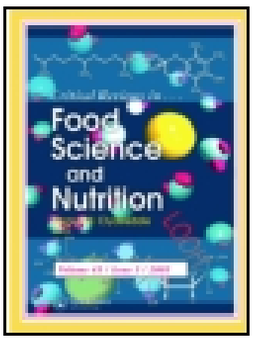

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REVIEW

Application of new emerging techniques in combination with classical methods for the determination of the quality and authenticity of olive oil: a review

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ABSTRACT

The determination of the quality and authenticity of olive oil becomes more and more required by producers, consumers, and authorities to thwarter falsification. Several analytical techniques including chemical, sensory, chromatography, and so on, are used for the determination of the quality and authenticity of olive oil. Although these methods are considered as the reference ones, they are cumbersome, time-consuming and destructive. Therefore, rapid analytical techniques such as fluorescence, ultraviolet-visible, near infrared, and mid infrared spectroscopies, electronic sensing, among others, are more and more used for the determination of the quality and authenticity of olive oils. This review will identify current gaps related to different analytical techniques in olive oil authentication and discuss the drawbacks of existing analytical methods concerning olive oil authenticity from 2010 up to now.

KEYWORDS

Authenticity; chemometry; olive oil; quality; traceability

Introduction

Olive oil (OO) is a liquid vegetable fat obtained by different systems from olive fruits of the olive tree (*Olea europaea L.*) that its cultivation dates back to biblical times. Under the Roman Empire, the olive tree contributed to the *Pax Romana* by offering the conquered countries a product of great commercial, economic, and health value. Indeed, from Greece to Spain via Egypt, Italy, Tunisia, France, Spain, and Morocco, the culture of the olive tree was established throughout the Mediterranean for two thousand years (Vossen 2007). Virgin olive oil (VOO) is: i) extracted directly from olive fruits by mechanical processes; ii) obtained by washing and crushing olives, mixing the dough, separating the phases by pressing and settling or by centrifugation, and iii) filtered to obtain pure VOO (COI. 2016) .

The olive sector presents an important economic activity in the Mediterranean countries. As an example, table olives and OO sector represent 5% of the Agricultural gross domestic product of Morocco with a production of 1 million tons of table olives and 330,000 tons of OO during the 2018/2019 season of which nearly 80,000 tons were exported to Europe(COI. 2018) .

The economic importance of OO is also linked to its high nutritional value since it is considered an important lipid source of the Mediterranean diet. Indeed, it provides essential fatty acids and gives beneficial properties for health thanks to its high proportion of unsaturated fatty acids such as oleic and linoleic acids. In addition, OO is known to present anti-inflammatory, anti-atherogenic, antimicrobial, antiviral, anti-aging (Cicerale et al. 2009; Covas 2007), and neuroprotective effects (Angelova et al. 2018).

Fatty acids representing ~ 99% of the total chemical composition of OO, the remaining 1% is made of minor compounds such as squalene, triterpenic alcohols, sterols, phenolic compounds, chlorophylls, β-carotenes, and tocopherols (COI. 2016). The important minor compounds of OO are phenolic and tocopherols compounds that are among the main compounds giving OO its high nutritional, pharmacological, and medicinal values. Indeed, polyphenol compounds are well known as natural antioxidants inhibiting the oxidation of olive fruit and oil basing on the mechanism of scavenging hydroxyl and lipid radicals by trapping intermediate peroxyl radical (Martín-Peláez et al. 2013). In this context, tocopherol and polyphenol compounds were used to predict the quality of OO knowing that qualitative and quantitative composition of these minor compounds depends on several factors such as the geographical origin, the cultivars, the attack of insects, the mode of irrigation of trees, the maturity index, the storage conditions of fruits and oil, the harvesting methods, and the extraction modes (Mraicha et al. 2010, Bajoub et al. 2014).

The OO labeled as protected designation of origin (PDO), protected geographical indication (PGI) or registered designation of origin (RDO) are typified by some specific characteristics related to variety, geographical origin, agronomic conditions, fruit ripeness milling/extraction process, and so on (Bajoub et al. 2014). Taking into consideration that foods presenting high-value are the most vulnerable products for falsification (Kamal and Karoui 2015), the OO labeled as PDO,

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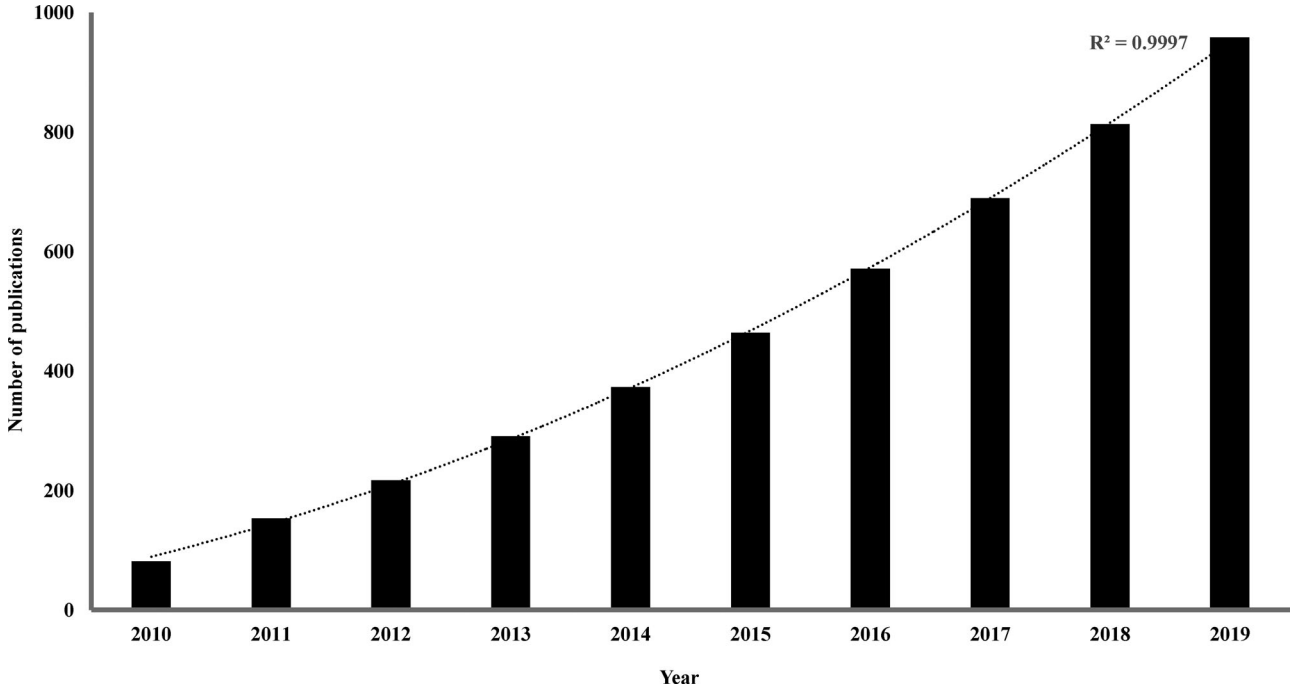

Figure 1. Cumulative number of studies investigating the determination of olive oil quality and authenticity using spectroscopic techniques since the year 2010. Information obtained from the database Scopus (Search criteria: Article title, Abstract, Keywords: Olive oil, AND Article title, Abstract, Keywords: Spectroscopy). The data were obtained on 12 June 2020.

PGI and RDO are subjected to different types of frauds. The temptations take several dimensions depending on the initial quality and composition of the adulterated OO.

Various analytical techniques have been developed for the determination of the quality and authenticity of OO. These techniques include chemical, sensorial, chromatographic, and so on (Arvanitoyannis and Vlachos 2007; Valli et al. 2016). However, most of the chemical and chromatographic techniques are subjected to certain limitations, as they are time-consuming and destructive. Therefore, the development of rapid, noncontact, nondestructive, and real-time measurements is of utmost importance (Zaroual, El Hadrami, and Karoui 2020). According to the Scopus database, the number of publications regarding the use of spectroscopic methods for determining the quality and authenticity of OO (Figure 1) increased from 80 publications in the year 2010 to more than 145 papers in the year 2019. This could be explained by: i) the growing awareness in the oil sector of the importance of these techniques compared to classical methods; and ii) spectroscopic techniques, known as un-targeted ones, give chemical and structure information allowing them to be considered as a fingerprint.

This present review paper will provide a comprehensive overview of the applications of different analytical techniques, in combination with multivariate data analysis, to determine the quality and authenticity of OO during the last 10 years (starting 2010).

Classical methods used for the determination of the quality and authenticity of olive oils

Chemical analysis

The well-known chemical parameters used for the determination of the quality and authenticity of OO and to classify VOO grade are the free acidity (FA), peroxide value (PV) and specific extinctions (k_{232}, k_{270} and Δk) obtained according to the official methods under the code COI/T.20/Doc/N°34, N°35, and N°19, respectively. Indeed, basing on these chemical parameters, VOO are classified into 4 classes: i) extra VOO (EVOO) (FA $\leq$ 0.8%, PV $\leq$ 20 meq O_2/kg oil, $k_{232} \leq 2.50$, $k_{270} \leq 0.22$, and $\Delta k \leq 0.01$), ii) VOO (0.8% $<$ FA $\leq$ 2, PV $\leq$ 20 meq O_2/kg oil, $k_{232} \leq 2.50$, $k_{270} \leq 0.30$, and $\Delta k \leq 0.01$), iii) ordinary VOO ($2 <$ FA $< 3.3\%$, PV $\leq$ 20 meq O_2/kg oil, $_{k232} \leq 2.50$, $k_{270} \leq 0.22$, and $\Delta k \leq 0.01$), iv) lampante VOO (FA $> 3.3\%$, and no limit of PV, k_{232}, k_{270} and Δk) (COI. 2016).

