra-virgin olive oil. *Molecules*, *24*(10). https://doi.org/10.3390/molecules24101986_rfseq1

López-Yerena, A., Ninot, A., Jiménez-Ruiz, N., Lozano-Castellón, J., Pérez, M., Escribano-Ferrer, E., … Vallverdú-Queralt, A. (2021). Influence of the Ripening Stage and Extraction Conditions on the Phenolic Fingerprint of ‘Corbella’Extra-Virgin Olive Oil. *Antioxidants*, *10*(6), 877.

López-Yerena, A., Ninot, A., Lozano-Castellón, J., Escribano-Ferrer, E., Romero-Aroca, A. J., Belaj, A., … Lamuela-Raventós, R. M. (2020). Conservation of native wild ivory-white olives from the Medes islands natural reserve to maintain virgin olive oil diversity. *Antioxidants*, *9*(10), 1009. https://doi.org/10.3390/antiox9101009

Lozano-Castellón, J., López-Yerena, A., Olmo-Cunillera, A., Jáuregui, O., Pérez, M., Lamuela-Raventós, R. M., & Vallverdú-Queralt, A. (2021). Total analysis of the major secoiridoids in extra virgin olive oil: Validation of an uhplc-esi-ms/ms method. *Antioxidants*, *10*(4), 1–13. https://doi.org/10.3390/antiox10040540

Lozano-Castellón, J., Vallverdú-Queralt, A., Rinaldi de Alvarenga, J. F., Illán, M., Torrado-Prat, X., & Lamuela-Raventós, R. M. (2020). Domestic Sautéing with EVOO: Change in the Phenolic Profile. *Antioxidants*, *9*(1), 77. https://doi.org/10.3390/antiox9010077

Manai-Djebali, H., Oueslati, I., Baccouri, B., Harzalli, Z., Martínez-Cañas, M. A., & Sánchez-Casas, J. (2021). Importance of Phytosterols in the Classification of Tunisian Olive Cultivars: Discrimination Between Varieties, Hybrids and Oleasters. In *Environmental Science and Engineering* (pp. 1109–1113). Springer International Publishing. https://doi.org/10.1007/978-3-030-51210-1_174

Manai-Djebali, H., Oueslati, I., Martínez-Cañas, M. A., Zarrouk, M., & Sánchez-Casas, J. (2018). Improvement of the Sterol and Triacylglycerol Compositions of Chemlali Virgin Olive Oils through Controlled Crossing with Mediterranean Cultivars. *Journal of Oleo Science*, *67*(4), 379–388. https://doi.org/10.5650/jos.ess17105

Mathison, B., & Holstege, D. (2013). A Rapid Method To Determine Sterol, Erythrodiol, and Uvaol Concentrations in Olive Oil. *Journal of Agricultural and Food Chemistry*, *61*(19), 4506–4513. https://doi.org/10.1021/jf400254k

Medina, S., Pereira, J. A., Silva, P., Perestrelo, R., & Câmara, J. S. (2019). Food fingerprints – A valuable tool to monitor food authenticity and safety. *Food Chemistry*, *278*, 144–162. https://doi.org/10.1016/j.foodchem.2018.11.046

Melucci, D., Bendini, A., Tesini, F., Barbieri, S., Zappi, A., Vichi, S., … Gallina Toschi, T. (2016). Rapid direct analysis to discriminate geographic origin of extra virgin olive oils by flash gas chromatography electronic nose and chemometrics. *Food Chemistry*, *204*, 263–273. https://doi.org/10.1016/j.foodchem.2016.02.131

Mendes, T. O., da Rocha, R. A., Porto, B. L. S., de Oliveira, M. A. L., dos Anjos, V. de C., & Bell, M. J. V. (2015). Quantification of extra-virgin olive oil adulteration with soybean oil: a comparative study of NIR, MIR, and Raman spectroscopy associated with chemometric approaches. *Food Analytical Methods*, *8*(9), 2339–2346.

Merás, I. D., Manzano, J. D., Rodríguez, D. A., & de la Peña, A. M. (2018). Detection and quantification of extra virgin olive oil adulteration by means of autofluorescence excitation-emission profiles combined with multi-way classification. *Talanta*, *178*, 751–762.

Mikrou, T., Pantelidou, E., Parasyri, N., Papaioannou, A., Kapsokefalou, M., Gardeli, C., & Mallouchos, A. (2020). Varietal and Geographical Discrimination of Greek Monovarietal Extra Virgin Olive Oils Based on Squalene, Tocopherol, and Fatty Acid Composition. *Molecules*, *25*(17). https://doi.org/10.3390/molecules25173818

Milanez, K. D. T. M., Nóbrega, T. C. A., Nascimento, D. S., Insausti, M., Band, B. S. F., & Pontes, M. J. C. (2017). Multivariate modeling for detecting adulteration of extra virgin olive oil with soybean oil using fluorescence and UV–Vis spectroscopies: A preliminary approach. *LWT-Food Science and Technology*, *85*, 9–15.

Millman, J. F., Okamoto, S., Teruya, T., Uema, T., Ikematsu, S., Shimabukuro, M., & Masuzaki, H. (2021). Extra-virgin olive oil and the gut-brain axis: influence on gut microbiota, mucosal immunity, and cardiometabolic and cognitive health.

Nutrition Reviews, *79*(12), 1362–1374. https://doi.org/10.1093/nutrit/nuaa148

Monasterio, R. P., Fernández, M. de los A., & Silva, M. F. (2013). Olive oil by capillary electrophoresis: Characterization and genuineness. *Journal of Agricultural and Food Chemistry*, *61*(19), 4477–4496.

Montealegre, C., Marina Alegre, M. L., & García-Ruiz, C. (2010). Traceability markers to the botanical origin in olive oils. *Journal of Agricultural and Food Chemistry*, *58*(1), 28–38.

Moore, J. C., Spink, J., & Lipp, M. (2012). Development and application of a database of food ingredient fraud and economically motivated adulteration from 1980 to 2010. *Journal of Food Science*, *77*(4), R118–R126.

