



Review

The use of analytical techniques coupled with chemometrics for tracing the geographical origin of oils: A systematic review (2013–2020)

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ABSTRACT

The global market for imported, high-quality priced foods has grown dramatically in the last decade, as consumers become more conscious of food originating from around the world. Many countries require the origin label of food to protect consumers need about true characteristics and origin. Regulatory authorities are looking for an extended and updated list of the analytical techniques for verification of authentic oils and to support law implementation. This review aims to introduce the efforts made using various analytical tools in combination with the multivariate analysis for the verification of the geographical origin of oils. The popular analytical tools have been discussed, and scientometric assessment that underlines research trends in geographical authentication and preferred journals used for dissemination has been indicated. Overall, we believe this article will be a good guideline for food industries and food quality control authority to assist in the selection of appropriate methods to authenticate oils.

1. Introduction

Nowadays, the issues of food quality and safety have gained great attention from both food processors and consumers (Janssen & Hamm, 2012; Valdés, Beltrán, Mellinas, Jiménez, & Garrigós, 2018). In this regard, the main role of quality and safety is achieved through the identification, certification, and traceability of foods (Lees, 2003; van Rijswijk, Frewer, Menozzi, & Faioli, 2008). In Europe, geographic verification is one of the key authenticity concerns for food products (Danezis, Tsagkaris, Brusica, & Georgiou, 2016). Accordingly, European countries have introduced legislation to protect 'mountain products' and 'products of island farming' (EU regulation 1151/2012). These two markers are the up-to-date companions to the protected designation of origin (PDO) and protected geographical indication (PGI) labels

(Danezis, Tsagkaris, Brusica, et al., 2016). High-quality foods with known geographical origin have a higher price and gain a higher financial profit to the PDO/PGI producers compared to other products (Gazeli, Bellou, Stefanis, & Couris, 2020; Nescatelli et al., 2014). Therefore it is important to protect such products from possible commercial fraud. These products (e.g. Kalamata olive oil PDO and Sabina PDO) are defined by production area, processing and preparation process, and use of certain farming systems. Food authentication is of great interest to consumers, legislation authorities, the scientific community, importers, and exporters. Production of fraudulent food and illegal food trade are widespread throughout the world.

Several selective conventional techniques (target analysis) such as high-performance liquid chromatography (HPLC), gas chromatography (GC), ultra-high-performance liquid chromatography (UHPLC), and

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inductively coupled plasma (ICP) coupled with the mass spectrometry are used to analyze the chemical composition of edible oils and fats (Borges et al., 2017; Longobardi et al., 2012; Rutkowska, Bialek, Adamska, & Zbikowska, 2015). Analytical tools that can produce multiple data for each sample tested such as infrared spectroscopy (IR), Raman spectroscopy, Nuclear Magnetic Resonance spectroscopy (NMRs), terahertz spectroscopy (THzS), ultraviolet–visible (UV–Vis) spectroscopy and fluorescence spectroscopy have also been applied to authenticate edible oils (Esteki, Shahsavari, & Simal-Gandara, 2018; Forina et al., 2015; Medina, Perestrelo, Silva, Pereira, & Câmara, 2019; Wang, Sun, Zhang, & Liu, 2016). Besides, previous studies have shown the rapid growth of research articles that used multi-elements and multi-isotope analyses for tracing the geographical origins of foods (Drivelos & Georgiou, 2012; Durante et al., 2015; Laursen, Schjoerring, Kelly, & Husted, 2014). Recently, the possibility of an electronic nose (e-nose) and electronic tongue (e-tongue) for verifying the geographical origin of oils have been reported (Cosio, Romano, & Scampicchio, 2016; Forina et al., 2015; Haddi et al., 2013). Accurate techniques for oils authenticity analysis does not benefit only consumers but also include regulatory authorities and food processors who are attempting to guarantee appropriate compliance and branding of oil labeling.

Research activities in geographical origin have increased substantially between 2013 and 2020, which reveals the great attention of researchers to the value of food geographical authentication (Fig. 1). Many reviews have been published on specific instrumental techniques combined with a chemometric approach for authentication of edible oils (Aparicio, Morales, Aparicio-Ruiz, Tena, & García-González, 2013; Bosque-Sendra, Cuadros-Rodríguez, Ruiz-Samblás, & de la Mata, 2012; Esteki et al., 2018; Luykx & van Ruth, 2008; Valdés et al., 2018). However, most of these reviews focused on botanical and adulteration identification. In this review, we provided extensive information on the latest studies published in the use of various analytical tools in conjunction with the chemometrics applied to declare the geographical origin of edible oils from 1/January/2013 to 15/December/2020. In order to show the status of the research in this field, section 3 indicated an overall outline of articles published in the above-mentioned period.

2. Methods

2.1. Literature search to find the relevant papers

A systematic review of published articles was used to report the application of analytical tools coupled with chemometrics for potential use in the authentication of the geographical origin of oils. A comprehensive database search was carried out on Web of Science, Scopus, Google scholar, and Pub Med databases between 1/January/2013 to 15/

December/2020. This process was performed as group work. Searches were limited by the following terms: Oil OR edible oil AND Multivariate OR chemometrics OR pattern recognition AND authentication OR geographical origin. Besides, the reference lists of all selected articles were reviewed to retrieve additional studies. Fig. 2 describes the systematic search and study selection procedure. The outcome measurement technique was restricted to the use of classification techniques (Table 1).

2.2. Qualification and data extraction

The studies were considered eligible as they fulfilled the following criteria: (i) reporting the application of analytical tools coupled with the chemometrics; (ii) application in oils of known geographical source; (iii) carried out between first January 2013 and 15 December 2020; (iv) published in the English language; and (v) full-text article available. Conversely, the studies that did not apply chemometrics methods for authentication of the geographical origin of oils, conferences, letters, abstracts, review papers, books, and chapters were excluded. The extracted characteristics of each study consist of detection techniques, oil types, number of samples, geographical origin, chemometric approaches used, and the percentage of discrimination rate (specifically for supervised algorithms) (Table 1).

3. Research status

The geographical difference is a clear intraspecific variation in oils. Oils produced in different environments always revealed differences in chemical composition. This makes the geographical variation a key factor in the study of the oil quality; as the oils' geographical origin is of great importance for food processors and consumers. The geographical source of agri-food products has become a vital tool of discrimination; it is a distinctive characteristic that makes products challenging to be reproduced and assumed to have a quality indication for agri-food products (Santeramo & Lamonaca, 2020). An evaluation of using analytical tools combined with the chemometrics for the geographical origin of foods authenticity can be done easily and quickly using the Web of Science, Scopus, Google scholar, and Pub Med database. This survey showed that oils are a category of foods often taken through geographical authentication (Fig. 1). Based on the database search, it is clear that oils geographical confirmation revealed the highest cases during 2017 and 2018 and followed by decreases during 2019–2020 (Fig. 3).

3.1. Research activity spreads to different countries

Research on oil geographical variations mostly comes from the main producing olive oil countries such as Tunisia, Portugal, Morocco, Australia, Argentina, Italy, Greece, and Spain. It is clear from Table 1 that European countries are active in this field for many years, African and Asia's countries showed less activity while no article was published from the USA. This could be attributed to the production of high-quality oil in Europe. Among the European countries, Spain and Italy showed the highest activities in the field and this correlated with the high number of PDO-PGI-PGI food products produced in these countries.

3.2. Journals

Based on the database information, the publications on oil geographical authentication are distributed among 21 journals. Several journals published oil authentication articles, although preferences for particular journals have been observed. A significant number of articles accounting for 52% of the total (Fig. 4) were published in the "Food Chemistry", "European Journal of Lipid Science and Technology" and "Food Research International" These journals are highly cited food journals. Other considerable numbers of articles were published in foods (7%),

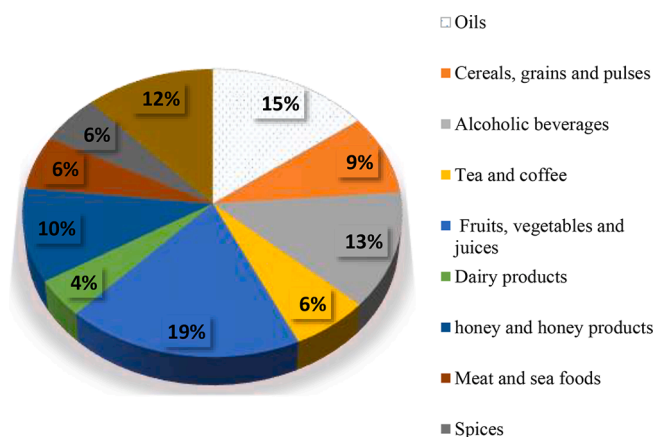


Fig. 1. Percentage of food geographical origin authentication cases from 2013 to 2020 based on the database information.