The temptations of using chemical analyses to authenticate VOO according to their geographical and botanical origins are well-abundant (Table 1). In this context, Houlali et al. (2014) differentiated between VOO samples collected from different altitudes using FA and PV parameters. Indeed, FA of VOO samples originating from the plain ($n = 41$) were significantly lower ($< 2\%$) than those collected from piedmont (FA = 2.6%) and mountain (FA = 2.7%) regions ($n = 101$). The PV showed an inverse trend since the highest values were observed for VOO belonging to plain (13.5 meq O_2/kg oil) *versus* 8.6 and 6.6 meq O_2/kg oil for those originating from the mountain and piedmont, respectively. Nevertheless, the obtained results are not in line with those of Tanouti et al. (2010) who reported that Moroccan VOO provided from high altitude orchards (+741 m) allowed to obtain VOO presenting low values of FA (0.12%) and high amount of PV (15.4%), while those originated from low altitude (342 m) exhibited higher and lower values of FA (0.62%) and PV (7 meq O_2/kg oil), respectively. The authors explained this trend by the fact that the high levels of antioxidant (Polyphenol and vitamin E) in VOO in

Table 1. A summary overview of chemical methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Investigated parameters	Objectives	Main results	Reference
VOO	24	FA, PV, k_{232}, k_{270}, & Δk and Follin-Ciocalteu (Phenolic compounds)	Characterisation of VOO of 6 varieties collected from 1 region during 1 crop season	Chemical parameters of VOO Moroccan varieties (Bouchouk Laghlid, Bouchouk Rguigue, Bakhboukh Beldi, Berri Meslal, Bouchouika and Moroccan Picholine) are influenced by agronomic and industrial factors.	(Boukachabine, Ajana, and El Antari 2011)
EVOO	298	FA, PV, k_{232}, k_{270}, Minguez-Mosquera (Pigments) and Follin-Ciocalteu (Phenolic compounds)	Characterisation of EVOO belonging to Moroccan Picholine variety and collected from 1 region (Meknes) during 4 crop seasons	The state of fruit before harvesting, as well as the agronomic and extraction modes affected significantly ($P < 0.05$) the chemical proprieties of EVOO of Moroccan Picholine collected from Meknes region.	(Bajoub et al. 2014)
VOO	20	FA, PV, k_{232}, k_{270}, and Δk	Characterisation of VOO according to agronomic factors (fertilization and irrigation)	The agronomic factors affected significantly ($P < 0.05$) FA since it ranged between 0.2 and 0.54 according to the conditions of fertilization and irrigation.	(Khlil, Mansouri, and Ben 2017)
EVOO	5	FA, PV, k_{232}, k_{270}, and Minguez-Mosquera (Pigments),	Effect of olive fruit fly infestation on the quality of Tunisian EVOO	-FA, PV, k_{232},and k_{270} values are significantly higher ($P < 0.05$) for EVOO of the affected olive fruits than those of non-affected ones. -Phenolic compounds are significantly lower ($P < 0.05$) in the EVOO of affected olive fruits.	(Mraicha et al. 2010)
VOO	27	Minguez-Mosquera (Pigments) and Follin-Ciocalteu (Phenolic compounds)	Influence of maturity index	-Chlorophylls, antioxidant activity, and polyphenols levels decreased with maturation since it passed, respectively, from 27.16 mg/kg, 92.4 DPPH*% and 2.07 mg/kg oil in September to 13.38 mg/kg, 74 DPPH*% and 0.37 mg/kg oil in the end of November.	(Zaringhalami et al. 2015)
EVOO	6	FA, PV, k_{232}, k_{270}, and Δk		-FA, PV, k_{232}, and k_{270} values increased with maturity since it passed from 0.24%, 2.34 meq O_2/kg oil, 1.44 and 0.08 during October to 0.46%, 4.30 meq O_2/kg oil, 1.66 and 0.13 in the end of December, respectively.	(Köseoğlu, Sevim, and Kadiroğ lu 2016)
VOO	10	FA, PV, k_{232}, k_{270}, Δk and Ollivier Denis (Phenolic compounds)	Effect of altitude	Moroccan Picholine variety samples originating from high altitude are richer in phenolic compounds (Tafoughalte: +839 m) compared to those belonging to low altitude (Taourirte: +269 m).	(Tanouti et al. 2010)
VOO	142	FA and PV		-Altitude had an effect on the chemical VOO samples. -FA of plain VOO are < 2%, while FA of piedmont and mountain samples are of 2.6% and 2.7%, respectively. -PV of plain VOO was of 13.5 meq O_2/kg oil, versus 8.6 and 6.6 meq O_2/kg oil for those originating from mountain and piedmont, respectively.	(Houlali et al. 2014)
EVOO	42	FA, PV, k_{232}, $k_{270}$and Follin-Ciocalteu (Phenolic compounds)		- Moroccan Picholine variety originating from high altitude were richer in phenolic compounds (+380 m) compared to those belonging to low altitude (+269 m).	(Boulfane et al. 2015)
VOO	8	FA, PV, k_{232} and k_{270}, and Δk	Effect of soil composition	VOO obtained from the olive trees cultivated on clay soils presented higher FA (0.34-0.41%) than those obtained from the trees of calcareous soils (0.22-0.32%).	(Essiari, Zouhair, and Chimi 2014)
EVOO and ROO	1 EVOO and 1 ROO	FA, PV, k_{232} and k_{270}	Evaluation of ROO and EVOO	- Chemical parameters of ROO were more affected by heat treatment than EVOO.	(Gharby et al. 2016)

(*continued*)

Table 1. Continued.

Product	Number of samples	Investigated parameters	Objectives	Main results	Reference
			resistance to heat treatment (110 °C)	- FA, PV, k_{232}, and k_{270} values of refined OO during heat treatment passed from 0.07%, 0.6 meq O_2/kg oil, 1.78 and 0.55 to 1.24%, 275.10 meq O_2/kg oil, 2.79 and 1.99, respectively. - FA, PV, k_{232}, and k_{270} values of EVOO during heat treatment passed from 0.64%, 2.3 meq O_2/kg oil, 1.57 and 0.13 to 0.82%, 32.43 meq O_2/kg oil, 2.59 and 0.33, respectively.	
EVOO	8	FA, PV, k_{232}, k_{270}, and Δk	Effect of harvesting system and fruit storage	- Harvesting system (Manual, hand-held combs, hand-held machine and straddle machine) and fruit storage time after harvesting induced some deterioration of the fruits due to the action of lipase. - Manual olive fruit harvesting appeared to be the best system mode since it gave better olive oil with low FA, PV, k_{232} and k_{270} Parameters. - FA passed from 0.23% to 3.23% for olive fruits stored for 1 week before extraction. - FA passed from 6.4 meq O_2/kg oil to 12.3 meq O_2/kg oil after 1 week of olive fruits storage.	(Famiani et al. 2020)
EVOO	24	FA, PV, k_{232}, k_{270}, Minguez-Mosquera (Pigments)	Influence of headspace oxygen and light exposure on the quality of EVOO during storage	-FA, PV, k_{232},and k_{270} increased significantly ($P < 0.05$) during exposure to sunlight since it passed from 0.32%, 6.05 meq O_2/kg oil, 1.4 and 0.12 to 0.99%, 21.80 meq O_2/kg oil, 2.52 and 0.32 , respectively, after 12 months of storage. -Oxygen headspace induced an increase in FA, PV, k_{232} and k_{270} after 12 months of storage. -At 21% of head-space oxygen, FA, PV, k_{232}, and k_{270} passed from 0.32%, 6.05 meq O_2/kg oil, 1.4 and 0.12 to 0.99%, 21.80 meq O_2/kg oil, 2.52 and 0.32, respectively, after 12 months of storage. -Pigment levels decreased significantly during storage and seemed to be affected by light exposure and the quantity of headspace oxygen.	(Iqdiam et al. 2020)

EVOO: Extra virgin olive oil; FA: Free acidity; OO: Olive oil; PV: Peroxide value; ROO: Refined olive oil; VOO: Virgin olive oil.

lower orchards limit their oxidation. By using chemical analyses, Bajoub et al. (2014) failed to differentiate between EVOO according to their geographical and botanical origins. Indeed, the authors depicted that the state of fruit before harvesting, as well as the agronomic and industrial parameters from harvesting to conditioning, are among the main factors that affect significantly ($P < 0.05$) the chemical properties. Indeed, Khlil, Mansouri, and Ben (2017) pointed out that the agronomic factors affected significantly ($P < 0.05$) FA, since it ranged between 0.2 and 0.54 according to the conditions of fertilization and irrigation. Furthermore, the soil composition influences the chemical composition of OO since Essiari, Zouhair, and Chimi (2014) showed, in their research study conducted during two crop seasons (2010–2011 and 2011–2012), that OO belonging to olive trees cultivated on clay soils presented higher FA ($0.35 - 0.41\%$) than those obtained from the trees of calcareous soils ($0.22 - 0.36\%$). In addition, the deterioration of the fruit before milling increase FA under the action of lipase (Famiani et al. 2020). Additionally, FA, PV, k_{232} and k_{270} values increased with maturity index since it passed from 0.24%, 2.34 meq O_2/kg oil, 1.44, and 0.08 during October to 0.46%, 4.30 meq O_2/kg oil, 1.66, and 0.13 at the end of December, respectively (Köseoğlu et al. 2016) .

Recently, Iqdiam et al. (2020) monitored the quality of VOO up to 12 months kept in different conditions (sunlight,

oxygen headspace). The authors depicted an increase of FA, PV, k_{232}, and k_{270} during storage (Table 1). The sunlight and headspace oxygen seemed to have a huge impact on the quality of VOO since EVOO stored in bottles with 2 to 5% headspace oxygen at room temperature (25–28 °C) presented FA, and PV values of 0.8% and 20 meq O_2/kg oil, respectively, even after 12 months of storage. Those kept with 21% headspace oxygen exceeded the FA and PV limits for EVOO category within 9 and 12 months, respectively. In a different approach, Gharby et al. (2016) pointed out the impact of heating (100 °C for 120 hours) on OO quality and found that after heating, the value of FA increased from 0.64% to 0.82% for VOO and from 0.07% to 1.24% for refined OO. The authors depicted that the higher content of active antioxidant compounds removed by refining could be the reason for better stability of VOO against hydrolysis during heat-treatment compared to refined OO.