Nikou, T., Witt, M., Stathopoulos, P., Barsch, A., & Halabalaki, M. (2020). Olive Oil Quality and Authenticity Assessment Aspects Employing FIA-MRMS and LC-Orbitrap MS Metabolomic Approaches. *Frontiers in Public Health*, *8*(September), 1–20. https://doi.org/10.3389/fpubh.2020.558226

Nocella, C., Cammisotto, V., Fianchini, L., D'Amico, A., Novo, M., Castellani, V., … Carnevale, R. (2018). Extra virgin olive oil and cardiovascular diseases: Benefits for human health. *Endocrine, Metabolic & Immune Disorders-Drug Targets (Formerly Current Drug Targets-Immune, Endocrine & Metabolic Disorders)*, *18*(1), 4–13.

Olmo-Cunillera, A., López-Yerena, A., Lozano-Castellón, J., Tresserra-Rimbau, A., Vallverdú-Queralt, A., & Pérez, M. (2020). NMR spectroscopy: a powerful tool for the analysis of polyphenols in extra virgin olive oil. *Journal of the Science of Food and Agriculture*, *100*(5), 1842–1851. https://doi.org/10.1002/jsfa.10173

Olmo-Cunillera, A., Lozano-Castellón, J., Pérez, M., Miliarakis, E., Tresserra-Rimbau, A., Ninot, A., … Vallverdú-Queralt, A. (2021). Optimizing the Malaxation Conditions to

Produce an Arbequina EVOO with High Content of Bioactive Compounds. *Antioxidants*, *10*(11), 1819. https://doi.org/10.3390/antiox10111819

Omwange, K. A., Al Riza, D. F., Saito, Y., Suzuki, T., Ogawa, Y., Shiraga, K., … Kondo, N. (2021). Potential of front face fluorescence spectroscopy and fluorescence imaging in discriminating adulterated extra-virgin olive oil with virgin olive oil. *Food Control*, *124*, 107906.

Orozco-Solano, M. I., Priego-Capote, F., & Luque de Castro, M. D. (2011). Influence of Simulated Deep Frying on the Antioxidant Fraction of Vegetable Oils after Enrichment with Extracts from Olive Oil Pomace. *Journal of Agricultural and Food Chemistry*, *59*(18), 9806–9814. https://doi.org/10.1021/jf2019159

Orsavova, J., Misurcova, L., Vavra Ambrozova, J., Vicha, R., & Mlcek, J. (2015). Fatty acids composition of vegetable oils and its contribution to dietary energy intake and dependence of cardiovascular mortality on dietary intake of fatty acids. *International Journal of Molecular Sciences*, *16*(6), 12871–12890. https://doi.org/10.3390/ijms160612871

Ozcan-Sinir, G. (2020). Detection of adulteration in extra virgin olive oil by selected ion flow tube mass spectrometry (SIFT-MS) and chemometrics. *Food Control*, *118*, 107433.

Perez, M., Lopez-Yerena, A., & Vallverdú-Queralt, A. (2020). Traceability, authenticity and sustainability of cocoa and chocolate products: a challenge for the chocolate industry. *Critical Reviews in Food Science and Nutrition*, *0*(0), 1–15. https://doi.org/10.1080/10408398.2020.1819769

Pitt, J., Roth, W., Lacor, P., Smith, A. B., Blankenship, M., Velasco, P., … Klein, W. L. (2009). Alzheimer’s-associated Aβ oligomers show altered structure, immunoreactivity and synaptotoxicity with low doses of oleocanthal. *Toxicology and Applied Pharmacology*, *240*(2), 189–197. https://doi.org/10.1016/j.taap.2009.07.018

Poiana, M.-A., Alexa, E., Munteanu, M.-F., Gligor, R., Moigradean, D., & Mateescu, C. (2015). Use of ATR-FTIR spectroscopy to detect the changes in extra virgin olive oil by adulteration with soybean oil and high temperature heat treatment. *Open Chemistry*, *13*(1).

Polari, J. J., Garcí-Aguirre, D., Olmo-García, L., Carrasco-Pancorbo, A., & Wang, S. C. (2018). Impact of industrial hammer mill rotor speed on extraction efficiency and quality of extra virgin olive oil. *Food Chemistry*, *242*, 362–368. https://doi.org/10.1016/j.foodchem.2017.09.003

Psaltopoulou, T., Kosti, R. I., Haidopoulos, D., Dimopoulos, M., & Panagiotakos, D. B. (2011). Olive oil intake is inversely related to cancer prevalence: a systematic review and a meta-analysis of 13800 patients and 23340 controls in 19 observational studies. *Lipids in Health and Disease*, *10*(1), 127. https://doi.org/10.1186/1476-511X-10-127

Quintanilla-Casas, B., Bertin, S., Leik, K., Bustamante, J., Guardiola, F., Valli, E., … Vichi, S. (2020). Profiling versus fingerprinting analysis of sesquiterpene hydrocarbons for the geographical authentication of extra virgin olive oils. *Food Chemistry*, *307*, 125556.

Quintanilla-Casas, B., Strocchi, G., Bustamante, J., Torres-Cobos, B., Guardiola, F., Moreda, W., … Vichi, S. (2021). Large-scale evaluation of shotgun triacylglycerol profiling for the fast detection of olive oil adulteration. *Food Control*, *123*. https://doi.org/10.1016/j.foodcont.2020.107851

Quintanilla-Casas, B., Torres-Cobos, B., Guardiola, F., Servili, M., Alonso-Salces, R. M., Valli, E., … Tres, A. (2022). Geographical authentication of virgin olive oil by GC–MS sesquiterpene hydrocarbon fingerprint: Verifying EU and single country label-declaration. *Food Chemistry*, *378*, 132104. https://doi.org/10.1016/j.foodchem.2022.132104

Rotondi, A., Morrone, L., Bertazza, G., & Neri, L. (2021). Effect of Duration of Olive Storage on Chemical and Sensory Quality of Extra Virgin Olive Oils. *Foods*, *10*(10), 2296. https://doi.org/10.3390/foods10102296

Ruiz-Aracama, A., Goicoechea, E., & Guillén, M. D. (2017). Direct study of minor extra-virgin olive oil components without any sample modification. 1 H NMR multisupression experiment: A powerful tool. *Food Chemistry*, *228*, 301–314. https://doi.org/10.1016/j.foodchem.2017.02.009

Salivaras, E., & McCurdy, A. R. (1992). Detection of olive oil adulteration with canola oil from triacylglycerol analysis by reversed-phase high-performance liquid chromatography. *Journal of the American Oil Chemists Society*, *69*(9), 935–938.