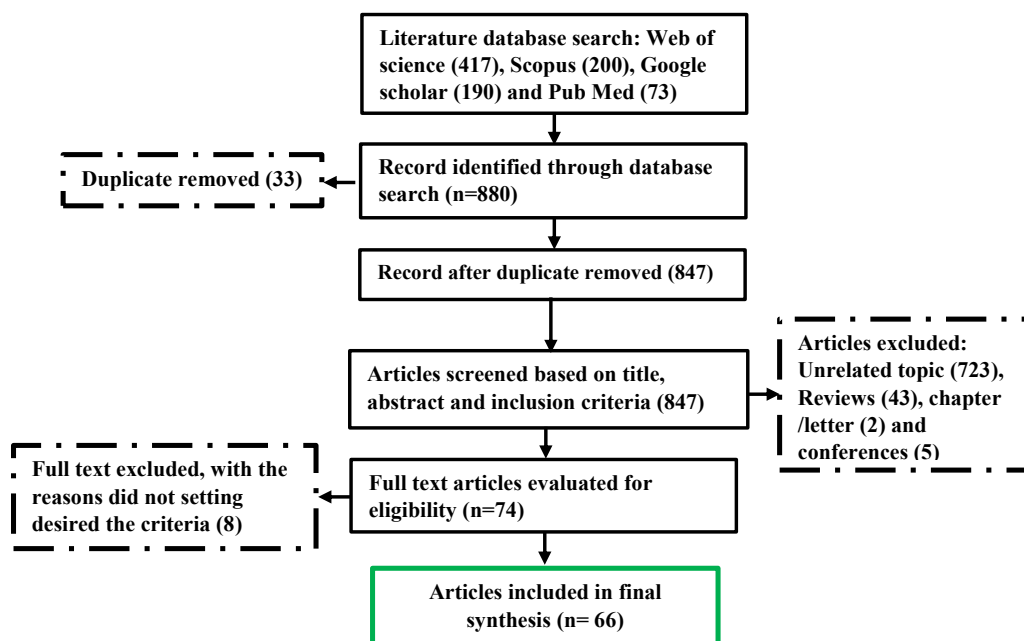


Fig. 2. Systematic literature search and study selection strategy.

Journal of The American Oil Chemists Society (7%), *Talanta* (5%), *European Food Research and Technology* (3%), and *Chemometrics and Intelligent Laboratory Systems* (3%) (Fig. 4).

3.3. Analytical tools used for oil authentication

Authentication of food includes various analytical techniques. This section provides brief information on analytical tools to verify the geographical origin of oil in the context of regulatory and more recently consumer and food processor requirements. Regarding geographical confirmation, analytical techniques depend mainly on the measurement of chemical components, which may be relatively similar even when compared to oils gathered from different production zones. Attempts had been made by measuring some components such as phenolic compounds, fatty acid profile, sterols, triacylglycerol (TAGs), volatile compounds, and color as a typical region. Chromatographic, inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) are the major methods for oil geographical verification. These three methodologies accounted for more than half of published articles (Table 1). UV–vis spectroscopy and infrared spectroscopies are also evident.

Some other technologies such as Raman, THz spectroscopy, laser-induced breakdown spectroscopy (LIBS), fluorescence spectroscopy, isotope ratio mass spectrometry (IRMS), differential scanning calorimetry (DSC), electronic tongue, and electronic nose have not been used to the maximum. However, our opinion is that they will find extensive use in the near future. This deserved the attention of food scientists, who must take advantage of cross-scientific collaboration.

In the past seven years, an emerging trend shows that chromatographic, NMR, and infrared spectroscopy methods are increasing in popularity for oil authentication. The combination of data from different techniques appears to be one of the most promising approaches to establish the geographical origin (Table 1).

3.4. Chemometrics in geographical verification of oils authentication

The term chemometrics can be described as the science of extracting information from chemical structures via mathematical modeling of data (Wold, 1972). In 1997, Massrat defined chemometrics as a chemical discipline that utilizes mathematics and formal logic in order to (1)

design optimum experimental methods; (2) to produce maximum chemical information by evaluating chemical data; and (3) to get information regarding chemical systems” (Massart et al., 1998). In this review, a brief introduction to supervised and unsupervised approaches used in the authentication of edible oils was presented. Other methodologies that transcend the scope of this work were discussed elsewhere. Unsupervised technique divided data sets into clusters without specifying groups of the samples; instead, trends between samples were recognized without previous information regarding patterns or groups (Jain, Duin, & Mao, 2000). Hierarchical cluster analysis (HCA) and principal components (PCA) are still the well-known unsupervised techniques applied in food authentication (Berrueta, Alonso-Salces, & Héberger, 2007). Supervised techniques split data set into a training set based on a prior allocation of samples to predefined groups to build calibration models and that are subsequently employed to predict unknown samples (Esteki et al., 2018). The majority of chemometrics used in food authentication begins with PCA (as exploratory data analysis) before supervised learning steps for detecting patterns in the analyzed data (Baker & Inventado, 2014; Lavine, 2006). PCA, PLS-DA, and LDA are the methods mostly used in testing the geographical origin of oils (Table 1). Recently, the use of innovative pattern recognized techniques is increasing in the field of food because of their power and ability to address complex problems associated with food classification. Support vector machine (SVM) and random forest (RF) were the most frequent techniques employed in both classification and calibration models (Jiménez-Carvelo, González-Casado, Bagur-González, & Cuadros-Rodríguez, 2019). However, their application is still rare in the field of food geographical origin authentication. Some studies have demonstrated their strengths as compared with conventional chemometrics (Table 1). In a comparative study, Gromski et al. (2015) applied PLS-DA and SVM; they found that SVM was not affected by the allocation of the different sample groups, but focused on the side of the support vectors that specific test samples fall on. Similarly, an ecological study demonstrated the advantages of RF models (Cutler et al., 2007). The chemometrics generated by decision tree methods such as RF, and SVM cannot separate data into different classes, but the samples were distributed into subclasses dependent on the value of certain variables, and this procedure was repeated on each resultant subclass of samples (Ai et al., 2014). Thus, the discriminations depend on a set of integrated decisions comparable to artificial neural networks (ANN). Previous works

Table 1

Various analytical tools combined with chemometrics to assess the authenticity of edible oils in recent years (2013–2020).