Chlorophyll level was used as a reliable indicator for identifying the optimum harvesting period of olive fruit (Giuliani, Cerretani, and Cichelli 2011) (Table 1). In this context, Boulfane et al. (2015) depicted that the highest level of chlorophyll was observed during the beginning of the season (259 and 251 mg/kg oil) and decreased with the maturity index of olive fruits achieving 0.12 mg/kg oil at the end of the season confirming previous findings of Motilva and Romero (2010) who reported that total chlorophyll levels of Spanish VOO decreased with maturity passing from 392 mg/kg oil for green olives to 1.6 mg/kg oil for black ones. These findings were in line with those of Zaringhalami et al. (2015) who studied three monovarietal Iranian cultivars (Green, Mari, and Shenge) and found that the chlorophyll levels decreased from September (40 mg/kg oil) to November (3.5 mg/kg oil). An explanation could arise from the fact that during maturation, the chlorophyll compounds originally found in the fruit, are irreversibly converted into more stable pigments called pheophytins, and subsequently pyropheophytins which are the products of carboxy-methyl group as depicted by Bajoub et al. (2014). Although several authors indicated a decrease in the level of chlorophyll compounds during maturation, this indicator failed to differentiate between OO according to their geographical origin and variety. This could be due to the fact that the level of chlorophyll is impacted by agricultural conditions (Boulfane et al. 2015).

Several research studies have assessed the potential of phenolic compounds to determine the authenticity and quality of OO. For instance, Boulfane et al. (2015) depicted that VOO samples originating from the mountain region presented the highest values of total phenolic compounds (1100–1200 mg/kg oil) than those collected from plain areas (455-982 mg/kg oil). An explanation could arise from the fact that VOO belonging to trees grown in the regions with the low altitude and high temperature had the highest level of polyphenols, while those cultivated in areas with high altitude and low temperature exhibited the lowest values (Issaoui et al. 2010).

As observed for chlorophyll, the level of total phenolic compounds failed to authenticate OO according to their geographical origin (Boukachabine, Ajana, and El Antari 2011). These findings were, recently, confirmed by Famiani et al. (2020) who did not accomplish to discriminate between VOO according to their geographical origin and botanic cultivar. One explanation could be due to the fact that various factors such as the pedoclimatic conditions, the maturity index, and the extraction modes impact significantly the phenolic composition of OO. For example, it has been observed during 4 successive crop seasons of Meknes EVOO samples that the total phenol content of green fruits ranged between 216.83 and 668.67 mg/kg oil and decreased considerably with maturity index achieving 2.1 mg/kg oil for ripe fruits (Bajoub et al. 2014). These results were confirmed, later, by Boulfane et al. (2015) who depicted that olive oils produced by traditional mills exhibited the highest polyphenol content during July and August (301 to 380 mg/kg oil) and the lowest one in January (97 mg/kg oil).

Liquid chromatography analyses

Liquid chromatography (LC) coupled with mass spectroscopy (MS) is used to determine the quality and the authenticity of OO (Table 2). LC coupled with electrospray ionization time of flight and MS (LC-ESI-TOF-MS) was used to study 7 EVOO samples collected from central and southern Tunisia. By applying principal component analysis (PCA) to 22 phenolic compounds, a clear differentiation between EVOO according to their 7 geographical origins was observed (Ouni et al. 2011). To improve the efficiency of this method, Bajoub et al. (2015) have explored: i) LC-ESI-TOF-MS and ii) LC-ESI-tandem MS (LC-ESI-IT MS) to characterize and quantify the phenolic compounds of 261 VOO samples collected from 7 regions in Morocco. By utilizing a series of chemometric tools such as PCA and stepwise linear discriminant analysis (SLDA) on the five main phenolic groups named secoiridoids, phenolic alcohols, lignans, flavonoids, and phenolic acids, a clear separation of VOO samples according to their geographical origins was obtained. Indeed, the classification model of SLDA achieved up to 90% of accuracy. These results were confirmed, later, by Bajoub et al. (2016) who succeeded to discriminate between 37 PDO, 42 PGI, and 57 Moroccan VOO samples, since linear discriminant analysis (LDA) applied to the phenolic compounds allowed 100% of correct classification of samples according to their geographical origin.

In a similar approach, Vera et al. (2019) used high-performance liquid chromatography (HPLC) coupled to a charged aerosol detector to characterize the phenolic compounds of 65 Spanish EVOO. By applying a series of chemometric tools named PCA, Soft independent modeling of class analogy (SIMCA), and partial least square discriminant analysis (PLS-DA), the authors succeeded to discriminate between the EVOO according to their geographical origins. Bajoub et al. (2017) used LC-MS to quantify phenolic compounds. By applying LDA to phenolic compounds acquired on Arbequina (n = 11), Arbosana (n = 15), Cornicabra (n = 16), Frantoio (n = 21), Languedoc Picholine (n = 23), Moroccan Picholine (n = 31), and Picual (n = 23) EVOO

samples, the authors succeeded to discriminate EVOO samples according to their variety. Later, Monasterio et al. (2017) used phenolic profiles obtained by LC-MS to differentiate between 25 VOO samples of different Argentinian and European cultivars (Arauco, Arbequina, Picual, Frantoio, Changlot, Empeltre, Nevadillo, Manzanilla, and Coratina). By applying PCA, the authors succeeded to differentiate between the investigated samples since a clear differentiation between the samples was observed on the PCA similarity maps.

Green et al. (2020) used ultra-HPLC with charged aerosol detection to detect EVOO adulteration (Table 2). For instance, the authors succeeded to detect the presence of grape seed, soybean, canola, high-oleic sunflower and high-oleic safflower oils in EVOO samples with a level of adulteration ranging between 5 to 95% by applying PCA on triglycerides profile data.

Gas chromatography analyses

GC is one of the most common separation techniques used in food research to analyze volatile substances, aromas, and pesticides (Kamal and Karoui 2015). The fatty acid profile obtained by GC was used as an indicator to discriminate VOO samples according to their geographical origin. Indeed, the application of canonical discriminant analysis on fatty acid profiles allowed clear discrimination of 47 samples originating from four Greek regions (Karabagias et al. 2013) (Table 2).

Fatty acid profile was used as a marker for differentiating between OO according to the altitude of orchards. Msallam Al-Shdiefat (2019) reported that VOO belonging to the olive trees of high altitude are characterized by their lowest levels of oleic and linoleic acid compared to those collected from a lower altitude.

Sterols and volatile organic compounds were investigated to differentiate between OO according to their geographical origin (Table 2). For instance, by applying LDA to volatile organic compounds data sets, Pouliarekou et al. (2011) obtained a correct classification rate of only 57.4% regarding 47 Italian VOO originating from Zakynthos, Kefalonia, Lefkada, and Kerkyra. A better classification was obtained, later, by Vera et al. (2019) who succeeded, by applying PLS-DA on fatty acid profiles, to differentiate between 65 Spanish EVOO samples collected from three different regions with correct classification rates varying between 96 to 100%. The obtained results are in agreement with the findings of Lukić et al. (2019) who accomplished to discriminate 30 Croatian VOO samples originating from Istria and Dalmatia according to their geographical origin following the application of PCA and SLDA to 256 volatile organic compounds. By using flash GC electronic nose (FGC-EN) and solid-phase micro-extraction GC-MS (SPME/GC-MS), Melucci et al. (2016) achieved to discriminate between 132 samples labeled as Italian and 119 samples labeled as non-Italian (Spanish and Maghrebian). These findings were, recently, confirmed by Cecchi et al. (2020) who, by applying LDA on volatile organic compounds data sets, succeeded to differentiate between 1217 EVOO according to their geographical origin (Spain, Italy, Greece, Tunisia, Portugal, Peru, Morocco, Australia, Albania, California, and Argentina) with 94.5% of correct classification.

Blasi, Pollini, and Cossignani (2019) pointed out that the application of LDA on triglycerides and volatile organic compounds obtained by GC-MS allowed to differentiate 16 EVOO samples originated from four Italian varieties (Dolce Agogia, Frantoio, Leccino, and Moraiolo) with 100% of correct classification (Table 2). However, the obtained results should be validated in a large number of samples. Recently, Maléchaux et al. (2020) applied PLS-DA to the GC data obtained from Chemlali (n = 187), Chetoui (n = 102), and Oueslati (n = 45) VOO samples and found 98% of correct classification of VOO samples according to their cultivars confirming previous findings of Essiari, Zouhair, and Chimi (2014) and Lukić et al. (2019) who succeeded, by applying PCA and SLDA to volatile organic compounds, to discriminate Croatian VOO samples belonging to five varieties (Buža, Istarska bjelica, Rosinjola, Oblica, and Lastovka).

Spectroscopic techniques used for the determination of the authenticity and the quality of olive oils

In the last decade, spectroscopic techniques in the different electromagnetic radiation spectrum (Ultraviolet-visible, infrared, nuclear magnetic resonance, fluorescence, and so on) have become more and more used in food research for determining the quality and authenticity of food products including OO. These techniques are fast, low-cost, and being most of them nondestructive (Karoui, Dufour, and De Baerdemaeker 2006b, Hassoun and Karoui 2017).

Infrared spectroscopy

The infrared radiations are absorbed by matter thanks to molecular vibrations that are separated by energies linked to the infrared part of the electromagnetic radiation (Casale and Simonetti 2014). The infrared part of electromagnetic radiation is divided into three areas: the near infrared (NIR) which extends from 14000 to 4000 cm^{-1} (0.7-2.5 µm in wavelengths); the mid infrared (MIR) which ranges from 4000 to 400 cm^{-1} (2.5-25 µm); and the far infrared covering the spectral range from 400 to 10 cm^{-1} (25-1000 µm). The principle of infrared spectroscopy is based on the implementation of the infrared radiation interaction with a sample, then the detection and spectral analysis (by transmission or by reflection) of this radiation since each frequency designates a specific molecular bond (C-H, N-H, O-H and C– N, …) (Kamal and Karoui 2015).