Sánchez-Hernández, L., Marina, M. L., & Crego, A. L. (2011). A capillary electrophoresis–tandem mass spectrometry methodology for the determination of non-protein amino acids in vegetable oils as novel markers for the detection of adulterations in olive oils. *Journal of Chromatography A*, *1218*(30), 4944–4951.

Sánchez-López, E., Sánchez-Rodríguez, M. I., Marinas, A., Marinas, J. M., Urbano, F. J., Caridad, J. M., & Moalem, M. (2016). Chemometric study of Andalusian extra virgin olive oils Raman spectra: Qualitative and quantitative information. *Talanta*, *156–157*, 180–190. https://doi.org/10.1016/j.talanta.2016.05.014

Sanmartin, C., Venturi, F., Sgherri, C., Nari, A., Macaluso, M., Flamini, G., … Zinnai, A.

(2018). The effects of packaging and storage temperature on the shelf-life of extra virgin olive oil. *Heliyon*, *4*(11), e00888. https://doi.org/10.1016/j.heliyon.2018.e00888

Sanz-Cortés, F., Parfitt, D. E., Romero, C., Struss, D., Llácer, G., & Badenes, M. L. (2003). Intraspecific olive diversity assessed with AFLP. *Plant Breeding*, *122*(2), 173–177.

Schneider, S. (2016). Quality analysis of virgin olive oils – Part 1 Thermal treatment analysis – determination of 3,5-stigmastadienes in olive oil using the Agilent Infinity 1220 LC System with Diode Array Detector. *Agilent Technologies*, 5991-1894EN. Retrieved from https://www.agilent.com/cs/library/applications/5991-1894EN.pdf

Shi, J., Yuan, D., Hao, S., Wang, H., Luo, N., Liu, J., … Chen, Z. (2019). Stimulated Brillouin scattering in combination with visible absorption spectroscopy for authentication of vegetable oils and detection of olive oil adulteration. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, *206*, 320–327.

Silva, S. F., Anjos, C. A. R., Cavalcanti, R. N., & Celeghini, R. M. D. S. (2015). Evaluation of extra virgin olive oil stability by artificial neural network. *Food Chemistry*, *179*, 35–43. https://doi.org/10.1016/j.foodchem.2015.01.100

Srigley, C. T., Oles, C. J., Kia, A. R. F., & Mossoba, M. M. (2016). Authenticity Assessment of Extra Virgin Olive Oil: Evaluation of Desmethylsterols and Triterpene Dialcohols. *Journal of the American Oil Chemists' Society*, *93*(2), 171–181. https://doi.org/10.1007/s11746-015-2759-4

Stadler, R. H., Tran, L.-A., Cavin, C., Zbinden, P., & Konings, E. J. M. (2016). Analytical approaches to verify food integrity: needs and challenges. *Journal of AOAC*

International, *99*(5), 1135–1144.

Tan, J., Li, R., Jiang, Z., Shi, M., Xiao, Y., Jia, B., … Wang, H. (2018). Detection of Extra Virgin Olive Oil Adulteration With Edible Oils Using Front-Face Fluorescence and Visible Spectroscopies. *Journal of the American Oil Chemists' Society*, *95*(5), 535–546.

Tapp, H. S., Defernez, M., & Kemsley, E. K. (2003). FTIR Spectroscopy and Multivariate Analysis Can Distinguish the Geographic Origin of Extra Virgin Olive Oils. *Journal of Agricultural and Food Chemistry*, *51*(21), 6110–6115. https://doi.org/10.1021/jf030232s

Tena, N., Wang, S. C., Aparicio-Ruiz, R., García-González, D. L., & Aparicio, R. (2015). In-Depth Assessment of Analytical Methods for Olive Oil Purity, Safety, and Quality Characterization. *Journal of Agricultural and Food Chemistry*, *63*(18), 4509–4526. https://doi.org/10.1021/jf5062265

Tfouni, S. A. V., Reis, R. M., Amaro, N. de P. L., Pascoal, C. R., de Camargo, M. C. R., Baggio, S. R., … Furlani, R. P. Z. (2017). Adulteration and presence of polycyclic aromatic hydrocarbons in extra virgin olive oil sold on the Brazilian market. *Journal of the American Oil Chemists' Society*, *94*(11), 1351–1359.

Tiryaki, G. Y., & Ayvaz, H. (2017). Quantification of soybean oil adulteration in extra virgin olive oil using portable raman spectroscopy. *Journal of Food Measurement and Characterization*, *11*(2), 523–529.

Toledo, E., Salas-Salvadó, J., Donat-Vargas, C., Buil-Cosiales, P., Estruch, R., Ros, E., … Martínez-González, M. A. (2015). Mediterranean Diet and Invasive Breast Cancer Risk Among Women at High Cardiovascular Risk in the PREDIMED Trial. *JAMA Internal Medicine*, *175*(11), 1752.

https://doi.org/10.1001/jamainternmed.2015.4838

Torrecilla, J. S., Rojo, E., Dominguez, J. C., & Rodríguez, F. (2010). A novel method to quantify the adulteration of extra virgin olive oil with low-grade olive oils by UV−vis. *Journal of Agricultural and Food Chemistry*, *58*(3), 1679–1684.