Detection techniques	Oil types	Number of samples	Geographical origin	Chemometric approaches	Accuracy %	References
UHPLC-QTOF-MS	EVOOs	57	Spain	PLS-DA, OPLS-DA	94.4	(Gil-Solsona et al., 2016)
UHPLC-ESI/QTOF	EVOOs	26	Tunisia, Italy	OPLS-DA, VIP	94	(Mohamed et al., 2018)
HPLC-ESI-IT-MS	EVOOs	136	Morocco	PCA, LDA	100	(Bajoub, Ajal et al., 2016)
HPLC/DAD, HPLC/ESI-MS	EVOOs	77	Italy	biPLS-GA- PLS-DA	85	(Nescatelli et al., 2014)
HPLC-RI	OOs	279	Morocco	PCA, SIMCA, PLS-DA, LDA	100	(Bajoub, Medina-Rodriguez et al., 2016)
UHPLC-ESI-MS/MS	EVOOs	50	Spain	PCA, LDA	92.4%	(Becerra-Herrera et al., 2018)
HPLC-ESI-TOF/MS, HPLC-DAD	EVOOs	32	Spain	DA	100	(Bakhouch et al., 2013)
HPLC-UV& HPLC-CAD	palm oil	102	Malaysia, Indonesia, Papua New Guinea, and Salomon Islands, Ghana, Guinea, Ivory Coast, Nigeria, and Cameroon Brazil.	PLS-DA, SIMCA	92	(Pérez-Castaño et al., 2015)
HPLC-UV, HPLC-CAD	Crude palm oil	100	Malaysia, Indonesia, Papua New Guinea, and Salomon Islands, Ghana, Guinea, Ivory Coast, Nigeria and Cameroon Brazil.	PLS-DA	100%	Obisesan et al. (2017)
GC-FID, HPLC	VOOs	6	Tunisia	PCA, HCA	–	(Hlima et al., 2017)
HPLC- CAD, (HT)GC-FID	EVOOs	65	Spain	SIMCA, PLS-DA	100	(Vera et al., 2019)
HTGC-MS& HPLC-CAD	palm oil	85	Malaysia, Indonesia, Papua New Guinea, Salomon Islands, Ghana, Guinea, Cote d'Ivoire, Nigeria, and Cameroon and Brazil	PLS-DA	100.	(Ruiz-Samblás et al., 2013)
GC-IMS	EVOOs	53	Greece, Italy, Spain	PCA-LDA	100	(Schwolow et al., 2019)
GC-FID	OOSs	242	French, Tunisia, Algeria, Morocco	PCA, SIMCA	96.51	(Laroussi-Mezghani et al., 2015)
GC-FID	Palm oils	94	Malaysia, Indonesia, Papua New Guinea, Salomon Islands, Ghana, Guinea, Cote d'Ivoire, Nigeria, and Cameroon and Brazil	PLS-DA	100	(Tres et al., 2013)
GC-FID, GC/MS	VOOs	255	Cyprus	PCA	–	(Kritioti et al., 2018).
GC × GC-TOF-MS and 1D-GC-MS	VOOs	30	Croatia	PCA, HCA, SLDA	100	(Lukić et al., 2019)
HS-SPME-GC-MS, GC-FID	VOOs	1217	Italy	LDA	97	(Cecchi et al., 2020)
HPLC-RI/ELSD/MS	Hazelnut oil	50	Turkey	PCA, HCA	–	(Kıralan et al., 2015)
HS-SPME-GC-MS	EVOOs	82	Croatia, Slovenia, Spain, Italy, Greece, Morocco Turkey	LS-DA	100	(Quintanilla-Casas et al., 2020)
GC- FID, Physicochemical properties, oxidative stability	OOs	33	Brazilia, Spain	PCA	–	(Borges et al., 2017)
SPME-GC/MS	OOs	74	Greece	LDA	87.8	(Kosma et al., 2017)
GC-FID	VOOs	101	France	PCA, Fuzzy ID3, C4.5	85.15	(Nasibov et al., 2016)
GC-MS, NIRS, MALDI-TOF/MS	EVOOs	49	Croatia	PCA, PLS-DA	96	(Peršurić et al., 2018)
GC-FID, ICP-AES and others	VOOs	33	Greece	MANOVA, LDA	94.5	(Karabagias et al., 2013)
SIFT-MS	Extra Virgin Argan Oils (EVAO)	95	Morocco	SIMCA, PLS-DA, SVM, KNN	100	(Mourad Kharbach et al., 2018)
SIFT-MS	OOs	130	Morocco, Greece, Spain, Italy	PCA, PLS-DA	100	(Bajoub et al., 2018)
ICP-OES	Sesame oil	8	Yemen	PCA and HCA	–	(Mohammed et al., 2018)
ICP-MS	OOs	49	Turkey	PCA, HCA	–	(Gumus et al., 2017)
ICP-MS	OOs	21	Tunisia	LDA	92.1	(Damak et al., 2019)
ICP-MS	VOOs	35	Spain	LDA	100	(Beltrán et al., 2015)
ICP-MS/OES	EVOOs	125	Spain	PCA, PLS-DA, LDA, SVM RF	98	(Sayago et al., 2018)
ICP-MS	EVOs	50	Italy	PCA, LDA	100	(Aceto et al., 2019)
ICP-MS	Pumpkin seed oil	660	Austria, China, and Russia	PCA	–	(Bandoniene et al., 2013)
¹ H NMR	VOOs	38	Romania, EU and China	PCA	–	(Popescu et al., 2015)
¹ H NMR, IRMS	VOOs	125	Italy, Spain, Greece, France, Turkey, and Cyprus	PCA, PLS-DA	93	(Alonso-Salces et al., 2015)
¹ HNMR	EVOOs	235	Italy, Spain and Portugal, Tunisia, Turkey, Chile, and Australia	PCA, OPLS-DA	90.5	(Rongai et al., 2017)
¹ H NMR, UF	EVOOs	49	Spain	LDA, SIMCA, PLS-DA, RF, ANNs, SVM	84.9	(Sayago et al., 2019)
¹ H NMR	EVOOs	383	Italy	LDA	90	(Ingallina et al., 2019)
¹ H NMR	EVOOs	126	Turkey and Slovenia	PCA, PLS-DA	92.50	(Ozdemir et al., 2018)
¹ H-NMR	EVOOs	1125	Italy, Greece, and Spain	PCA, CA, KNN	96	(Ole Winkelmann et al., 2019)
¹ H NMR, IRMS	OOs	263	Italy, Tunisia	PCA, RF	100	(Camin et al., 2016)
¹ H and ¹³ C NMR	EVOOs	65	Malta, Italy, Spain, France Sicily	PCA, PLS-DA, ANN	100	(Liá, Vella et al., 2020)
NIR	EVOOs	217	Italy	LDA	100	(Forina et al., 2015)

(continued on next page)

Table 1 (continued)

Detection techniques	Oil types	Number of samples	Geographical origin	Chemometric approaches	Accuracy %.	References
NIR & FT-IR	OOs	910	Italy	PLS-DA, SVM	86.3	(Devos et al., 2014)
NIR, MIR	OOs	57	Italy	PLS-DA	100	(Bevilacqua et al., 2013)
FT-NIR	OOs	765	Spain, Greece, Portugal, and Italy	LDA, KNN	72.7	(Gertz et al., 2019)
FT-IR	Argan oils	120	Morocco	PCA, PLS-DA	100	Exclude for meta-analysis (M. Kharbach et al., 2017)
Raman	EVOOs	145	Spain	DA	89.0	(Sánchez-López et al., 2016)
IRMS, Raman	EVOOs	38	Italy	PCA, LDA	82	(Portarena et al., 2017)
Front-face fluorescence	Patchouli oils	43	Indonesia	PCA	–	(Al Riza et al., 2019)
3D-Fluorescence Spectroscopy	EVOOs	65	Malta, Italy, Spain, France Sicily	DN-PLSR	98.58	(Lia et al., 2020a; Lia et al., 2020b)
Fluorescence, NIR	OOs	104	Argentina	PLS-DA	100	(Jiménez-Carvelo et al., 2019)
UV-Visible and others	EVOOs	40	Spain	PCA, LDA, PLS-DA	100	(Pizarro et al., 2013)
UV-vis	Palm oils	60	Nigeria	PCA, OPLS-DA	95	(Jolayemi et al., 2018)
THz spectroscopy	EVOOs	80	Australia, Spain, Greece, and Italy	PCA, LS-SVM, BPNN, RF	96.25	(Liu et al., 2018)
IRMS	Palm oil	33	Malaysia	OPLS-DA	71.43	(Muhammad et al., 2018)
FGC e-nose	EVOOs	251	Italy	PCA, HCA, PLS-DA	100	(Melucci et al., 2016)
D S C	EVOOs	27	Greece	LDA	90.5	(Chatziantoniou et al., 2014)
DSC	EVOOs	39	Italy, Tunisia, USA, Turkey, Albania, Greece, Andalusia and Spain	PCA	–	(Mallamace et al., 2017)
LIBS	OOs	36	Greece	k-NN, SVC and PCA-LDA	94	(Gyftokostas et al., 2020)
LIBS	OOs	400	Greece	PCA, LDA, SVM and RFC	99.2 %	(Gazeli et al., 2020)
E-nose, e-tongue	VOOs	25	Morocco	PCA, CA, SVMs	100	(Haddi et al., 2013)
E-ongue	OOs	60	Tunisia	LDA-SA	100	(Souayah et al., 2017)

demonstrated the potential of these chemometric techniques in the authentication of oils (Aparicio & McIntyre, 1998; Aparicio, Conte, & Fiebig, 2013; Bosque-Sendra et al., 2012).

3.5. Oils geographical authentication

Fingerprinting detection methods used for the declaration of the geographical origin of oil consist of numerous analytical tools such as chromatographic, e-nose, e-tongue, spectroscopy, spectrometry, and differential scanning calorimetry approaches which are presented in Table 1. In the next sections as well as in Table 1, we indicated a general outline of the analytical tools coupled with the chemometrics reported from 2013 to 2020 to certify oil geographical origin.

3.6. Separation techniques

Several chromatographic systems have been effectively applied for testing the geographical origin of foods (Luykx & van Ruth, 2008), including high-performance liquid chromatography (HPLC), gas chromatography (GC), and ultra-high performance liquid chromatography (UHPLC). HPLC and GC were the most frequently implemented techniques for the identification of targeted fingerprinting for the authentication of oils (Table 1). MS together with the chromatographic system is the most approach employed in metabolic investigation taking advantage of the strengths of both techniques; the high resolution of the chromatographic method and the high sensitivity of MS detectors (Zhang, Sun, Wang, Han, & Wang, 2012). The combination of chromatographic with MS enables many metabolites present in oil samples to be separated from each other. This allows an increase in the number of metabolites to be analyzed with the MS system without losing sensitivity (Zeki, Eylem, Reçber, Kir, & Nemutlu, 2020).