3.1.1. Near infrared spectroscopy

The response of the molecular bonds O-H, C-H, C-O, and N-H can be characterized by the NIR region of the electromagnetic spectrum. For each wavelength, the part of the radiation absorbed by the sample is measured and

Table 2. A summary overview of chromatographic methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
EVOO	22	LC-MS (Phenolic compounds)	PCA and HCA	Geographical authentication of EVOO according to 8 Tunisian regions	Phenolic fraction could be used as marker since it allowed clear differentiation of EVOO samples.	(Ouni et al. 2011)
VOO	47	GC (FATs)	CDA	Geographical authentication of VOO according to 4 Greek regions	FATs profiles could be used as geographical markers	(Karabagias et al. 2013)
VOO	136	LC-MS (Phenolic compounds)	PCA and LDA	Geographical authentication of VOO according to 3 Moroccan regions	Phenolic compounds could be used as geographical markers since 100% of correct classification of VOO.	(Bajoub et al. 2016)
EVOO	278	Flash GC and SPME/GC-MS (VOC)	PCA and PLS-DA	Geographical authentication of EVOO according to 3 Italian origins	VOC could be used as geographical markers of EVOO since R^2 of 0.91 was observed.	(Melucci et al. 2016)
EVOO	65	HPLC (Phenolic compounds), GC (FATs)	PCA, SIMCA and PLS-DA	Geographical authentication of EVOO according to 3 Spanish regions	Phenolic compounds and FATs could be used as markers of VOO geographical origin since 96 to 100% of correct classification was observed.	(Vera et al. 2019)
VOO	30	GC (VOC)	PCA and SLDA	-Geographical authentication of VOO according to 2 Croatian regions -Authentication of 5 varieties (Buža, Istarska bjelica, Rosinjola, Oblica and Lastovka).	VOC could be used as indicator to determine geographical origin and variety since SLDA allowed clear differentiation of VOO samples.	(Lukić et al. 2019)
EVOO	1217	Head-space SPME/GC-MS (VOC)	ANOVA and LDA	Geographical authentication of EVOO according to 9 Mediterranean and 1 Argentinean regions	VOC could be used as marker for EVOO authentication since 94.5% of correct classification was obtained.	(Cecchi et al. 2020)
VOO	142	GC (FATs)	PCA	Authentication of VOO according to their altitude	FATs composition could be used as altitude marker since a clear differentiation of samples was observed.	(Houlali et al. 2014)
VOO	12	GC (FATs)	not mentioned		VOO samples collected from tree of high altitude were characterized by long shelf life compared to those of low altitude.	(Msallam Al-Shdiefat 2019)
VOO	51	GC (VOC)	PCA and LDA	-Geographical authentication: 4 Greek regions -Authentication of 7 varieties (Koroneiki, Ntopia of Zakynthos, Thiaki, Mouzolia, Asprolia, and Lianolia)	-VOC could not be used with success as tool to determine geographical and cultivar origin since 57.4 and 52% of correct classification, respectively was obtained.	(Pouliarekou et al. 2011)
VOO	24	GC (FATs) and HPLC (Triglycerides)	ANOVA	Authentication of 7 varieties (Bouchouk Laghlid, Bouchouk Rguigue, Bakhboukh Beldi, Berri Meslal, Bouchouika and	-Significant difference ($P < 0.05$) of OO according to their variety was obtained. -Oleic acid of OO was found to be higher in Moroccan Picholine compared to Bouchouika variety.	(Boukachabine, Ajana, and El Antari 2011)

(*continued*)

Table 2. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
				Moroccan Picholine)		
EVOO	25	LC-MS (Phenolic compounds)	PCA	Authentication of 9 varieties (Arauco, Arbequina, Picual, Frantoio, Changlot, Empeltre, Nevadillo, Manzanilla, and Coratina)	Phenolic compounds could be used as a valuable indicator of the varietal origin.	(Monasterio et al. 2017)
EVOO	203	HPLC (Phenolic compounds)	PCA, k-NN and PLS-DA	Authentication of 9 varieties (Arauco, Arbequina, Picual, Frantoio, Changlot, Empeltre, Nevadillo, Manzanilla, and Coratina).	A discriminant approach based on phenolic profile was considered as a promising and efficient tool for differentiating EVOO according to their varietal origin with 99% of correct classification.	(Bajoub et al. 2017)
EVOO	60	GC-MS (VOC)	LDA	Authentication of 4 varieties (Dolce Agogia, Frantoio, Leccino, and Moraiolo)	VOC could be used as varietal origin markers with 100% of correct classification.	(Blasi, Pollini, and Cossignani 2019)
EVOO	234	GC (FATs)	PLS-DA	Varietal authentication : 3 varieties (Chemlali, Chetoui and Oueslati)	FATs profiles could be used as varietal markers since 98% of correct classification of EVOO was obtained.	(Maléchaux et al. 2020)
EVOO	140	Ultra-HPLC (Triglycerides)	PCA	Detection of the adulteration of EVOO with grape seed, soybean, canola, high-oleic sunflower and high-oleic safflower oils with level of adulteration ranging between 5 to 95%	Triglyceride profiling could be used to detect the adulteration of EVOO by grape seed, soybean, canola, high-oleic sunflower and high-oleic safflower oils	(Green et al. 2020)

ANOVA: Analysis of variance; CDA: Canonical discriminant analysis; EVOO: Extra virgin olive oil; FATs: Fatty acids; GC: Gas chromatography; HCA: Hierarchical cluster analysis; HPLC: High performance liquid chromatography; HS-SPME: Head space solid phase microextraction; k-NN: K-nearest neighbors; LC-MS: Liquid chromatography-Mass spectroscopy; LDA: Linear discriminant analysis; OO: Olive oil; PCA: Principal component analysis; PLS-DA: partial least squares discriminant analysis; PLSR: Partial least squares regression; SIFT-MS: Selected ion flow tube mass spectrometry; SIMCA: Soft independent modeling of class analogy; SLDA: Stepwise linear discriminant analysis; VOC: Volatile organic compounds; VOO: Virgin olive oil.

constitutes the spectrum which can be considered as a global fingerprint (Casale and Simonetti 2014). This technique is well-used in the authentication of food products. For example, NIR spectroscopy was utilized for: i) controlling the quality of potato chips (Shiroma and Rodriguez-Saona 2009); ii) authenticating fish filet (Alamprese and Casiraghi 2015), iii) differentiate between soft cheeses according to their manufacturing process and sampling zone (Karoui et al. 2006f), iv) Classification of soil texture classes (Mouazen et al. 2005), and so on.

Regarding OO, Karunathilaka et al. (2016) accomplished, by applying a series of chemometric tools named standard normal variate (SNV), PCA, and SIMCA on NIR spectra (6200-4600 cm^{-1}) acquired on 88 OO samples of different brand quality (62 EVOO and 26 non-EVOO), to classify them according to their quality with an accuracy of 100% (Table 3). These findings are in line with those of Yan, Stuijvenberg, and Ruth (2019) who reported that by applying PCA to NIR data sets recorded in the 1350–1570 nm, a clear separation between 80 EVOO, 40 refined OO and 10 pomace OO samples was observed.

Houlali et al. (2014) assessed the potential of NIR (10000–4500 cm^{-1}) to differentiate 200 VOO samples originating from Marrakech and Meknes in Morocco. At the naked eye, the spectra were found to be very similar and very difficult to be distinguishable from each others. Thus, the authors applied PCA allowing a clear discrimination of samples according to their geographical origin, in agreement with previous findings of Casale et al. (2012) who succeeded to differentiate between 23 PDO *Chianti Classico* VOO samples and 34 samples produced from the same olive varieties but under different pedoclimatic conditions. By applying PCA, Unequal class models (UNEQ), SIMCA, and partial least square regression (PLSR) to the NIR spectra in the

Table 3. A summary overview of near and mid-infrared spectroscopy methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
EVOO and non-EVOO	62 EVOO and 26 non-EVOO	NIR 6200-4600 cm^{-1}	PCA, SNV and SIMCA	Classification of OO according to their class	100% of correct classification of samples was observed according to their class.	(Karunathilaka et al. 2016)
EVOO, ROO, and POO	80 EVOO, 40 ROO, and 10 POO	NIR 1350-1570 nm	PCA		PCA similarity maps allowed clear differentiation of samples according to their class.	(Yan, Stuijvenberg, and Ruth 2019)
EVOO	140	NIR 800 and 1075 nm	DOSC-GA-PLS	Geographical authentication of EVOO originating from 1 Spanish, 1 Turkish and 1 Italian regions	EVOO samples were well discriminated since R^2 of 0.987 was observed.	(Lin, Chen, and He 2012)
EVOO	57	NIR 10000-4000 cm^{-1}	PCA, UNEQ and SIMCA	Geographical authentication of EVOO according to 3 Italian regions	VOO were clearly discriminated on the maps according to their geographical origins.	(Casale et al. 2012)
EVOO	910	NIR 1100–2498 nm	PLS-DA and SVM	Geographical authentication of Ligurian and non-Ligurian samples (Italy)	Good classification of OO according to their geographical origin with 85.1% and 82.2% of correct classification by applying SVM and PLS-DA, respectively.	(Devos, Downey, and Duponchel 2014)
VOO	142	NIR 10000-4500 cm^{-1}	PCA	Authentication of VOO according to their altitude	Samples are not clearly discriminated according to the altitude since great overlapping was observed.	(Houlali et al. 2014)
EVOO	247	NIR 10000-4500 cm^{-1}	PCA, SNV and SIMCA	Authentication of 6 Tunisian varieties (Chemlal, Zarrazi, Chemchali, Oueslati and Zalmati)	The NIR succeeded to separate samples according to their variety since 98.5% of correct classification was observed.	(Laroussi-Mezghani et al. 2015)
EVOO	63	NIR 10000-4400 cm^{-1}	PLSR and PCR	Detection of the adulteration of EVOO by mild deodorized and ROO with level ranging between 2.5 and 75%	Excellent detection of EVOO adulteration since R^2 of 0.97 and 0.98 was obtained respectively with PLSR and PCR.	(Wójcicki et al. 2015)
EVOO	10	NIR 12000-4000 cm^{-1}	PCA	Detection of the adulteration of EVOO by corn, sunflower, soybean, and canola oils with level of adulteration ranging between 10 and 20%	Clear differentiation of samples was observed.	(Vanstone et al. 2018)
EVOO	40	NIR 12000-4000 cm^{-1}	BSS-PLS	Detection of the adulteration of EVOO by canola and sunflower oils with level of	Excellent detection of adulteration of EVOO since R^2 of 0.99 and RMSEP	(Jiang and Chen 2019)

(*continued*)