Tresserra-Rimbau, A., Rimm, E. B., Medina-Remón, A., Martínez-González, M. A., De la Torre, R., Corella, D., … Arós, F. (2014). Inverse association between habitual polyphenol intake and incidence of cardiovascular events in the PREDIMED study. *Nutrition, Metabolism and Cardiovascular Diseases*, *24*(6), 639–647.

Tsopelas, F., Konstantopoulos, D., & Kakoulidou, A. T. (2018). Voltammetric fingerprinting of oils and its combination with chemometrics for the detection of extra virgin olive oil adulteration. *Analytica Chimica Acta*, *1015*, 8–19.

Ün, İ., & Salim, O. K. (2018). Analysis of olive oil for authentication and shelf life determination. *Journal of Food Science and Technology*, *55*(7), 2476–2487.

Uncu, O., & Ozen, B. (2019). A comparative study of mid-infrared, UV–Visible and fluorescence spectroscopy in combination with chemometrics for the detection of adulteration of fresh olive oils with old olive oils. *Food Control*, *105*, 209–218.

Uncu, O., & Ozen, B. (2020). Importance of some minor compounds in olive oil authenticity and quality. *Trends in Food Science & Technology*, *100*, 164–176.

Valli, E., Milani, A., Srbinovska, A., Moret, E., Moret, S., Bendini, A., … Lucci, P. (2021). In-House Validation of an SPE-GC-FID Method for the Detection of Free and Esterified Hydroxylated Minor Compounds in Virgin Olive Oils. *Foods*, *10*(6), 1260. https://doi.org/10.3390/foods10061260

Van Durme, J., & Vandamme, J. (2016). Non-thermal plasma as preparative technique to evaluate olive oil adulteration. *Food Chemistry*, *208*, 185–191.

Varona, E., Tres, A., Rafecas, M., Vichi, S., Barroeta, A. C., & Guardiola, F. (2021). Composition and Nutritional Value of Acid Oils and Fatty Acid Distillates Used in Animal Feeding. *Animals*, *11*(1), 196. https://doi.org/10.3390/ani11010196

Vávra, A., Hájek, M., & Kocián, D. (2021). The influence of vegetable oils composition on separation of transesterification products, especially quality of glycerol. *Renewable Energy*, *176*, 262–268. https://doi.org/10.1016/j.renene.2021.05.050

Vera, D. N., Jiménez-Carvelo, A. M., Cuadros-Rodríguez, L., Ruisánchez, I., & Callao, M. P. (2019). Authentication of the geographical origin of extra-virgin olive oil of the Arbequina cultivar by chromatographic fingerprinting and chemometrics. *Talanta*, *203*, 194–202. https://doi.org/10.1016/j.talanta.2019.05.064

Violi, F., Loffredo, L., Pignatelli, P., Angelico, F., Bartimoccia, S., Nocella, C., … Carnevale, R. (2015). Extra virgin olive oil use is associated with improved post-prandial blood glucose and LDL cholesterol in healthy subjects. *Nutrition & Diabetes*, *5*(7), e172–e172. https://doi.org/10.1038/nutd.2015.23

Violino, S., Benincasa, C., Taiti, C., Ortenzi, L., Pallottino, F., Marone, E., … Costa, C. (2021). AI-based hyperspectral and VOCs assessment approach to identify adulterated extra virgin olive oil. *European Food Research and Technology*, *247*(4), 1013–1022.

Violino, S., Pallottino, F., Sperandio, G., Figorilli, S., Antonucci, F., Ioannoni, V., … Costa, C. (2019). Are the innovative electronic labels for extra virgin olive oil sustainable, traceable, and accepted by consumers? *Foods*, *8*(11), 529.

Wang, Y., Zhu, M., Mei, J., Luo, S., Leng, T., Chen, Y., … Xie, M. (2019). Comparison of Furans Formation and Volatile Aldehydes Profiles of Four Different Vegetable Oils During Thermal Oxidation. *Journal of Food Science*, *84*(7), 1966–1978.

https://doi.org/10.1111/1750-3841.14659

Weesepoel, Y., Alewijn, M., Wijtten, M., & Müller-Maatsch, J. (2021). Detecting food fraud in extra virgin olive oil using a prototype portable hyphenated photonics sensor. *Journal of AOAC International*, *104*(1), 7–15.

World Health Organization. (2005). Summary and conclusions of the sixty-fourth meeting of the joint FAO/WHO Expert Committee on Food Additives. *World Health Organization: Rome*.

Yahay, M., Heidari, Z., Allameh, Z., & Amani, R. (2021). The effects of canola and olive oils consumption compared to sunflower oil, on lipid profile and hepatic steatosis in women with polycystic ovarian syndrome: a randomized controlled trial. *Lipids in Health and Disease*, *20*(1), 7. https://doi.org/10.1186/s12944-021-01433-9

Yang, H., & Irudayaraj, J. (2001). Comparison of near-infrared, Fourier transform-infrared, and Fourier transform-Raman methods for determining olive pomace oil adulteration in extra virgin olive oil. *Journal of the American Oil Chemists' Society*, *78*(9), 889–895.

Yang, Y., Ferro, M. D., Cavaco, I., & Liang, Y. (2013). Detection and identification of extra virgin olive oil adulteration by GC-MS combined with chemometrics. *Journal of Agricultural and Food Chemistry*, *61*(15), 3693–3702. https://doi.org/10.1021/jf4000538

Youseff, R., Soubh, L., & Alassaf, Z. (2014). Detection of vegetable oils adulteration using desmethylsterols composition. *International Journal of Pharmaceutical Sciences Review and Research*, *28*(2), 229–233.

Yubero-Serrano, E. M., Lopez-Moreno, J., Gomez-Delgado, F., & Lopez-Miranda, J. (2019). Extra virgin olive oil: More than a healthy fat. *European Journal of Clinical*

Nutrition, *72*(Table 1), 8–17. https://doi.org/10.1038/s41430-018-0304-x

Zhou, X., Zhang, Q., Chen, X., Li, X., & Han, C. (2021). In-situ assessment of olive oil adulteration with soybean oil based on thermogravimetric-gas chromatography/mass spectrometry combined with chemometrics. *Food Control*, *130*, 108251.