3.6.1. Gas chromatography

A significant number of oils were analyzed using GC to evaluate the volatile profiles and fatty acid composition. The GC technique used for the analysis of vegetable oils has been discussed (Cserháti, Forgács, Deyl, & Miksik, 2005). Owing to the higher number of chemical compounds, the GC data were mostly assessed by various mathematical techniques such as unsupervised and supervised pattern recognition (Bosque-Sendra et al., 2012). GC associated with chemometrics has been widely used for the measurement of volatile compounds and fatty acid

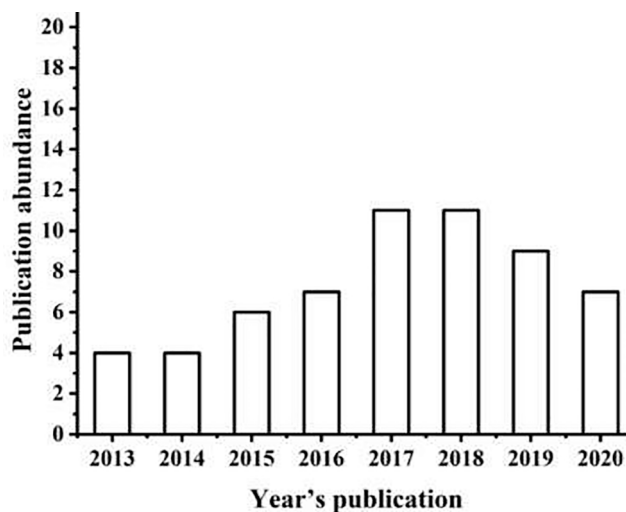


Fig. 3. Publications using various analytical tools combined with chemometrics for authentication of geographical origin of oils based on the database searches (January 2013 to December 2020).

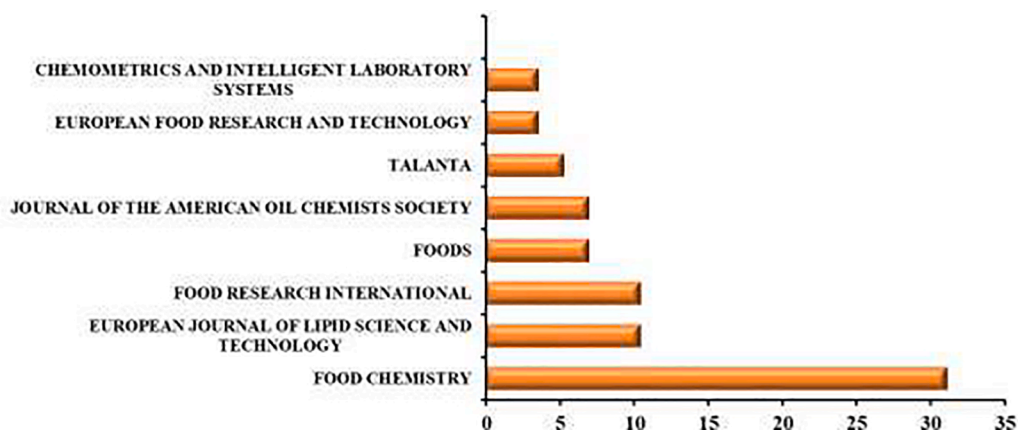


Fig. 4. Top seven Journals for oil geographical authentication.

composition of the oil. The majority of these studies employed chemometrics for processing the data set (Table 1).

The potential use of fatty acid measured by GC-MS, triacylglycerol (TAGs) through matrix-assisted laser desorption ionization time-of-flight/mass spectrometry (MALDI-TOF/MS), and nondestructive analysis by NIRS combined with the chemometrics for testing the geographical origin of Croatian extra virgin olive oil (EVOO) from three locations (Istria, Kvarner, and Dalmatia) were reported (Peršurić, Saftić, Mašek, & Kraljević Pavelić, 2018). TAGs showed a better classification for EVOO in the PCA, while the PLS-DA technique offered the best correct classification based on NIR spectra significant variable or TAGs data. The parameters that were considered for oil origin verification were free fatty acid including oleic acid (C18:1n-9c) and C18:2n-6c, and TAGs including PLP, PLL, POL, POO, LLO/LOL, LOO, and OOO. Furthermore, the PLS-DA model has been established based on these selected important components. PCA based on TAG achieved the best classification result while the PLS-DA based on the NIRS showed the highest discrimination rate (96%). The authors suggested that NIRS could be used as a rapid and inexpensive technique for the authenticity study of EVOOs while MALDI-TOF/MS was the key technique for complete profiling of EVOOs.

In a study, 256 volatile compounds were detected using GC × GC-TOF-MS and 1D-GC-MS to discriminate OOs collected from different locations in Croatia (Buža, Istarska bjelica, Rosinjola, Oblica and Lastovka). The results showed the superiority of GC × GC-TOF-MS over 1D-GC-MS regarding the sensitivity and the quantity of information. Among the identified volatile component, 131 were used for variety identification, while 60 components were found to be useful for geographical origin. Forward stepwise linear discriminant analysis (SLDA) based on the volatile components demonstrated to be a potent multivariate tool for the differentiation of VOOs according to the geographical origin (Lukić, Carlin, Horvat, & Vrhovsek, 2019). The main compounds that contributed to confirming geographical origin were 1-heptanol, 1-octen-3-ol, (E)-2-heptenal, benzyl alcohol, (E)-3-octen-2-one, dihydroactinidiolide, and nerolidol.

In another study, HS-SPME-GC-MS was used to analyze 1217 samples of olive oil (OO) collected from the main worldwide producing countries. By combining GC-MS and GC-FID (Flame Ionization Detecto) the authors developed an LDA model based on the combined data of fatty acid and VOCs profiles. Model-based on VOCs and FAs does not distinguish all samples well, especially that of Spain and Portugal. The correct classifications were very acceptable and even greater than those obtained by other authors who used the quantitative data of VOCs and FAs (Kosma et al., 2017). In order to obtain a better discrimination rate between Spain and Portugal samples, LDA only based on selected 25 VOCs (include alcohols (11), aldehydes (6), esters (4), ketones (2), carboxylic acids (1), and hydrocarbons (1) was constructed. The

combined use of FAs and VOCs obtained the best model in terms of a higher discrimination percentage of (97.2%). However, the best model in terms of percentage of correctly classified samples (88.1%) was reached, where LDA was built on the selected VOCs, with only a little lower discrimination rate of samples (94.5% in external validation) (Cecchi et al., 2020).

Borges et al. (2017) used 18 variables of fatty acid profile and other physicochemical parameters coupled with PCA to test the geographical origin of olive oil (OO) produced in different areas of Brazil and Spain; some differences were found between and within oil samples that were evaluated. However, the differences between the samples based on the variables measured in the present study do not obtain a clear grouping of the oils according to their geographical origin. Similarly, it has been reported in the literature that GC-FID and GC-MS together with PCA and HCA have been used to discriminate VOOs based on their geographical origin (Cypriot and Koroneiki) (Kritioti, Menexes, & Drouza, 2018). The geographical classification, through altitude and district factors, was not achieved, mostly due to the different parameters such as rain-fed/irrigation systems and degree of olive ripeness in the four districts, the three altitude zones, and the small land size of the country, all affected the fatty acid composition of VOOs. Fatty acid measured by GC-FID combined with T-operators in Fuzzy decision tree methodology was successfully employed for geographical authentication of French VOO obtained from North Aegean, South Aegean, South East Aegean, and the Mediterranean (Nasibov, Kantarcı Savaş, Vahaplar, & Kınay, 2016).

Recently, the geographical origin of OOs has been successfully identified using the low-level data fusion of headspace gas chromatography-ion mobility spectrometry (GC-IMS) and Fourier transform infrared spectroscopy (FT-IR) spectra combined with PCA-LDA and PLS-DA models (Schwolow, Gerhardt, Rohn, & Weller, 2019). Validation results showed that the discrimination models were compared with the results achieved by the individual technique separately. GC-IMS models showed better results compared to the FT-IR. However, integrating the data of the two techniques improved both the robustness of the resulting discrimination model, compared to models based on the GC-IMS and FT-IR individually.

In 2015, a study by Laroussi-Mezghani et al. (2015) proved the possibility of using the combination of geographical origins by PCA and SIMCA applied on fatty acid methyl esters (FAME) and NIR, which presented a high potential to identify the origin of Tunisian oils. With NIR data, samples from Chemlali Sfax, Chetoui, and Oueslati oils were discriminated with the correct classification of 89.55%, 92.53%, and 98.50%, respectively. With GC-FID data, better discrimination percentages were achieved for Chemlali Sfax (95.52%), Chetoui (94.77%), and Oueslati (99.25%). While all the imported oils (i.e. Algerian, French and Moroccan) were not distinguished. Overall, both fatty acids and NIR have the potential to classify the origin of Tunisian VOOs.