Table 3. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
				adulteration ranging between 2 and 50%	of 1.44% were observed.	
VOO	54	NIR 12000-4000 cm^{-1}	2D-COS and k-NN	Detection of the adulteration of EVOO by canola, soybean and corn oils with level of adulteration ranging between 5 and 95%	Detection of canola, soybean, and corn oils in EVOO with 85 % accuracy.	(Sohng et al. 2020)
EVOO	478	Visible-NIR 400-2500 nm	PLSR	Prediction of FA, PV, k_{232}, and k_{270} of EVOO	-Excellent prediction of PV with R^2 of 0.97. -Approximate prediction of FA, k_{232} and $k_{270}$with R^2 of 0.68, 0.69 and 0.62, respectively.	(Garrido-Varo et al. 2017)
VOO	73	NIR 12000-4000 cm^{-1}	PLSR	Prediction of tocopherols and FATs of VOO	-Good prediction of tocopherols with R^2 of 0.83. -Excellent prediction of FATs with R^2 of 0.99.	(Özdemir et al. 2018a)
EVOO, VOO and non-VOO	56 EVOO, 5 VOO and 40 non-VOO	NIR 1000-1800 nm	PLSR	Prediction of FATs of EVOO, VOO, and non-VOO	Excellent prediction of FATs with R^2 of 0.95.	(Cayuela-Sánchez et al. 2019)
EVOO	5000	NIR 11500-4000 cm^{-1}	PLSR	Prediction of FA, PV, k_{232} and, k_{270} of EVOO	Excellent prediction of FA, PV, k_{232} and k_{270} with R^2 of 0.99, 0.92, 0.9 and 0.9, respectively.	(Willenberg, Matthäus, and Gertz 2019)
EVOO	57	MIR 4000 – 700 cm^{-1}	PCA, UNEQ, SIMCA and PLSR	Geographical authentication of EVOO according to 3 Italian regions	EVOO are clearly discriminated according to their geographical origins.	(Casale et al. 2012)
EVOO	910	MIR 4000 – 600 cm^{-1}	PLS-DA and SVM	Geographical authentication of Ligurian and non-Ligurian samples	Good classification of samples according to their geographical origin with 82.7% and 78.2% of correct classification by applying SVM and PLS-DA, respectively.	(Devos, Downey, and Duponchel 2014)
VOO	41	MIR 4000 – 700 cm^{-1}	PCA, FDA and PLSR	-Geographical authentication of EVOO according to 5 Moroccan regions -Varietal authentication of EVOO according to 4 cultivars P-rediction of FA, PV, k_{232} and k_{270} parameters and chlorophyll level	-Good discrimination of VOO samples according to their geographical origin and variety with 91.87% and 91.87% of correct classification, respectively. -FA, PV, k_{232}, k_{270} and chlorophyll level of VOO samples are predicted with of R^2 of 0.99, 0.97, 0.84, 0.98 and 0.93, respectively.	(Zaroual et al. 2020)
EVOO	112	MIR 4000 – 400 cm^{-1}	LDA and SIMCA	Authentication of 3 Italian varieties (Leccino, Casaliva and Frantoio)	Excellent discrimination of samples with 98% and 100% of correct	(Sinelli et al. 2010a)

(*continued*)

Table 3. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
					classification according to their variety.	
EVOO	27	MIR $4000-500\,cm^{-1}$	PLSR and PCR	Detection of the adulteration of EVOO by palm oil with level ranging between 1 and 50%	Pure EVOO samples are clearly discriminated from the adulterated ones with R^2=0.99.	(Rohman and Man 2010)
EVOO	104	MIR $4000-600\,cm^{-1}$	PLSR	Detection of the adulteration of EVOO by sunflower oil with level ranging between 1 and 24 %	Excellent detection of the adulteration of EVOO samples by sunflower oil with less than 2% prediction error and R^2 of 0.99.	(Oussama et al. 2012)
EVOO	63	MIR $4000-650\,cm^{-1}$	PLSR and PCR	Detection of the adulteration of EVOO by mild deodorized and ROO with level ranging between 2.5 and 75%	Excellent detection of EVOO adulteration since R^2 of 0.99 and 0.98 was observed, respectively, with PLSR and PCR.	(Wójcicki et al. 2015)
VOO	54	MIR $4000-700\,cm^{-1}$	PLSR	Prediction of water content, phenol levels and antioxidant activity	-The prediction of water content and phenol levels were good (R^2=0.87). -Approximate discrimination was observed for antioxidant activity (R^2=0.63).	(Cerretani et al. 2010)
VOO	64	MIR $4000-650\,cm^{-1}$	PLSR	Prediction of oxidative stability, chlorophyll, some major FATs and total phenolic contents	Excellent prediction for all the parameters: oxidative stability (R^2=0.99), chlorophyll content (R^2=0.98), some major fatty acids named palmitic (R^2=0.87), oleic (R^2=0.94), linoleic (R^2=0.97), saturated (R^2=0.91), mono-unsaturated (R^2=0.94) and poly-unsaturated (R^2=0.97), hydroxytyrosol as a phenolic compound (R^2=0.97) and total phenolic content (R^2=0.99)	(Uncu and Ozen 2015)
VOO	146	MIR $4000-600\,cm^{-1}$	PLS-DA	Prediction of 4 main sensorial defects: musty, winey, fusty and rancid	-Good prediction of musty defect with predictive ability around 87 %. -Approximate determination of winey, fusty and rancid defects since only 77% of correct classification.	(Borràs et al. 2015)

(*continued*)

Table 3. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
VOO	343	MIR $4000-600\,cm^{-1}$	PLS-DA	Prediction of 10 sensory attributes: 6 positive (fruity, bitter, pungent, grassy, sweet and astringent) and 4 off-flavors (musty, fusty, winey-vinegary and rancid)	-MIR failed to predict fruity (R^2=0.42), bitter (R^2=0.50), pungent (R^2=0.24), grassy (R^2=0.36), sweet (R^2=0.28) and astringent (R^2=0.35) attributes and fusty (R^2=0.35), winey (R^2=0.39) and rancid (R^2=0.28) defects. -Approximate prediction of musty defect was observed (R^2 =0.64).	(Borras et al. 2016)

2D-COS: Two-dimension correlation; BSS-PLS: Bootstrapping soft shrinkage-*partial least squares; DOSC-GA-PLS: direct orthogonal signal correction-genetic algorithms-partial least squares; EVOO: Extra virgin olive oil; FATs: Fatty acids; FDA: Factorial discriminant analysis; k-NN: K-nearest neighbors; LDA: Linear discriminant analysis; MIR: Mid infrared spectroscopy; NIR: Near infrared spectroscopy; OO: Olive oil; PCA: Principal component analysis; PCR: Principal component regression; PLS-DA: Partial least squares discriminant analysis; PLSR: Partial least squares regression; SIMCA: Soft independent modeling of class analogy; SNV: Standard normal variate; SVM: Support victor machine; VOO: Virgin olive oil; UNEQ: Unequal class models.*

10000–4000 cm^{-1}, the authors accomplished to differentiate between samples since 100% of correct classification was obtained by applying UNEQ. These findings were in line with Jiménez-Carvelo, Lozano, and Olivieri (2019) who succeeded to differentiate 65 EVOO as a function of their geographical origins. Indeed, by applying PLS-DA on NIR spectra data, 100% of correct classification was achieved, confirming previous findings of Lin, Chen, and He (2012). The latter succeeded to authenticate 140 EVOO according to their country (Spain, Turkey and Italy) by applying direct orthogonal signal correction-genetic algorithms-PLS (DOSC-GA-PLS) on visible-NIR spectra (325-1075 nm) since R^2 value of 0.98 was observed (Table 3). However, some studies revealed that NIR did not succeed to discriminate between: i) Maghrebian and French VOO samples according to their geographical origin since some samples originating from Tunisia, Morocco, Algeria and France were overlapped (Laroussi-Mezghani et al. 2015); and ii) Moroccan VOO according to the orchards altitude since great overlapping of 142 VOO was observed (Houlali et al. 2014). Contrary to the previous study, Laroussi-Mezghani et al. (2015) succeeded by using NIR (10000–4500 cm^{-1} spectral range) coupled with PCA to separate correctly 133 Tunisian VOO samples according to their six cultivars named Chemlali Sfax (n = 63), Chetoui (n = 41), Zarrazi (n = 5), Chemchali (n = 5), Oueslati (n = 14) and Zalmati (n = 5).

Reported studies on the use of NIR spectroscopy to detect adulteration were, recently, extensively explored. Wójcicki et al. (2015) studied the spectral region ranged between 12500-4000 cm^{-1} of pure EVOO (n = 3), EVOO mixed with mild deodorized (n = 30), and refined OO (n = 30) at different levels of adulteration (2.5 to 75% with 2.5% step). The authors depicted satisfactory results since R^2 of 0.97 and 0.98 was observed, respectively, by applying PLSR and principal component analysis (PCR) on NIR spectra data. These findings are in line with those of: i) Vanstone et al. (2018) who pointed out that applying PCA to NIR data set (12000-4000 cm^{-1}) yielded a better separation between pure EVOO and the adulterated samples with canola oil at 20% of adulteration; and ii) Jiang and Chen (2019) who succeeded to detect the adulteration of EVOO with sunflower and canola oil at levels of adulteration ranging between 2 and 50% by applying bootstrapping soft shrinkage PLS modeling. Indeed, the authors found R^2 and root mean square prediction error (RMSEP) of, respectively, 0.99 and 1.44%. These findings are supported, recently, by Sohng et al. (2020) who succeeded to detect the adulteration of VOO samples by canola, soybean, and corn oils at 5% with 95% accuracy (Table 3).

Some research studies attempted to use NIR to predict fatty acid levels and minor components such as tocopherols. Indeed, 73 EVOO samples obtained from 21 different cultivars and 4 geographical regions were analyzed by NIR (12000-4000 cm^{-1}), HPLC and GC-MS (Özdemir et al. 2018b). By applying PLSR to the data tables, values of R^2 of 0.83 and 0.99 were obtained for tocopherols and fatty acids, respectively. The obtained results are, later, confirmed by Cayuela-Sánchez et al. (2019) who by applying NIR in the 1000–1800 nm spectral range on 56 EVOO, 5 VOO and 40 non-VOO succeeded to predict fatty acid levels with R^2 ranging between 0.9 and 0.95 (Table 3). In a research study conducted on a large number of EVOO (n = 478), Garrido-Varo et al. (2017) accomplished to obtain an excellent and approximate prediction for some chemical parameters named FA, PV, k_{232} and k_{270} with R^2 of 0.97, 0.68, 0.69 and 0.62 was observed, respectively. These findings are in line with those of Willenberg, Matthäus, and Gertz (2019) who obtained excellent prediction of FA, PV, k_{232} and k_{270}

parameters with R^2 of 0.99, 0.92, 0.9 and 0.9, respectively was observed.