Tables

Table 1. Summary of the trending or novel analytical techniques, biomarker compounds, and data processing methods for the detection of EVOO adulterants.

Methods	Determination	Data processing	Adulterants	Ref
GC-FID	FA composition	PCA	Geographical varieties	(Mikrou et al., 2020)
GC-FID	FA composition	Direct comparison	Corn, sunflower, soybean, and canola oils	(Aykas et al., 2020)
GC-MS	FA composition	CARS-PLS-LDA and MCTree	Corn, peanut, rapeseed, and sunflower oils	(Yang et al., 2013)
SPE-GC-FID	DAG and FFA	Direct comparison	Soft deodorized oil	(Gómez-Coca et al., 2020)
GC-FID	FA composition	Direct comparison	Refined soybean oil	(da Silveira et al., 2017)
MALDI-TOF MS	Complete lipidomic profile	UHC, PCA and Pearson’s correlation	Corn oil	(Di Girolamo et al., 2015)
FIA-HESI-HRMS	TAG profile	PLS-DA	High linoleic and high oleic vegetable oils	(Quintanilla-Casas et al., 2021)
FIA-MRMS	TAG, DAG and FFA	PCA and OPLS-DA	Geographical varieties	(Nikou et al., 2020)
FT-NIR	FA composition	PLS	Oils high in linoleic or oleic acid, palm olein, and refined olive oil	(Azizian et al., 2016)
FT-NIR	Volatile compounds and FA	PCA and SIMCA, PLS	Soybean, sunflower, corn, canola, hazelnut, high oleic acid safflower, peanut and refined olive oils, and palm olein	(Azizian et al., 2015)
FT-NIR	1,2-DAG, 1,3-DAG and FFA	PLS	Oils high in linoleic or oleic acid, palm olein, and refined olive oil	(Azizian et al., 2018)
FT-Raman	FA composition	PLS	Geographic location, olive variety, harvest year and PDO	(Sánchez-López et al., 2016)
FT-Raman	FA composition	iPLS and SiPLS	Waste cooking oil	(Li et al., 2018)
MALDI-TOF-MS	Phospholipids	Direct comparison	Hazelnut oil	(Calvano et al., 2012)
FL	Tocopherols, phenolic compounds and chlorophylls	PLSR and ANN, PCA	Corn, soybean, linseed, or sunflower oils	(Lia et al., 2018)
FL	Tocopherols, phenolic compounds and pigments	LDA-PARAFAC and UPLS-DA	ROO, RPOO	(Merás et al., 2018)
EEM-FL	Tocopherols, tocotrienols, phenolic compounds and oxidation products	SVM and PLSR	EVOO vs VOO	(Omwange et al., 2021)

HPLC-FL	Tocopherols (α/β-tocopherol ratio)	Direct comparison	Sunflower, hazelnut and peanut oils	(Chen et al., 2011)
Voltametric analysis	Phenolic compounds and tocopherols	PCA, PLS-DA, SIMCA, PLSR	Sunflower, soybean, and corn oils	(Tsopelas et al., 2018)
e-tongue	Phenolic compounds and tocopherols	PLS-DA and PLSR	Sunflower, soybean and corn oils	(Apetrei & Apetrei, 2014)
HPLC-DAD	Phenolic compounds	PLSR	Different cultivars, sunflower oil and ROO	(Carranco et al., 2018)
GC-FID, HPLC-FL, HPLC-DAD-ESI/MS, UV	Volatile substances, polar phenolic substances, antioxidant activity, FA composition, and α-tocopherol	PCA	Low price vs high price EVOO	(Fiorini et al., 2018)
LC-DAD/ESI–MS/MS, GC-FID	Phenolic compounds and sterol profile	PCA and UHC	RPOO	(Drira et al., 2020)
HPLC-DAD	Squalene and tyrosol	Scatter diagram of standard scores (z)	Sunflower and grapeseed oils	(Hayakawa et al., 2020)
SIFT-MS	Volatile compounds	SIMCA, PLSR	Corn, sunflower, high oleic sunflower, and olive oils	(Ozcan-Sinir, 2020)
GC-MS, GC-FID, GC-O-MS, ADEA	Volatile compounds and FA	PCA	RPOO	(Drira et al., 2021)
HS-SPME-GC–MS	Volatile compounds	PCA and SIMCA	Sunflower oil	(Van Durme & Vandamme, 2016)
vis-NIR	Volatile compounds and pigments	Artificial intelligence model	Corn, sunflower, rice, peanut, hazelnut, virgin wheat germ and virgin cornstarch oils	(Violino et al., 2021)
GC-IMS, FGC E-nose	Volatile compounds	SIMCA	ROO	(Damiani et al., 2020)
UV-vis	Pigments, phenolic compounds and tocopherols	PLS-JK, SW-MLR, GA-MLR	Soybean oil	(Milanez et al., 2017)
SBS-vis	Pigments	Linear and surface regressions	Rapeseed, soybean, peanut, and sunflower oils	(Shi et al., 2019)
FL spectroscopy	Pigments and oxidation compounds	PCA, PLSR	Sunflower oil	(Ali et al., 2018)
Frontface-FL and vis	Pigments, phenolic compounds and tocopherols	PLSR	Corn, soybean and sunflower oils	(Tan et al., 2018)
vis	Pigments, phenolic compounds and tocopherols	ANN, MLP	ROO	(Aroca-Santos et al., 2016)
vis	Pigments, phenolic compounds and tocopherols	ANN	ROO, PO, sunflower and corn oils	(Aroca-Santos et al., 2015)
vis	Chlorophylls and carotenoids	CR, PCA and LDA	OO, PO	(Ferreiro-González et