A study on geographical authentication of 82 EVOO from seven different countries was conducted by Quintanilla-Casas et al. (2020). HS-SPME-GC-MS was used to determine sesquiterpene hydrocarbons as targeted compounds (profiling) and non-targeted (fingerprinting) approaches to compare their effectiveness in the authentication of the geographical origin. After preprocessing, PCA and PLS-DA have been applied for both profiling and fingerprinting data individually. For the profiling approach, accurate classification was obtained for samples from Slovenia (100%), Turkey (100%) Morocco (93.3%), while samples from the other regions showed a non-satisfactory model, mainly in the sample of Croatia (45.5%). This could be attributed to the limited number of compounds that can be identified or tentatively identified according to their mass spectrum and linear retention index (Quintanilla-Casas et al., 2020). For the fingerprint approach, pretreatment and PCA were applied to the two-way unfolded matrix obtained (82 samples \times 22,203 variables), then PLS-DA was employed. In this case, the sample classification based on their origin was significantly enhanced compared to the profiling model. A 100% of correct discrimination was achieved for each of the seven countries of origin. Although accurate classification was obtained, the authors suggested further study with a large number of oil samples from various origins and different harvest years to establish a more robust and elaborated model.

In many cases, GC and HPLC methods are employed together to improve accurate, precise, and complete identification of the chemical compounds of oil (Table 1). In a study, fatty acids and phenolic composition were used to distinguish VOOs from six regions in Tunisian Coasts (Hlima, Ayed, Ennouri, & Smaoui, 2017). PCA and CA techniques were successfully verified the geographical origin of samples. According to this study, the fatty acid profile is highly associated with the production sites of VOOs, since samples from Gabes and Medenine are characterized by the best fatty acid profile and the highest oxidative stability whereas Sfax samples are characterized with the lowest oxidative stability and the worst composition in the context of fatty acids.

3.6.2. Liquid chromatography

It was also reported that phenolic profiles were used to identify the geographical origin of Arbequina EVOO obtained from southern Catalonia (Bakhouché et al., 2013). In this study, the phenolic composition of various oil samples was characterized by HPLC-ESI-TOF/MS. Discriminant analysis (DA) was applied to all phenolic compounds determined without variable selection. From the results of the DA classifier, it was clear the analyzed compounds might be used as a marker to distinguish these OOs according to their production area. Phenolic compounds measured by HPLC-ESI-IT-MS and HPLC-DAD were used to distinguish between Moroccan EVOO samples collected from three locations (Meknès, Ouazzane, and Tryout-Chiadma) (Bajoub, Ajal, Fernández-Gutiérrez, & Carrasco-Pancorbo, 2016). With the PCA technique, no clear classification based on geographical origin was observed. Consequently, the LDA technique was applied for a better classification rate. The forward stepwise variable selection procedure was employed to choose more relevant compounds for the correct classification of geographical origin. In this study, all phenolic compounds were used for classification except for oleuropein aglycone, ligstroside aglycone, oleuropein aglycone-isomer 4, ligstroside aglycone-isomer 2, ligstroside aglycone-isomer 3, (+)-pinoresinol, (+)-1-acetoxypinoresinol, tyrosols, *p*-coumaric, and quinic acid. The results reported in this study demonstrated the feasibility of correct classification (100%) of samples according to their production regions. Thus, the projected method could be easily applied to other OOs cultivation regions certified under geographical origin indication systems. Similarly, UHPLC-ESI-MS/MS has been used for the determination of phenolic compounds in 50 Spanish EVOO samples (Becerra-Herrera et al., 2018). PCA and LDA were applied successfully to discriminate the samples using the selected variables which include six polyphenols (tyrosols, hydroxytyrosol, 3,4-DHPEA-EDA, 3,4-DHPEA-EA, *p*-HPEA-EDA, pinoresinol, and total

phenolic compounds. The selected compounds used in LDA could be used as a fingerprint to classify OOs according to their geographical origin.

In a study, fingerprints from HPLC-CAD and high-temperature GC-FID, in both phases, were achieved. PCA was used as exploratory techniques, while SIMCA and PLS-DA were used as authentication techniques (Vera, Jiménez-Carvelo, Cuadros-Rodríguez, Ruisánchez, & Callao, 2019). Low and high-level data fusion approaches were used to enhance the discrimination models when the data obtained from individual techniques were separately used. The classification was accurate for the PLS-DA model with the low-level fusion of two procedures. Sensitivity and specificity were 100% in almost all classes and error was 0.0% for all classes. Therefore, the combination of data has confirmed that there is synergy between these methods, which give complementary information to distinguish between the classes considered. Although the results of developed models were very satisfactory, however, working with more than one sophisticated analytical tool involves an extra cost.

Nescatelli et al. (2014) investigated the potential of tracing the geographical origin of EVOOs by using HPLC/ESI-MS and HPLC-DAD (254 nm, 280 nm, and 340 nm) and PLS-DA. The combination of backward-interval-PLS and genetic algorithms (biPLS-GA) were used for selecting the most relevant area of the chromatogram (i.e. 280 nm and 340 nm), which provide 85 % of correct classification. Based on the results of BiPLS-GA and HPLC-MS, vanillic acid, *p*-coumaric acid, luteolin, pinoresinol, acetoxypinoresinol, apigenin, and metoxyluteolin are the main compounds that contributed to the authentication geographical origin of the EVOOs. Thus, HPLC-MS provided compounds that mainly contributed to the correct classification of Sabina PDO from other oils. To obtain a better classification model, mid-level data fusion was applied to the combined regions (280 nm and 340 nm), which allowed further to improve the specificity of the traceability model (90%) for PDO Sabina towards other oils (Nescatelli et al., 2014).

In 2018, the potential of phenolic acid and sterolic composition to distinguish between Tunisian and Italian EVOOs was investigated (Mohamed et al., 2018). In this study, a non-targeted metabolomics procedure using UHPLC-ESI/QTOF was performed for authentication of Spanish EVOO collected from different areas in Spain (Gil-Solsona et al., 2016). After the collection of 90 samples from 6 Spanish locations, the polar and nonpolar metabolites were extracted and extracts were kept at $-24\text{ }^{\circ}\text{C}$ until analysis. The proposed method identified 12 compounds in EVOOs. After identification of metabolites, PLS-DA and orthogonal PLS-DA (OPLS-DA) were applied to verify the geographical origin of samples. The results showed that the hydroxybenzoic acids and cholesterol derivatives were the main markers associated with geographic origin. OPLS-DA (90%) was selected as the best classification model due to its better recognition of EVOOs geographical origin. These results proved the feasibility of using untargeted fingerprints in the oils geographical authentication.

Two studies (Obisesan, Jiménez-Carvelo, Cuadros-Rodríguez, Ruisánchez, & Callao, 2017; Pérez-Castaño et al., 2015) compared their results obtained using liquid chromatography coupled to two detectors-ultraviolet (HPLC-UV) and charged aerosol (HPLC-CAD) combined with the chemometrics strategies, for testing the geographical origin of palm oil collected from the main palm-producing countries.

In the study of Obisesan et al. (2017), the high- and mid-level data fusion strategies were applied to the chromatographic data (obtained from 100 samples). For mid-level implementation, interval partial least squares and PCA were applied for variables selection. For high-level data fusion, fuzzy aggregation connective operators were applied to combine the PLS-DA attained from each chromatographic technique. The discrimination rates in this study were ranged between 73% and 98% for the single techniques and between 87%, 100%, 93%, and 100% for the mid- and high-level data fusion approaches, respectively. In the study of Pérez-Castaño et al. (2015), soft independent modeling by class analogy (SIMCA) and PLS-DA were used for developing the classification models. For HPLC-UV data, the SIMCA model has obtained the best classification

results, while for the HPLC-CAD data both of the classification models showed similar behavior. Comparison of the two previous studies showed that data-fusion approaches generally have better authentication ability than the individual models, which, indicates that the information attained from both chromatograms has a synergistic effect.

In another study carried by Ruiz-Samblás, Arrebola-Pascual, Tres, van Ruth, and Cuadros-Rodríguez (2013), they applied PLSR to TAGs measured by HPLC-CAD and high-temperature GC-MS for authentication of the geographical origin of palm oil samples collected from different continents. They found that the rates of success in the identification of the geographical origin of the 85 samples were ranged between 70% and 100%. Thus, these methods could be used as simple and rapid techniques for confirming the production area of palm oil and could be adapted to verify the labeling compliance of the oil.