3.1.2. Mid Infrared spectroscopy

Several papers were published in the last decade regarding the effectiveness of MIR spectroscopy to authenticate food products (Valli et al. 2016) (Table 3). For instance, the technique was utilized to: i) authenticate fish products (Karoui et al. 2007b, Boughattas, Le Fur, and Karoui 2020a); ii) monitor the quality of pound cakes during storage (Nhouchi and Karoui 2018), iii) differentiate sheep milk according to feeding system (Karoui et al. 2011), iv) predict some physico-chemical parameters of Emmental cheeses (Karoui et al. 2006e), and so on.

Regarding OO, MIR spectroscopy was used to classify Moroccan olive fruits according to their geographical origin and variety (Table 3). Indeed, by applying a series of chemometric tools named PLS-DA and PCA, De Luca et al. (2011) succeeded to separate 29 Moroccan Picholine VOO originating from four different areas of central Morocco (Meknes, Brida, Fqih Bensaleh and Ksiba) since R^2 and RMSEP of 0.99 and 0.049 was observed, respectively. These results arre in agreement with those of Devos, Downey, and Duponchel (2014) who by applying PLS-DA and genetic algorithm for the simultaneous optimization of support vector machine (SVM) on MIR spectra ($4000-600\,cm^{-1}$) on 210 Ligurian and 700 non-Ligurian EVOO noted 82.7% and 78.6% of correct classification, respectively. These results were confirmed, recently, by Zaroual et al. (2020) who succeeded to discriminate 41 Moroccan VOO according to their geographical origin (Fez/Meknes, Marrakech/Safi, Northern, Western and Beni-Mellel/Khenifra) and variety (Moroccan Picholine, Languedoc Picholine, Arbosana and Arbequina) since 91.78 and 91.78% of correct classification was obtained. These results are in line with those of Sinelli et al. (2010a) who succeeded to discriminate EVOO according to their botanic cultivars since 94.2% and 86.6% of correct classification for the calibration and prediction was obtained, respectively. The authors stated that the best classification was observed for Leccino cultivar compared to Casaliva and Frantoio cultivars (Table 3).

In a different approach, MIR spectroscopy was used to detect the adulteration of EVOO by other oils of lower quality. For instance, Oussama et al. (2012) applied PLSR on MIR spectra ($4000-600\,cm^{-1}$) acquired on 104 EVOO samples and succeeded to detect the presence of sunflower and soybean oil in EVOO at a level of 2% with R^2 of 0.99 (Table 3). These findings are in agreement with the previous findings of: i) Rohman and Man (2010) who accomplished to detect the adulteration of EVOO with palm and rice bran oil with levels of adulteration ranging between 1 and 50%; and ii) Wójcicki et al. (2015) who achieved to detect the adulteration of EVOO by deodorized OO with 96% accuracy by applying PCR on MIR spectra obtained in the 4000-650 cm^{-1}.

Regarding the ability of MIR to predict some chemical parameters, Uncu and Ozen (2015) succeeded to determine the oxidative stability (R^2=0.99), chlorophyll content (R^2=0.98), some major fatty acids named palmitic (R^2=0.87), oleic (R^2=0.94), linoleic (R^2=0.97), saturated (R^2=0.91), mono-unsaturated (R^2=0.94), and poly-unsaturated (R^2=0.97), hydroxytyrosol as a phenolic compound (R^2=0.97) and total phenolic content (R^2=0.99) of 64 Turkish VOO. These findings revealed the ability of MIR to be used as a screening method for a rapid determination of some chemical parameters of OO.

Limited studies were focused on the determination of sensory properties of OO from MIR. Borràs et al. (2015) found satisfactory results for the prediction of the musty defect with predictive ability around 87%; while the MIR failed to determine winey, fusty and rancid defects since only 77% of correct classification was observed. The same research group tried to predict sensory properties of 343 VOO by applying PLS-DA on MIR spectra scanned in the spectral range 4000- 600 cm^{-1}. Nevertheless, the authors failed to predict with high accuracy the fruity (R^2=0.42), bitter (R^2=0.50), pungent (R^2=0.24), grassy (R^2=0.36), sweet (R^2=0.28), and astringent (R^2=0.35) attributes and fusty (R^2=0.35), winey (R^2=0.39), rancid (R^2=0.28), and musty (R^2=0.64) defects (Borràs et al. 2016).

Fluorescence spectroscopy

Fluorescence spectroscopy is the electromagnetic analytical technique that measures the fluorescence of a sample based on the principle of the interaction between light and matter (Andueza et al. 2015). Fluorescence spectroscopy is one of the techniques that is continuously growing, due to its high specificity, convenience, quick response, being nondestructive, noninvasive, and cost-effective (Karoui, Cartaud, and Dufour 2006a, Karoui, Dufour, and De Baerdemaeker 2007a). Indeed, front-face fluorescence spectroscopy was used to authenticate different food products. For instance, the technique has shown its ability to: i) monitor the freshness of fish (Hassoun and Karoui 2016, Boughattas et al. 2020b) and egg (Karoui et al. 2006c, 2006d, 2007c, Karoui, Nicolaï, and de Baerdemaeker 2008, Karoui and Blecker 2011); ii) authenticate tuna species (Boughattas, Le Fur, and Karoui 2019); iii) monitor milk coagulation (Blecker, Habib-Jiwan, and Karoui 2012), and so on.

Regarding OO, Jiménez-Carvelo, Lozano, and Olivieri (2019) succeeded to differentiate 65 EVOO according to their eight geographical origins by applying PCA and PLS-DA to fluorescence excitation–emission matrix, since 100% of correct classification was observed. One of the main conclusions of this study was that fluorescence spectroscopy could be used as a rapid screening method for the determination of the authenticity of EVOO. These findings are in line with those of: i) Lia et al. (2020) who succeeded to authenticate 65 Maltese EVOO according to their geographical regions since 73, 80 and 93% of correct classification by applying LDA, PARAFAC and discriminant multi-way partial least squares regression (DN-PLSR), respectively, were obtained; ii) Al Riza et al. (2021) who by applying SVM on 87 EVOO samples obtained 94 and 90% of accuracy according to their geographical origin and variety, respectively; and

iii) Zaroual, El Hadrami and Karoui (2020) who achieved to discriminate 41 Moroccan VOO according to their geographic origin and variety with 96.72 and 95.12% of correct classification.

By using the same approach, Dankowska, Małecka, and Kowalewski (2013) succeeded to discriminate OO according to their grades. Indeed, the application of successive projection algorithm (SPA) and LDA method to synchronous fluorescence spectra allowed the classification of 36 EVOO, 12 VOO and 12 pomace OO according to their quality with low classification error ranging between 0.9 and 6.4%. These results are, later, confirmed by: i) Mabood et al. (2016) who, by applying PLSR to the synchronous fluorescence spectra, succeeded to detect the presence of pomace olive oil (2 to 20%) in EVOO with R^2 of 0.94; and ii) Wójcicki et al. (2015) who detected, by applying PLSR and PCR to the excitation-emission fluorescence spectroscopy, the presence of mild deodorized and refined OO at different levels of adulteration with R^2 of 0.98 and 0.98, respectively (Table 4). Recently, Li et al. (2020) applied PCA, LDA and PLSR to laser-induced fluorescence spectra in the aim to detect the presence of soybean oil in EVOO at a level of 2%. The authors stated that heating treatment before scanning spectra enhance the efficiency of detecting adulteration since R^2 of 0.92 and RMSEP of 0.27% were observed. These results confirmed previous findings of Milanez et al. (2017) who succeeded to detect soybean oil in EVOO at 1.33% with R^2 of 0.98 and Durán Merás et al. (2018) who detected the presence of pomace OO in EVOO since 99% of accuracy is obtained.

In a different approach, Guzmán et al. (2015) accomplished to predict PV, k_{232}, k_{270} and FA by applying PLSR and N-PLS to fluorescence spectra. Indeed, R^2 of 0.93, 0.96, 0.90 and 0.98 were observed for FA, PV, k_{232} and k_{270}, respectively. In the same context, Squeo et al. (2019) succeeded to predict phenolic level by applying PLSR to fluorescence spectra since R^2 of 0.95 was observed. One of the main conclusions of this study was the ability to use fluorescence spectroscopy as a rapid technique for the determination of chemical parameters of OO.

Raman spectroscopy

Raman spectroscopy is used to determine vibrational modes of molecules, although rotational and other low-frequency modes of systems may also be observed. Raman spectroscopy is commonly used in chemistry to provide a structural fingerprint by which molecules can be identified. The technique was used as a tool for the authentication of edible oils. For instance, Portarena, Baldacchini, and Brugnoli (2017) succeeded to discriminate EVOO samples originating from 7 Italian regions (Apulia, Latium, Ligury, Molise, Sardinia, Siciliy, Tuscany) according to their geographical origins. The application of LDA to Raman spectra scanned in the 1800-900 cm^{-1} spectral range allowed classification rate of 82%, in line with the findings of Sánchez-López et al. (2016) who succeeded to authenticate: i) 412 EVOO samples according to their geographical origin (Jaen, Cadiz, Cordoba, Malaga, Granada and Seville) with 89% of correct classification; and ii) botanic cultivar (Frantoio, Hojiblanca, Picual and Gorda) with 84% of good classification. Some misclassifications were observed between Hojiblanca and Picual varieties which were ascribed by the authors to the botanic similarities between these two cultivars (Table 5).

In a different approach, Raman spectroscopy (1800-900 cm^{-1}) was used to detect OO adulteration with other oils. Georgouli, Martinez Del Rincon, and Koidis (2017) detected the presence of hazelnut oil in EVOO samples at different levels (1-90%) and obtained correct classification rate of 82% by applying continuous locality preserving projections (CLPP) coupled with k-NN; less interesting results were obtained when the authors coupled CLPP, PCA, SIMCA, LDA, PLS-DA and PLSR since correct classification rates of ranging between 24 and 66% were obtained, respectively. In a similar approach, 96 VOO samples adulterated with waste cooking oil (2.5%, 5%, 10%, 20%, 30% and 50%) were scanned by Raman spectroscopy (Li et al. 2018); following the application of SNV processing, the authors succeeded to detect the adulteration of OO with validation R^2 of 0.98 and an RMSEP of 0.05% demonstrating the ability of this technique to be used as a rapid tool for the determination of OO quality.

Ultraviolet and visible spectroscopy

UV-Vis spectroscopy is an electronic spectroscopy technique involving photons where wavelengths are in the ultraviolet (100 nm − 400 nm) and visible (400 nm − 800 nm) range.