				al., 2017)
MidIR, UV-vis, FL	Pigments, phenolic compounds, tocopherols, FA and oxidation products	OPLS-DA and PLSR	Aged EVOOs	(Uncu & Ozen, 2019)
GC-FID	Campesterol and stigmasterol	Direct comparison	Corn, soybean, sunflower, and cotton oils	(Al-Ismail et al., 2010)
SPE-GC-FID	Uvaol, sterol and erythrodiol	Direct comparison	Pomace olive oil	(Mathison & Holstege, 2013)
HPLC-DRI, GC-FID	FA and sterol profile	LDA	Soybean, sunflower and corn oils	(Jabeur et al., 2014)
TLC-GC-FID	Desmethylsterols	Direct comparison	Sunflower and soybean oils	(Youseff et al., 2014)
GC-FID	Desmethylsterols and triterpene dialcohols	Direct comparison	Canola, corn, peanut, safflower, soybean, and sunflower oils	(Srigley et al., 2016)
^{1}H NMR	Acyl groups, squalene, sterols, triterpene acids/esters, fatty alcohols, wax esters and phenols (lignans, tyrosol, hydroxytyrosol, oleocanthal, oleacein, oleokoronal, oleomissional, ligstrodials and oleuropeindials)	Direct comparison	Different cultivars	(Ruiz-Aracama et al., 2017)
HPLC-DRI, GC-FID	FA and sterol profiles and ΔECN42	PCA	Cotton and sunflower oils	(Kesen, 2019)
GC-FID	Sterol profile	PCA, UHC	Different cultivars and EVOO from oleasters	(Manai-Djebali et al., 2021)
SPE-GC-FID	Free and esterified sterols and free and esterified triterpenic alcohols	Linear regression	Sunflower oil	(Valli et al., 2021)

GC-FID: gas chromatography flame ionization detector; FA: fatty acid; PCA: principal component analysis; GC-MS: gas chromatography mass spectrometry; CARS-PLS-LDA: competitive adaptive reweighted sampling partial least square linear discriminant analysis; MCTree: Monte Carlo tree; SPE: solid phase extraction; DAG: diacylglycerides; FFA: free fatty acids; MALDI-TOF-MS: matrix-assisted laser desorption/ionization mass spectrometry; UHC: unsupervised hierarchical cluster; FIA-HESI-HRMS: flow injection analysis-heated electrospray ionization high resolution mass spectrometry; TAG: triacylglycerides; PLS-DA: partial least square discriminant analysis; FIA-MRMS: flow injection analysis magnetic resonance mass spectrometry; OPLS-DA: orthogonal projection to latent structures discriminant analysis; FT-NIR: Fourier transform near infrared spectroscopy; PLS: partial least square; SIMCA: soft independent modeling of class analogy; 1,2-DAG: diacylglycerides in position 1 and 2; 1,3-DAG: diacylglycerides in position 1 and 3; PDO: protected designation of origin; iPLS: interval partial least square; SiPLS: synergy interval partial least square; FL: fluorescence spectroscopy; PLSR: partial least squares regression; ANN: artificial neural network; ROO: refined olive oil; RPOO: refined pomace olive oil; LDA-PARAFAC: linear discriminant analysis parallel factor analysis; UPLS-DA: unfolded partial least squares discriminant analysis; EEM-FL: excitation emission matrix fluorescence spectroscopy; EVOO: extra virgin olive oil; VOO: virgin olive oil; SVM: support-vector machine model; HPLC-FL: high-performance liquid chromatography-fluorescence spectroscopy; e-tongue: electronic tongue; HPLC-DAD: high-performance liquid chromatography diode array detector; HPLC-DAD-ESI/MS: high-performance liquid chromatography diode array detector electrospray ionization mass spectrometry; UV: Ultraviolet spectroscopy; LC DAD/ESI–MS/MS: liquid chromatography diode array detector electrospray ionization mass spectrometry in tandem; SIFT-MS: selected ion flow tube mass spectrometry; GC-O-MS: gas chromatography olfactometry mass spectrometry; ADEA: aroma dilution extraction assay; HS-SPME-GC-MS: headspace solid phase microextraction gas chromatography mass spectrometry; vis: visible spectroscopy; GC-IMS: gas chromatography ionic mobility spectrometry; FGC E-nose: flash gas chromatography electronic nose; PLS-JK: partial least squares Jack-Knife; SW-MLR stepwise multiple linear regression; GA-MLR: genetic algorithm multiple linear regression; SBS- vis: stimulated Brillouin scattering visible spectroscopy; MLP: multilayer perception model; PO: pomace olive oil; CR: multivariate curve resolution method; LDA: linear discriminant analysis; MidIR: Mid infra-red spectroscopy; HPLC-DRI: high-performance liquid chromatography differential refractometer detector; TLC-GC-FID: thin layer chromatography gas chromatography flame ionization detector; 1H NMR: proton nuclear magnetic resonance.

Table 2 Identification of EVOO adulteration by assessing VOCs.

Samples	**Analysis**	**Findings**	**Ref**
EVOO and adulterated EVOO (1, 2.5, 5, 10, and 20% of corn oil, sunflower oil, high oleic sunflower oil, and olive oil)	- SIFT-MS, SIMCA algorithm, PLSR model. - The excellence of the final model was calculated based on the number of latent variables, loading vectors, SECV, the coefficient of determination (R-value), SEP, and outlier diagnostics.	1-Octanol, 1-penten-3-one, 2-phenylethanol, dodecane, anisole, ethyl nonanoate, isobutanoic acid, ocimene, phenol, and toluene were the compounds that most successfully classified adulteration.	(Ozcan-Sinir, 2020)
EVOO adulterated with RPOO (1, 2, 3, 5, 6, 10 and 20%)	- GC-MS, GC-FID, GC-O-MS and AEDA	Thirty-four relevant aroma compounds and twenty-one key odorants were quantified in EVOO, RPOO, and EVOO adulterated with 1–20% of RPOO.	(Drira et al., 2021)
EVOO and EVOO adulterated with sunflower oil (1%)	- NTP, PCA and SIMCA	NTP treatments of 60 min (Ar/O_2 0.1%) resulted in the formation of a unique set of secondary volatile lipid oxidation products enabling classification of adulterated oil samples.	(Van Durme & Vandamme, 2016)
EVOO and EVOO adulterated with RPOO	- FT-NIR, PLS1 and FT-NIR	FT-NIR cannot distinguish between naturally occurring volatile carbonyl-type compounds in edible oils and those derived from subsequent oxidation.	(Azizian et al., 2015)
EVOO, olive oil, pure seed oils and olive oil samples adulterated with 7 different seed oils in different ratios	- vis–NIR and artificial intelligence model	Analyzing the data produced by the instruments using artificial intelligence methods accurately distinguished between EVOO adulterated with sophisticated techniques and pure EVOO	(Violino et al., 2021)
EVOO blended with soft-refined olive oils	- GC-IMS, FGC E-nose and SIMCA	Volatile fraction analysis might be the right strategy to overcome the lack of clear and specific process-related markers formed in soft-refinement processes.	(Damiani et al., 2020)