Correct classifications for North Moroccan OO samples (Chefchaouane, Fès, Meknès, Ouazzane, Sefrou, Taouate, and Taza) was investigated by Bajoub, Medina-Rodríguez et al. (2016). Their study showed that correct classifications were obtained when PCA, LDA, SIMCA, and PLS-DA were applied to TAGs data. HPLC-RI data combined with chemometrics seem to be a powerful tool to verify the geographical origin of Moroccan OOs. Among the chemometrics applied, LDA and PLS-DA obtained the best models (in respect of accuracy for both training and test sets), while SIMCA gave satisfactory and correct geographical classification models. Based on the findings in this study, it can be concluded that the approach adopted in this work could be a useful method for traceability and verification of Moroccan PDO's VOOs (or for PODs from some other countries). Similarly, TAGs structures of Turkish hazelnut oils (collected from Giresun, Trabzon, Ordu, and Bolu) were successfully discriminated according to their geographical origin (Kıralan, Yorulmaz, Şimşek, & Tekin, 2015). The TAGs were analyzed using HPLC coupled with various detectors (including RI, light-scattering detectors (ELSD), and MS). The most important contributors in the classification were *sn-2* C18:1, *sn-2* C18:2, C18:2, OLL OOO, POO, *sn-2* C16:0 *sn-2* C18:1, and *sn-2* C18:2. Discriminant analysis clustered the samples according to their geographical source with the correct discrimination rate of 93.88 %. Among 50 samples, only three samples were misclassified; one sample from Ordu was clustered with Trabzon and two samples from Trabzon were misclassified with Giresun, whereas Giresun samples were discriminated correctly.

3.7. Inductively coupled plasma mass spectrometry (ICP-MS)

Multi-elements are the composition of oils that indicate the location where the plant is grown. The availability of trace elements was influenced by several factors such as soil, pH, humidity, and clay complexes (Kim & Thornton, 1993). Thus, the bioavailable elements indicated that soil could provide direct information regarding the production areas of oils. Previously, flame atomic absorption spectrometer (AAS) was the major analytical tool used in elements analysis in food. Recently, inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) were exclusively applied because of their capacity for multi-component analysis (Danezis, Tsagkaris, Camin, Brusica, & Georgiou, 2016; Laursen et al., 2014). Nowadays, several studies have shown the effectiveness of ICP-MS and ICP-AES in the authentication of the geographical origin of oils (Danezis, Tsagkaris, Camin et al., 2016; Gumus, Celenk, Tekin, Yurdakul, & Ertas, 2017).

In 2013, VOOs samples from four Western Greek regions (Zakynthos, Kefalonia, Lefkada, and Kerkyra) using trace elements and several physicochemical parameters coupled with the chemometrics were authenticated (Karabagias et al., 2013). LDA indicated that when various physicochemical parameters were pooled, accurate discrimination rates of 94.5% and 82.6% were attained using the original and cross-validation procedure, respectively. The results of this study indicated that various physicochemical parameters combined with the LDA model have the potential for classification of OOs based on geographical

origin even in a location where such origins are in proximity. Another study on tracing the geographical origin was conducted by Damak et al. (2019), who discriminated OOs from four Tunisian regions (Sfax, Monastir, Gafsa, and Medenine) by ICP-MS combined with LDA. Based on the stepwise LDA model, Fe, Mg, Sr, Rb, Mn, and Pb were the most elements that contributed to geographical confirmation. From this study, the discrimination of samples based on their origin via a multi-element fingerprinting method was demonstrated to be successful. LDA produced 92.1% and 87.3% discrimination and prediction rates, respectively; indicating that multi-elements are promising authentication markers of Tunisian OOs from various counties.

Beltrán, Sánchez-Astudillo, Aparicio, and García-González (2015) developed a method for testing the geographical origin of oils from southwestern Spain. Olive oils, olive pomaces, and soils were examined by ICP-MS. The results presented some correlation in the selection of elements in soils (W, Fe, Na), olive-pomace (W, Fe, Na, Mg, Mn, Ca, Ba, Li), and olive oils (W, Fe, Mg, Mn, Ca, Ba, Li, Bi), which enabled their utility in the authentication. In reference to OOs, 93% of them were classified according to their geographical origin using the LDA model. This result indicated that ICP-MS could be used as a powerful tool for conducting backward traceability of olive oils. However, further investigations are needed to acquire knowledge about what extent can the natural concentration of some elements be modified by utilizing fertilizers (with either foliar or soil application) and fungicides before generalizing the approach and its application as a possible standard for OOs authenticity in the future.

A discussion of the relationship between lanthanides analyzed by ICP-MS and EVOO from different geographical sources has been reported by (Aceto, Calà, Musso, Regalli, & Oddone, 2019). Lanthanides seem to be a unique marker for discrimination between EVOOs of Taggiasca cultivar from other Italian and foreign EVOOs. The LDA technique presented a 100% correct classification

Sayago, González-Domínguez, Beltrán, and Fernández-Recamales (2018) also successfully used multi-elements fingerprinting for testing the geographical origin of EVOOs. In this regard, the concentrations of 55 elements in 125 samples from various Spanish geographic locations were analyzed. PCA, PLS-DA, LDA, SVM, and RF were employed to develop discrimination methods and examined the relationship between the mineral content of oils and their geographical origin. The outcomes showed that the multi-element composition of Spanish EVOOs from three geographical origins (Atlantic coast, Mediterranean coast, and inland) enabled their geographical authentication. Furthermore, chemometrics presented high sensitivity and specificity, particularly when SVM and RF techniques were used, therefore indicating the advantage of these mathematical models in oil traceability and authenticity.

A study to evaluate the distribution of element traces in pumpkin seed and pumpkin seed oils in relation to the geographical origin (Austria, China and Russia) was reported by Bandoniène, Zettl, Meisel, and Maneiko (2013). ICP-MS combined with PCA successfully distinguished oils from various sources (Austria, China, and Russia). A preliminary study to discriminate eight Yemeni sesame oils according to their production areas from different provinces (Al Bayda, Marib, Ibb, Taiz, Abyan, Shabwa, Hadhramout, and Hodeida) using the concentration of minerals, quantified by ICP-OES was reported (Mohammed et al., 2018). In this study, PCA showed that P, Ca, Zn, Mg, Mn, and K, Cu, Cd, Pb, and Fe were the major elements that contributed to the declaration of the geographical origins of Yemeni Sesame Oil.

3.8. PTR-MS and SIFT-MS

Proton transfer reaction mass spectrometry (PTR-MS) and selected ion flow tube mass spectrometry (SIFT-MS) technologies utilize rapid flow-tube or drift-tube reactors combined with chemical ionization to measure trace compounds in a variety of media samples (such as air, food, cell, and bacterial cultures, etc.) and can achieve accurate determination even for compounds at very low concentrations (Smith &

Španěl, 2011). From a technical perspective, GC is the reference technique for the measurement of VOCs in food. However, there is growing interest to develop a simple and easy tool to overcome its shortcoming, as GC is still a time-consuming technique. Among the various analytical tools evaluated for rapid determination of VOCs in food is PTR-MS, which enable direct and highly sensitive on-line assessment of VOCs, with the capacity to detect numerous volatile organic compounds (VOCs) via proton-transfer-reaction with hydroxonium ions (H_3O^+) and identified by the mass spectrometer (Biasioli, Gasperi, Yeretian, & Märk, 2011). Comparing GC-MS or GC-FID (as reference method) with PTR-MS or in oil parallel with GC-FID can give information on the compounds that are associated with a particular PTR-MS peak and on probable inferences (Smith et al., 2011). PTR-MS technique has proved to be a potent tool for tracing the geographical origin of oil and fats (Biasioli et al., 2011). In recent years, Tres, Ruiz-Sambblas, van der Veer, and van Ruth (2013) investigated the potential use of VOCs measured by PTR-MS and fatty acid profile analyzed by GC-FID combined with the PLS-DA model for the classification of 94 crude palm oil samples obtained from South East Asia, South America, and Africa. GC-FID data presented three misclassified samples. PTR-MS models showed the highest classification models for East Asian, African, and American classes and the discrimination rates were 88%, 100%, and 100%, respectively. The results of the studies showed that the identification of geographical of crude palm oil is possible by GC-FID and PTR-MS. Since this is the first study that used PTR-MS for authentication of palm oil; further studies are required using a large sample size from different countries to validate the method and to increase its spatial specificity to country scale.