Pizarro et al. (2013) succeeded to discriminate 40 EVOO samples originating from Catalonia, Andalusia and La Rioja by applying LDA (75% of correct classification) and PLS-DA (90% of correct classification) to the UV-Vis spectra scanned in the 190-1090 nm spectral range. The obtained findings are in agreement with those of Casale et al. (2012) who depicted the ability of UV-Vis technique to authenticate 57 PDO EVOO since correct classification amounting to 97.5% and 100% was observed by using SIMCA and UNEQ, respectively (Table 5).

Recently, Ferreiro-González et al. (2017) applied PCA and LDA on 118 OO samples belonging to different brand quality (81 VOO, 10 refined OO and 17 pomace OO) by scanning UV-Vis spectra in the 380-730 nm spectral range. By applying PCA to the spectra, the authors pointed out a clear differentiation between samples. The obtained results were, recently, confirmed by Aroca-Santos et al. (2019) who used the UV-Vis to detect adulteration of 141 pure EVOO samples with soybean and sweet almond oils (20 of each). By applying PLSR and multilayer perceptron (MLP) to the UV-Vis spectra, EVOO adulterated at a level of 1% was detected, confirming previous findings of Torrecilla et al. (2010) who succeeded to quantify the adulteration of EVOO with refined OO, pomace OO, sunflower and corn oils with R^2=0.97 and RMSEP = 0.007% by applying PLSR (Table 5).

Regarding the determination of fatty acid, diacylglycerol and pigment amountsR^2 ranging between 0.5 and 0.8 were

Table 4. A summary overview of fluorescence spectroscopy methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
EVOO	65	EEFS	PCA and PLS-DA	Geographical authentication of EVOO according to 7 Argentinian regions	100% of correct classification of EVOO samples according to their geographical origin.	(Jiménez-Carvelo, Lozano, and Olivieri 2019)
EVOO	65	EEFS	PCA, PARAFAC, LDA, DN-PLSR	Geographical authentication of EVOO according to 2 Maltese regions	73, 80 and 93% of correct classification of EVOO samples according to their geographical origin by applying LDA, PARAFAC and DN-PLS, respectively.	(Lia et al. 2020)
EVOO	87	FFFS	PCA, LDA, SVM and k-NN	-Geographical authentication of EVOO according to 5 Italian regions -Authentication of VOO according to their altitude -Varietal authentication of EVOO according to 4 Italian cultivars	−94%, 96%, and 90% of accuracy of EVOO samples according to their geographical origin, altitude, and cultivars, respectively.	(Al Riza et al. 2021)
VOO	41	FFFS	PCA, FDA, CCSWA and PLSR	-Geographical authentication of EVOO according to 5 Moroccan regions -Varietal authentication of EVOO according to 4 cultivars -Prediction of FA, PV, k_{232} and k_{270} parameters and chlorophyll level	-Good discrimination of VOO samples according to their geographical origin, cultivar, with 96.72% and 95.12% of correct classification, respectively. -FA, PV, k_{232}, k_{270} and chlorophyll level of VOO samples are predicted with of R^2 of 0.98, 0.96, 0.88, 0.88 and 0.89, respectively.	(Zaroual et al. 2020)
EVOO	36	FFFS	PARAFAC	Control of olive cultivar irrigation	Good discrimination of EVOO samples according to their irrigation mode since R ranging between 0.85 and 0.89 was obtained.	(Cabrera-Bañegil et al. 2018)
EVOO, VOO and POO	12 EVOO, 12 VOO and 12 POO	SFS	SPA and LDA	Classification of different class of OO	Excellent discrimination of samples with 99% of correct classification.	(Dankowska, Małecka, and Kowalewski 2013)
EVOO	99	SFS	PLSR	Detection of the adulteration of EVOO by ROO with level of adulteration ranging between 2 and 20%	Detection of high accuracy of the presence of ROO in EVOO (R^2=0.94).	(Mabood et al. 2016)
EVOO	39	LIFS	PLSR	Detection of the adulteration of EVOO by soybean oil with level of adulteration ranging between 1 and 30%	Detection of high accuracy of the presence of soybean oil in EVOO (R^2 =0.98).	(Milanez et al. 2017)
EVOO	25	SFS	LDA-PARAFAC, UPLS and DA-UPLS	Detection of the adulteration of EVOO by POO with level of adulteration ranging between 3 and 33%	Detection of high accuracy of the presence of POO in EVOO (99% accuracy).	(Durán Merás et al. 2018)
EVOO	120	LIF	PCA, LDA and PLSR	Detection of the adulteration of	Detection of high accuracy of the presence of	(Li et al. 2020)

(*continued*)

Table 4. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
				EVOO by soybean oil with level of adulteration ranging between 3 and 20%	soybean oil in EVOO (R^2 =0.92).	
EVOO	63	EEFS	PLSR and PCR	Detection of the adulteration of EVOO by mild deodorized and ROO with level ranging between 2.5 and 75%	Detection of high accuracy since R^2 of 0.98 and 0.98 was observed, respectively, with PLSR and PCR.	(Wójcicki et al. 2015)
VOO	90	SFS	PLSR and N-PLS	Prediction of FA, PV, k_{232} and k_{270} parameters	Excellent prediction of FA, PV, k_{232} and k_{270} with R^2 of 0.93, 0.96, 0.9 and 0.98, respectively.	(Guzmán et al. 2015)
VOO	52	FFFS	PLSR	Prediction of phenolic level	Excellent prediction of phenolic level since R^2 of 0.95 was observed.	(Squeo et al. 2019)

CCSWA: common components and specific weight analysis; DN-PLSR: Discriminant multi-way partial least squares regression; EEFS: Excitation-emission fluorescence spectroscopy; EVOO: Extra virgin olive oil; FA: Free acidity; FDA: Factorial discriminant analysis; FFFS: Front-face fluorescence spectroscopy; k-NN: K-nearest neighbors; LDA: Linear discriminant analysis; LIFS: laser-induced fluorescence spectroscopy; N-PLS: N-way partial least squares; OO: Olive oil; PARAFAC: Parallel factor analysis; PCA: Principal component analysis, PLS-DA: Partial least squares discriminant analysis; PLSR: Partial least squares regression; POO: Pomace olive oil; PV: Peroxide value; ROO: Refined olive oil; SFS: Synchronous fluorescence spectroscopy; SPA: Successive projection algorithm; SVM: Support vector machine; UPLS: Unfolded partial least squares: VOO: Virgin olive oil.

obtained following the application of PLSR to UV-Vis spectra scanned on 89 VOO (Uncu, Ozen, and Tokatli 2019).

Nuclear magnetic resonance spectroscopy

Nuclear Magnetic Resonance Spectroscopy (NMR) is recognized as a fundamental analytical tool that is applied in organic chemistry to investigate the molecular structure and to study kinetics, dynamics, and interactions between molecules (Moro et al. 2020). In the food sector, the technique was used to: i) assess the freshness level of fish (Tan et al. 2018); ii) predict chemical properties of crude oil properties (Moro et al. 2020); and so on.

Regarding OO, the NMR technique was used to authenticate VOO samples according to their geographical origin. Longobardi et al. (2012) succeeded by applying PCA, CA and nearest class mean (PCA-CA-NCM) to the ^{1}H NMR spectra to differentiate between 104 EVOO originating from 7 different regions (Italy − 3 regions, Greece − 4 regions). Recently, Özdemir et al. (2018a) accomplished to discriminate monovarietal OO obtained from Turkish (Gemlik, Ayvalık and Memecik) and Slovenian (Bianchera) cultivars by applying PCA on ^{1}H NMR profiles. Indeed, the PCA similarity map showed clear discrimination between 89 Turkish and 37 Slovenian EVOO samples according to their botanical origins; however, the authors stated that to consider NMR spectroscopy as a tool to differentiate OO according to their botanical cultivars, it is necessary to check its reliability on different varieties provided from the same geographical location (Table 5).

Hatzakis et al. (2010) coupled ^{1}H NMR and ^{31}P NMR data with Bland and Altman statistical analysis to detect refined hazelnut oil at different levels (1% to 95%) in refined OO. The obtained results showed good detection of the presence of refined OO and hazelnut oil in EVOO with 100% of correct classification. Šmejkalová and Piccolo (2010) succeeded also to detect the presence of sunflower, soybean, hazelnut and peanut oils in EVOO at levels varying in the 10-50% range by applying DA on NMR spectra data.

NMR spectroscopy is used to quantify volatile organic compounds. For instance, Dourou et al. (2020) applied PLS-DA to NMR and an accuracy of 98% was obtained. In another hand, Hatzakis et al. (2010) succeeded to predict total, free and esterified sterols in EVOO by using ^{1}H NMR and ^{31}P NMR spectroscopy coupled with Bland and Altman statistical analysis (BASA) since 96% of good prediction was obtained (Table 5).

Other techniques: calorimetric and electronic sensing techniques

Other analytical techniques were used to determine the quality of OO. For instance, differential scanning calorimetry (DSC) was applied to study the action of heat on OO molecules. The changes due to warming are called thermal transitions like melting. It is a major analytical technique that measures the emission or absorption of heat from a biomolecule during a controlled increase or decrease of temperature (Valli et al. 2016).