FGC E-nose: flash gas chromatography electronic nose; FT-NIR: Fourier transform near infrared; GC-IMS: gas-chromatography ion mobility spectrometry; NTP: non-thermal plasma; PCA: principal component analysis; PLS1: partial least squares; GC-O-MS: gas chromatography–olfactometry-mass spectrometry; PLSR; partial least squares regression; SECV: standard error of cross-validation; RPOO: refined pomace olive oil; SEP; standard error of prediction; SIFT-MS: selected ion flow tube mass spectrometry; SIMCA: soft independent modeling of class analogy; VOCs: volatile organic compounds.

Table 3. General regimen for food and other agricultural products based on Regulation 510/2006

General regimen	**Origin**	**Characteristics**	**Restriction**
Protected Designation of Origin (PDO)	Region, specific place, or country	Quality essentially or exclusively due to a particular geographical area.	Produced, processed, and prepared in a given geographical area.
Protected Geographical Indication (PGI)	Region, specific place, or country	Slightly less strict; food reputation of a product from a given region is sufficient.	One of the stages of production, processing or preparation takes place in the area.

Data take from (COMMUNITY, 2006)

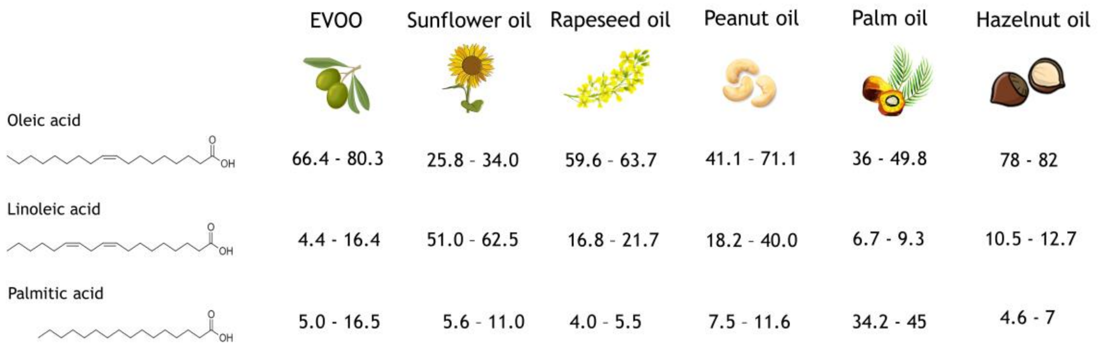
Table 4. Relevant compositional markers for establishing the botanical traceability of olive oils.

Compositional marker	Relevant chemical marker analysed	Olive/VOO variety example	Ref
FA	C18:0, C18:1	Cornicabra	(Aranda et al., 2004)
	C16:0, C17:1	Arbequina	
	C17:1 and C18:0	Hojiblanca	
	C16:0, C18:0, C18:1	Picual	
TAG	OOO and SLO + POO	Manzanilla Cacereña	(Diaz et al., 2005)
	SOO, LOO and PLO	Non manzanilla Cacereña	
Sterols	Stigmasterol	Cobrançosa	(Alves et al., 2005)
	β-sitoesterol/ Δ^5-avenasterol	Madural	
	Campesterol	Verdeal	
Phenolic compounds	p-HPEA-EDA and ligstroside aglycon	Cornicabra	(Gómez-Alonso et al., 2002)
	1-Acetoxypinoresinol + trans-cinnamic acid and 3,4-DHPEA-AC	Arbequina	
	3,4-DHPEA-AC	Hojiblanca	
	ligstroside aglycon	Picual	
Volatile compounds	(E)-Hex-2-enal	Bidh Hman, Rekhami, Jarboui 1, Regregui	(Haddada et al., 2007)
	Ethanol, 2-Methylpentane, (E)-Hex-2-enol and Hexanol	Jarboui 2	
	(Z)-pent-2-enol and two isomers of 3,4-diethylhexa-1,5-diene	Ain Jarboua, Chétoui 1 and 2, Neb Jmel	
Pigments	Lutein/β-carotene ratio=0.27	Cerasuola	(Giuffrida et al., 2007)
	Lutein/β-carotene ratio=0.4	Nocellara	
	Lutein/β-carotene ratio=0.17	Biancolilla	
Hydrocarbons	C29:0 to C34:0	Cacereña	(Bueno et al., 2005)
	C13:1	Carrasqueña	
	C24:0 to C29:0, C13:1	Corniche	
	C13:1	Picual	

FA: Fatty Acid; TAG: Triacylglycerol; L, O, P, and S: linoleoyl, oleoyl, palmitoyl, and stearoyl fat acid radicals, respectively; VOO: virgin olive oil; 3,4-DHPEA-AC: hydroxytyrosol acetate.