For the first time, the geographical origin of 130 VOOs collected from Greece, Italy, Morocco, and Spain were investigated by selected ion flow tube mass spectrometry (SIFT-MS) (Bajoub et al., 2018). PCA and PLS-DA were built on H_3O^+ , NO^+ , and O_2^+ data to discriminate OOs according to the country of origin and within the same country. The multi-class PLS-DA presented an excellent performance for fitting accuracy (98.90–100%) as well as for the prediction set (96.70–100%) and test set (97.30–100%). In this study, PLS-DA based on the Spanish oils presented 100% sensitivity, specificity, and accuracy in calibration, cross-validation, and external validation, while the model for Moroccan samples also exhibited a good model. The application of mathematical models demonstrated that the SIFT-MS could be used as a rapid and appropriate tool for routine analysis of origin-labeled olive oils. Similarly, Mourad Kharbach et al. (2018) compared the SIFT-MS fingerprinting to chemical data (including acidity, peroxide value, spectrophotometric indices, fatty acids, tocopherols and sterols composition) for geographic traceability of 95 EVOOs collected from various sources in Moroccan. PLS-DA, SIMCA, KNN, and SVM, were compared. Accurate models were developed from the chemical data as well as from the H_3O^+ and the NO^+ spectra using PLS-DA, SVM, and SIMCA. The SIFT-MS data in conjunction with the linear (SIMCA, PLS-DA, and KNN) and nonlinear (C-SVM and nu-SVM) multivariate techniques were also appropriate for verifying the geographical origin of EVOOs as a substitute for the chemical profiling.

3.9. Isotope ratio mass spectrometry

Isotope ratio mass spectrometry (IRMS) is a system that can discriminate chemical identical substances according to their isotope content (Brenna, Corso, Tobias, & Caimi, 1997). Mostly, the isotope profile of the ingredients of agricultural products (proteins, carbohydrates, minerals, and oils/fats) relies on several factors with the geographical origin being the major one, while others are associated with the production factors, namely fertilizer, feedingstuffs, seasonal differences and soil composition (Luykx & van Ruth, 2008).

Recently, the geographical origin verification of crude palm oil from different locations in Malaysia was investigated using a stable isotope. The bulk $\delta^2\text{H}$, $\delta^{18}\text{O}$, and $\delta^{13}\text{C}$ of oils were computed by HCA, PCA, and

OPLS-DA. In HCA and PCA techniques, the samples were clustered according to the corresponding state (Muhammad et al., 2018). $\delta^{18}\text{O}$ was identified as the main discriminatory marker for differentiating crude palm oil samples from different production areas. OPLS-DA provided a robust predictive model with an overall correct classification of about 71.43%. Although a satisfactory result was obtained, further work with large sample size is required to improve the accuracy of the predictive model. Gumus et al. (2017) used the combination of trace elements and stable carbon isotope ratios coupled with chemometrics, to verify the geographical origin of Turkish VOOs. The results showed that the ICP-MS and IRMS systems together with chemometrics might be an excellent tool for the authentication of the geographical origin of VOOs.

A comprehensive investigation of trace element contents and stable carbon isotope ratios of 49 VOOs from different sources in Turkey (İzmir, Manisa, Aydın, Muğla, Bursa, and Edremit Bay) was conducted using ICP-MS and EA-IRMS Gumus et al. (2017). Based on the results of PCA and HCA, Fe and Zn on the Manisa and $\delta^{13}\text{C}$ play an important role in the authentication of OOs according to the geographical origin. The authors concluded that this method could be employed to verify other geographical sites of OOs in Turkey too. Portarena, Baldacchini, and Brugnoli (2017) proposed methods based on the combination of IRMS and Resonant Raman (RR) coupled with the LDA model to identify the geographical origin of EVOOs from the Italian coasts (Apulia, Latium, Liguria, Molise, Sardinia, Sicily, Tuscany). The combination of IRMS and RR enabled correct identification (82%) of samples, whereas separate IRMS and RR analyses allowed the classification of only samples from Sicily and Latium, respectively. From this work, it is clear that the combination of data allows the use of synergy of the individual information to achieve an optimized model.

3.10. Spectroscopic techniques

In recent years, spectroscopic such as Nuclear Magnetic Resonance (NMR) near-infrared (NIR) and mid-infrared (MIR), Fourier transform spectroscopy (FTIR), UV-visible, fluorescence, and other spectroscopies have appeared as very effective and general tools for testing the geographical origin of foods (Li et al., 2020; Luykx & van Ruth, 2008; Tahir et al., 2020). These technologies are simple, rapid, non-destructive, and relatively low-cost analytical tools (Arslan et al., 2021; Tahir et al., 2019).

3.10.1. Vibrational spectroscopy

The combination of NIR and MIR techniques in conjunction with PLS-DA has been used for the classification of OOs (Bevilacqua, Bucci, Magri, Magri, & Marini, 2013). PLS-DA presented 100% discrimination rates in both cross-validation and external validation test-set, indicating a high level of confidence in the future results. The same authors applied the fusion of NIR and MIR together with the PLS-DA model to verify the geographical origin of Italian OOs (mainly from Sabina and other areas of Italy or Mediterranean countries) (Bevilacqua et al., 2013).

NIR and FT-IR techniques coupled with chemometrics were successfully used to distinguish Ligurian OOs from non-Ligurian OOs (Devos, Downey, & Duponchel, 2014). The GA was used as a pre-processing technique before the chemometrics application. NIR spectra and SVM provided better models (86.3%) as compared with the combination of NIR and PLAS-DA (82.8%). Regarding the MIR dataset, SVM presented the most accurate model (82.2%) as compared with the one achieved by PLS-DA (78.6%). Furthermore, this research indicated that even SVM techniques have to be established based on well-corrected spectral data to attain higher discrimination rates. Sánchez-López et al. (2016) developed the DA model with data obtained from the use of Raman spectroscopy to verify the geographical origin of Andalusian EVOOs; they concluded that the DA enabled correct classification of 89%.

3.10.2. UV–vis spectroscopy

UV–visible spectroscopy (UV–vis) coupled with multivariate analysis such as PCA, LDA, and PLS-DA is used in food authentication. Pizarro and co-authors mentioned examples of the possibility of authenticating EVOOs from Spanish regions (Aceite de La Rioja, Andalusia, Siurana, and Les Garrigues) based on their geographical origin using data fusion of UV–vis and chemical parameters (Pizarro, Rodríguez-Tecedor, Pérez-del-Notario, Esteban-Díez, & González-Sáiz, 2013). The spectra and quality descriptors were pretreated individually and then were merged to measure the effect of synergy on the information acquired from different methodologies. The results depicted the accurate classification between the defined groups after applying PLS-DA on the fused data, achieving 100% of correct discriminations and indicated a clear upgrading in the overall prediction rates (92.5%), thus demonstrating the influence of the synergy. This correct classification confirms the feasibility of the projected method and encourages the application of similar strategies based on UV–vis in OOs quality determination and authentication. In another study, UV-visible spectroscopy and quality parameters combined with PCA and OPLS-DA were applied to discriminate palm oils collected from three locations in Ondo State, Nigeria (North, Central, and South) (Jolayemi, Ajatta, & Adegeye, 2018). PCA score plots of both quality parameters and UV–visible data (250–550 nm) matrices exhibited geographical dissimilarity between the palm oil samples. Furthermore, OPLS-DA models presented over 95% and 85% correct classification for UV–visible and quality parameters, respectively. Although the chemometrics showed satisfactory results, the authors concluded that these results cannot be deemed exhaustive due to the smaller sample size (Jolayemi et al., 2018).

3.10.3. Fluorescence spectroscopy

Al Riza and co-authors evaluated the discriminatory power of front-face fluorescence spectroscopy combined with PCA to verify patchouli oil collected from seven Indonesian regions (including Masamba, Konawe, Kolaka, Bogor, Garut, Aceh, and Jambi) (Al Riza, Widodo, Purwanto, & Kondo, 2019). The results revealed the possible use of the face fluorescence technique as a supporting method for testing the geographical origin of patchouli oils. Similarly, the combination of fluorescence and NIR spectroscopy coupled with PLS-DA was used to discriminate individual Argentinean OOs from six registered designations of origin (RDO) (mainly from La Rioja, Mendoza, Catamarca, Córdoba, San Juan, and Buenos Aires) and unknown (RDO) (Jiménez-Carvelo, Lozano, & Olivieri, 2019). The authors found that the NIR correctly classified the oil samples by 100%, while fluorescence spectroscopy was not able to group the samples according to their origin. Several other studies on the use of spectroscopy to discriminate the geographical origin of oils can be found in the literature (Gertz, Gertz, Matthäus, & Willenberg, 2019; Ou et al., 2015). Discrimination of Maltese and non-Maltese EVOOs using 3D-Fluorescence spectroscopy combined with discriminant multi-way partial least squares regression (DN-PLSR) has been reported (Lia, Formosa, Zammit-Mangion, & Farrugia, 2020a; Lia, Formosa, Zammit-Mangion, & Farrugia, 2020b). DN-PLSR was correctly classified 98.58% of the original data and 93.18% of the cross-validated group. These results indicated that the DN-PLSR presented the discriminatory potential for confirming the authenticity of Maltese EVOO samples.