Regarding OO, DSC was applied to detect the adulteration of OO (Table 6). For instance, Van Wetten et al. (2015) succeeded to detect the presence of sunflower at 2% in EVOO samples with 95% confidence level. In a different approach, Cerretani et al. (2012) applied PLSR to VOO thermal profiles to quantify palmitic, stearic, oleic and linoleic acids. R^2 values of 0.93, 0.77, 0.9 and 0.95 were obtained, respectively, with error of prediction less than 1%. The authors suggested the use of the

Table 5. A summary overview of Raman, ultraviolet-visible and nuclear magnetic resonance spectroscopy methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
EVOO	412	Raman spectroscopy 3100-100 cm^{-1}	PCA and FDA	Harvesting year (n = 2), variety (n = 4: Frantoio, Hojiblanca, Picual and Gorda) and geographical origin (n = 12 Spanish regions) authentication	Samples are clearly discriminated with 94.3%, 84.0%, and 89.0% of correct classification of samples according to their harvesting year, olive variety and geographical origin, respectively.	(Sánchez-López et al. 2016)
EVOO	38	Raman spectroscopy 1800 – 900 cm^{-1}	PCA and LDA	Geographical authentication of EVOO according to 7 Italian regions	Samples are clearly discriminated with 82 % of correct classification.	(Portarena, Baldacchini, and Brugnoli 2017)
EVOO	20	Raman spectroscopy 1800 – 900 cm^{-1}	CLPP, PCA, Pearson's correlation SIMCA, PLS-DA, PLSR, UHC and k-NN	Detection of the adulteration of EVOO by hazelnut oil with level of adulteration ranging between 1 and 90%	Good detection of the presence of hazelnut oil in EVOO with 82% of correct classification.	(Georgouli, Martinez Del Rincon, and Koidis 2017)
VOO	16	Raman spectroscopy 1800 – 900 cm^{-1}	iPLS, SiPLS and SNV	Detection of the adulteration of VOO by waste cooking oil with level of adulteration ranging between 2.5 and 50%	Excellent detection of VOO adulteration by waste cooking oil since R^2 of 0.98 was obtained.	(Li et al. 2018)
EVOO	57	UV-Vis spectroscopy 190 – 1100 nm	PCA, UNEQ and SIMCA	Geographical authentication of EVOO according to 3 Italian regions	Excellent discrimination of EVOO according to their geographical origin since 100% of correct classification was obtained.	(Casale et al. 2012)
EVOO	40	UV-Vis spectroscopy 190 – 1090 nm	LDA and PLS-DA	Geographical authentication of EVOO according to 3 Spanish regions	Excellent discrimination of EVOO according to the geographical origin with 92.5% of correct classification.	(Pizarro et al. 2013)
VOO, POO and ROO	81 VOO, 10 ROO and 17 POO	UV-Vis spectroscopy 380 – 730 nm	PCA and LDA	Classification of OO according to their quality	Excellent classification of OO according to their quality with 95% of correct classification.	(Ferreiro-González et al. 2017)
EVOO	817	UV-Vis spectroscopy 190 – 900 nm	PLSR	Detection of the adulteration of EVOO with ROO, POO, sunflower and corn oils at a level of 5%	Excellent detection of ROO, POO, sunflower and corn oils in EVOO with R^2 of 0.97 and RMSEP of 0.007%.	(Torrecilla et al. 2010)
EVOO	141	UV-Vis spectroscopy 380 – 800 nm	PLSR and MLP	Detection of the adulteration of EVOO with soybean and sweet almond oils at a level of 1%	Excellent detection of soybean and sweet almond oils in EVOO with mean absolute error of 2.84%.	(Aroca-Santos et al. 2019)
EVOO	104	NMR ^1H	PCA and PCA-CA-NCM	Geographical authentication of	PCA coupled with CA and NCM	(Longobardi et al. 2012)

(*continued*)

Table 5. Continued.

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
				EVOO according to 7 regions (Italy: 3 regions, Greece: 4 regions)	approach allowed to better differentiation between EVOO according to their 7 geographical origins than PCA approach.	
EVOO	126	NMR ^{1}H	PCA and PLS-DA	Varietal authentication: Gemlik, Ayvalık, Memecik and Bienchera	Excellent differentiation between EVOO according to their 4 varieties since 92.5% of correct classification was obtained.	(Özdemir et al. 2018a)
EVOO	15	NMR ^{1}H	LDA	Detection of the adulteration of EVOO by sunflower, soybean, hazelnut and peanut oils with level ranging between 10 and 50%	Excellent detection of the presence sunflower, soybean, hazelnut and peanut oils in EVOO with correct classification ranging between 98 and 100%.	(Šmejkalová and Piccolo 2010)
EVOO	24	NMR ^{1}H and ^{31}P	BASA	Prediction of total, free and esterified sterols in EVOO	Excellent prediction of total, free and esterified sterols since 96% of predictive accuracy was observed.	(Hatzakis et al. 2010)
EVOO	45	NMR ^{1}H	OPLS-DA	Prediction of VOC in EVOO	Excellent prediction of VOC in EVOO since 98% of predictive accuracy was observed.	(Dourou et al. 2020)

BASA: Bland and Altman statistical analysis; CLPP: Continuous locality preserving projections; EVOO: Extra virgin olive oil; FDA: Factorial discriminant analysis; iPLS: Interval partial least squares; k-NN: K-nearest neighbors; LDA: Linear discriminant analysis; MLP: Multilayer perceptron; NCM: Nearest class mean; NMR: Nuclear magnetic resonance spectroscopy; OO: Olive oil; PCA: Principal component analysis, OPLS-DA: Orthogonal projections to latent structure discriminant analysis; PLS-DA: Partial least squares discriminant analysis; PLSR: Partial least squares regression; POO: Pomace olive oil; ROO: Refined olive oil; SIMCA: Soft independent modeling of class analogy; SiPLS: Synergy interval partial least squares; SNV: Standard normal variate; UHC: Unsupervised hierarchical clustering; UNEQ: Unequal class models; UV-Vis: Ultraviolet-Visible spectroscopy; VOC: Volatile organic compounds; VOO: Virgin olive oil.

technique for determining fatty acid levels in order to propose it as an alternative tool to the chromatographic methods.

Electronic sensing technologies (e-sensing) including electronic noses, eyes and mouth are the technologies based on the capability of reproducing human senses using sensor arrays and pattern recognition systems. Indeed, electronic noses and mouth are the techniques that could detect and recognize odors and flavors, while electronic eye is the technique that simulates the humane eye by recognizing structures. The technique was used for the determination of the quality of OO (Table 6). For instance, Haddi et al. (2013) succeeded to differentiate VOO according to their geographical origin. The same research group succeeded, later, by applying LDA to recognize 27 VOO originating from 5 regions of Morocco (Ouarzazate, Ouazzane, Taounate, Mrir't and Sidi Ali).

In a different approach, electronic sensing technologies were also utilized to: i) detect the presence of sunflower, soybean and corn oils in EVOO at different levels (2-25%) by using voltammetric e-tongue (Apetrei and Apetrei 2014); and ii) monitor the freshness level of Italian VOO during storage at 40 and 60 °C (Buratti et al. 2018) by applying k-NN on electronic senses (noses, mouth and eye). The obtained results were confirmed, recently, by Martínez Gila et al. (2020) who succeeded to predict sensory attributes of VOO extracted from olive fruits stored at different conditions and during different times. The authors suggested the application of electronic nose as a complementary tool to the official method developed by the Olive International Council (OIC) for the determination of the sensory attributes of OO. These findings are in agreement with those of Laddomada et al. (2013) who succeeded to obtain good correlation between thermal properties and PV, single free FATs (palmitic, oleic and linoleic acids) and unsaturated/saturated FATs ratio.

Conclusion

Considerable interest is paid to OO quality and methods of production due not only to its nutritive quality but also to the crises and scandals in the food industry, which have

Table 6. A summary overview of differential scanning calorimetry and electronic-sensing methods used for determining quality and authenticity of virgin olive oils (VOO) and extra virgin olive oils (EVOO).

Product	Number of samples	Materials & Methods	Data Analysis	Objectives	Main results	Reference
VOO	15	Electronic nose and tongue	PCA, CA and SVM	Geographical authentication of VOO according to 5 Moroccan regions	100% of correct classification of EVOO according to their geographical origin was obtained.	(Haddi et al. 2013)
EVOO	20	Electronic noses	PLS-DA and PLSR	Detection of the adulteration of EVOO by sunflower, soybean and corn oils with level of adulteration ranging between 2 and 55%	Excellent detection of EVOO adulteration with 100% of correct classification.	(Apetrei and Apetrei 2014)
VOO	24	Electronic senses (noses, mouth and eye)	k-NN	Monitoring the quality of VOO during storage	94% of correct classification of VOO according to their storage time.	(Buratti et al. 2018)
EVOO	12	DSC	not mentioned	Detection of the adulteration of EVOO by sunflower oil	Excellent detection of sunflower oil in EVOO with 95% confidence level.	(Van Wetten et al. 2015)
VOO	63	DSC	PLSR	Prediction of FATs in VOO	Excellent prediction of FATs with R^2 of 0.95	(Cerretani et al. 2012)
VOO	18	MASC	ANOVA	Prediction of PV, FATs, unsaturated/ saturated FATs ratio, FA, linolenic acid, polyphenol and o-diphenol contents	-Good correlation between thermal properties and PV, single free FATs (palmitic, oleic and linoleic acids) and unsaturated/ saturated FATs ratio -No significant correlation ($P > 0.05$) was observed between thermal properties and FA, linolenic acid, polyphenol and o-diphenol content.	(Laddomada et al. 2013)
VOO	82	Electronic noses	NB, PLS-DA and MLP	Monitoring the quality of VOO during storage	90.24% of correct classification of VOO according to their storage time.	(Martínez Gila et al. 2020)

ANOVA: Analysis of variance; CA: Canonical analysis; CDA: Canonical discriminant analysis; DSC: Differential scanning calorimetry; EVOO: Extra virgin olive oil; FA: Free acidity; FATs: Fatty acids; k-NN: K-nearest neighbors; LDA: Linear discriminant analysis; MASC: Modulated adiabatic scanning calorimeter; MLP: Multilayer perceptron; NB: Naïve Bayes, PCA: Principal component analysis, PLS-DA: Partial least squares discriminant analysis; PLSR: Partial least squares regression; SVM: Support victor machine.

seriously undermined consumer confidence and grown the need for rapid analytical techniques to determine OO quality and authenticity. This great need for authentication and quality assurance requires appropriate analytical tools for OO analysis before production (olive fruits analysis), and during both extraction and storage.

Targeted methods such as chemical, sensory, and chromatographic techniques considered as the official OIC methods were used to assess quality and authenticity of OO. However, most of these methods are either time consuming, destructive and demand trained personnel. This requires the development of fast, environment-friendly and simple analytical techniques such as the spectroscopic ones that are relatively low-cost and can be applied in both fundamental researches and the factory as on-line sensors for the determination of OO quality. In this review, recent advances in OO quality evaluation and authenticity by NIR, MIR, fluorescence and NMR are presented and discussed. From the results presented between 2010-2020 in the present review, it could be mentioned that spectroscopic techniques coupled with chemometric tools have the potential to determine the quality and authenticity of OO. Up-to-date, the combination of these spectroscopic techniques for the evaluation of the quality of OO are scarce. Thus, their combination would make them more convenient and effective for the analysis of OO allowing them to be used as rapid screening techniques to the standard reference methods for determining the quality and authenticity of OO.

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