Figures

Figure 1:



	EVOO	Sunflower oil	Rapeseed oil	Peanut oil	Palm oil	Hazelnut oil
Oleic acid	66.4 - 80.3	25.8 - 34.0	59.6 - 63.7	41.1 - 71.1	36 - 49.8	78 - 82
Linoleic acid	4.4 - 16.4	51.0 - 62.5	16.8 - 21.7	18.2 - 40.0	6.7 - 9.3	10.5 - 12.7
Palmitic acid	5.0 - 16.5	5.6 - 11.0	4.0 - 5.5	7.5 - 11.6	34.2 - 45	4.6 - 7

Figure 1. Main FA components of vegetable oils, ranges in % obtained from: (Alves et al., 2019; Chen et al., 2020; Feizabadi et al., 2021; Holey et al., 2021; Konuskan et al., 2019; Król, Gantner, & Piotrowska, 2021; Rotondi, Morrone, Bertazza, & Neri, 2021b; Vávra, Hájek, & Kocián, 2021b; Wang et al., 2019; Yahay et al., 2021)

Figure 2:

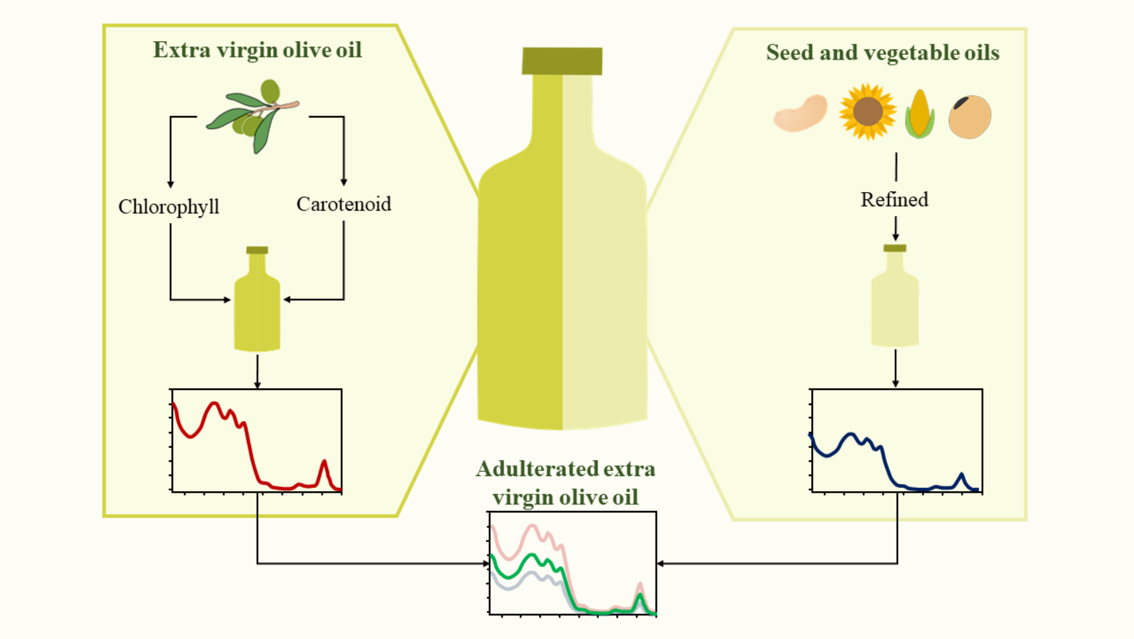


Figure 2. Comparison of pigment profiles in EVOO and seed and vegetable oils, in red pure EVOO, in green EVOO adulterated with refined oils, in blue or grey refined seed and vegetable oils.

Figure 3:

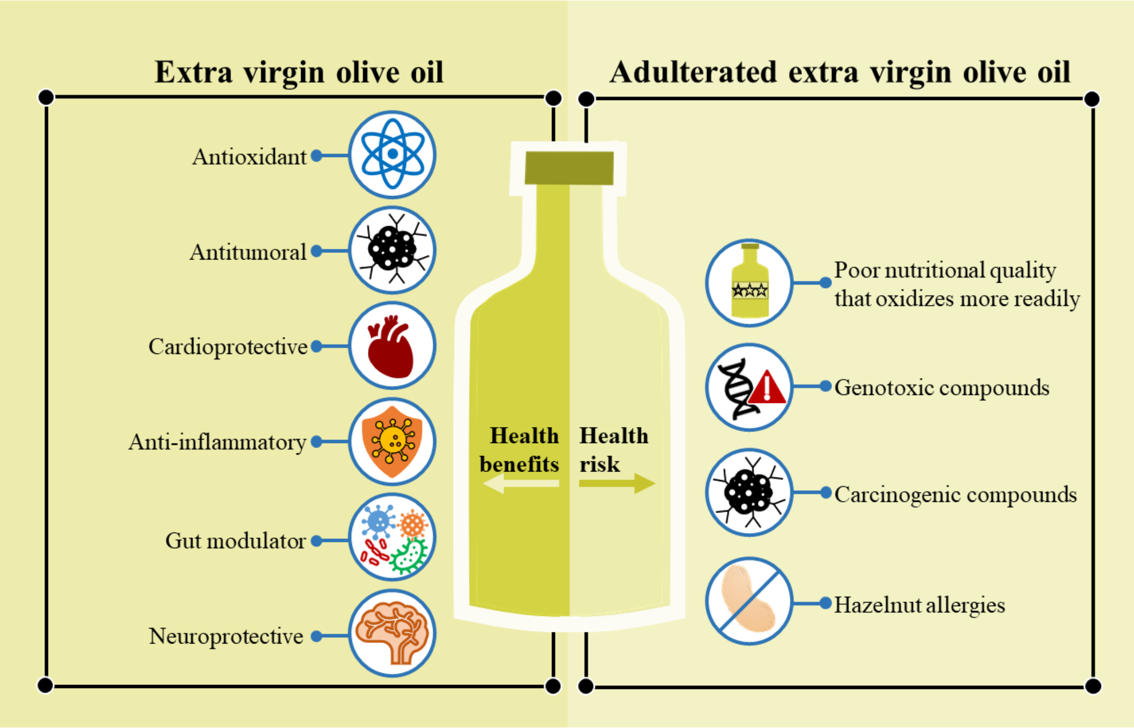


Figure 3. General description of health benefits of extra virgin olive oil and health risk derived from the consumption of adulterated extra virgin olive oil.