3.10.4. Laser-induced breakdown spectroscopy

Laser-induced breakdown spectroscopy (LIBS) is a semi-quantitative analytical tool suitable for rapid elemental analysis in a variety of fields such as biological (Gaudiuso et al., 2019) and analysis of minerals in geological research (Harmon, Russo, & Hark, 2013). Recently, few works on the use of the LIBS technique together with chemometrics for testing the geographical origin of OOs have been reported (Table 1). Most recently, Gazeli and co-authors used LIBS together with chemometrics to test the geographical origin of samples from different locations in Greece (Gazeli et al., 2020). The results showed that LDA,

support vector classifiers (SVC), and RF models revealed excellent classification rates that ranged between 90% and 99%; thus, they were able to discriminate samples from different places (namely, north, south, east, west, and central Rethymno). Similarly, Gyftokostas, Stefanis, and Couris (2020) applied the LIBS technique to OOs that were collected from various places in Greece, Rethymnon, Heraklion, and Lasithi. PCA-LDA model based on LIBS presented an excellent discrimination rate higher than 94%. From the results of mathematical models, it is evident that LIBS could be used as a powerful tool for the authentication of foods.

3.10.5. Nuclear Magnetic Resonance (NMR) spectroscopy

¹H NMR is a powerful tool for quantification of fatty acid profiles although mathematical models are required for quantification of total sums of saturated, monounsaturated, and polyunsaturated fatty acids from suitable signal intensities (Aparicio, Morales et al., 2013). As a result of signal overlap, this technique cannot quantify individual fatty acids with exception of linolenic acid (whose methyl protons are detected at δ 0.96). The NMR enables direct analysis of samples without any preparation or extraction process but it requires a large initial outlay and their procedure is time-consuming when compared with the GC method (Aparicio, Morales et al., 2013; Mannina & Segre, 2010). ¹H NMR spectroscopy combined with the LDA model was employed to verify the geographical origin of OOs with a discrimination rate higher than 85% both in calibration and in validation, with the predictive capability on the test set being about 90% (Ingallina et al., 2019). (Merchak et al., 2017) successfully employed NMR coupled with chemometrics to verify the geographical origin of green and black OOs samples (187) coming from across Lebanon at various altitudes and latitudes. Reports indicated that the NMR technique together with the LDA successfully discriminated the geographical origin of 44 vegetable oils which were collected from different botanical and geographical sources (Popescu et al., 2015). H-1-NMR in combination with PCA and CA (canonical analysis) were successfully used to classify OOs from Italy, Greece, and Spain based on their geographical origins (Winkelmänn & Kuchler, 2019). ¹H and ¹³C NMR coupled with multivariate techniques has shown to be powerful tools for the authentication of Maltese EVOOs (Lia, Vella, Mangion, & Farrugia, 2020). EVOOs were collected from various Italian locations (Tuscany, Sicily, and Apulia), Spain, Portugal, Tunisia, Turkey, Chile, and Australia. To verify their sources, NMR combined with PCA or OPLS-DA was used for classification (Rongai et al., 2017). Results demonstrated the potential use of NMR-based metabolic profiling together with chemometrics for clarifying the geographical origin of OO samples. NMR, GC-FID, and LC-DAD combined with chemometric techniques (LDA, SIMCA, PLS-DA, RF, ANNs, and SVM) were successfully used to distinguish Spanish OOs collected from Beas, Gibralforte, Niebla, and Sanlúcar de Guadiana. The developed models of these data presented obvious varietal differences with some extent of geographical origin (Sayago, González-Domínguez, Urbano, & Fernández-Recamales, 2019).

Chemometrics (PCA, CA, KNN) of NMR data presented clear discrimination between Italy, Greece, and Spain OO samples (Winkelmänn & Kuchler, 2019). The overall correct discrimination rate of the developed model was 96% for samples analyzed, demonstrating the suitability of the method for the purpose. By the use of ¹H NMR and IRMS data together with the RF technique allowed the classification of 177 Italian OO samples and 86 Tunisian samples, with an optimal correct classification rate of 98.5% (Camin et al., 2016). The discrimination of EVOOs produced in Turkey and Slovenia was investigated by Özdemir et al. (2018) using presaturation ¹H NMR. The NMR data were evaluated by PCA and PLS-DA. Aldehydes, phenolics, terpenes, and diacylglycerols were the key calcifiers. The authors demonstrated that this approach could be valuable for further declaration of the geographical origin of Turkish and Slovenian OOs.

A platform of fingerprinting tools, including ¹H NMR and IRMS, was applied to classify VOOs according to their geographical origin (Alonso-

Salces et al., 2015). PCA and PLS-DA were applied to evaluate the classification and recognition of important markers. Models based on IRMS data did not perfectly declare the geographical origin of VOOs. However, $\delta^2\text{H}$ and/or $\delta^{13}\text{C}$ data offered complementary geographical data to ^1H NMR data in the PLS-DA binary discrimination models.

3.10.6. Terahertz spectroscopy

Although many spectroscopies have been used in food quality control, little attention has been given to the utilization of terahertz spectroscopy (THzS). This is because it remains a challenge to reach a fast classification of oils according to their geographical origin in a non-destructive and efficient method. A few works based on THzS have been reported for the quantitative and qualitative analysis of edible oil quality (Liu et al., 2019; Yin, Tang, & Tong, 2016; Zhan, Xi, Zhao, Bao, & Xiao, 2016). However, up-to-date studies on the rapid identification of the geographical origin of oils are limited (Table 1). THzS technique combined with chemometrics such as least squares-support vector machines (LS-SVM), backpropagation neural network (BPNN), and RF combined with data reduction tools (PCA and GA) were applied to discriminate EVOOs from Australia, Spain, Greece, and Italy. The highest classification accuracy of 96.25% for the prediction set was achieved by the combination of LS-SVM and GA. It was concluded that THz spectroscopy is an effective rapid method for the classification of EVOOs (Liu et al., 2018).

3.11. An electronic nose (e-nose) and electronic tongue (e-tongue)

The feasibility of flash gas chromatography electronic nose (FGC e-nose) and chemometrics to discriminate Italian EVOOs from different geographical areas declared on the labels were studied (Melucci et al., 2016). Results exhibited that the applied technique is appropriate to affirm the geographical origin based on PCA and DA analyses applied to the volatile compounds. The selected variables were appropriate for discriminating Italian oil from non-Italian samples. PLS-DA also permitted the prediction of the degree of membership of unknown samples to the classes studied. Recently, the use of artificial nose was compared with the NIR, and UV-visible spectroscopy to classify OOs from the entire Chianti Classico production area and other Italian PDO areas (Forina et al., 2015). The data obtained from the three technologies were employed both separately and in combination, after integration of the respective variables, in order to develop a model for the Chianti Classico PDO olive oil authentication. From this study, it can be concluded that the NIR technique is the most potent analytical tool, while the artificial nose although provides complementary information, appeared to be less informative, and besides this system is costly (Forina et al., 2015).

In another study, electronic nose and tongue have been employed for authentication of 25 Moroccan VOO samples collected from various VOOs, but not clear classification could be obtained when the two instruments were used separately. Therefore, low-level data fusion was applied to combine electronic nose and tongue data. SVM model on the combined data correctly discriminated samples based on their geographical sources. Overall, the model based on data fusion results outperformed those based on the electronic nose and tongue alone. Electronic Tongue combined with the multivariate analysis was also employed for the rapid testing of the geographical origin of Tunisian OO samples (Souayah et al., 2017).

The combination of e-nose and e-tongue was used to discriminate Moroccan VOOs collected from five different locations (i.e. Ouarzazate, Ouazzane, Taounate, Mrir't, and Sidi Ali). The results showed how the results achieved with the combination of PCA, CA, and SVM and low-level abstraction were good enough to define the geographical origin of Moroccan VOOs (Haddi et al., 2013).

3.12. Differential scanning calorimetry

The geographical origin of four cultivars (Koroneiki, Lianolia, Adramitiani, and Thasitiki) of EVOOs coming from four regions in Greece (mainly from Crete, Peloponnesus, Ionian Sea, and the Aegean Sea) was subjected to Differential Scanning Calorimetry (DSC) combined with LDA model (Chatziantoniou, Triantafyllou, Karayannakidis, & Diamantopoulos, 2014). The results based on LDA demonstrated the potential use of DSC for testing the geographical origin of oils from different regions with a correct classification of 88.5%. Furthermore, the results of Koroneiki cultivars from different production areas were accurately (90.5%) discriminated according to their geographical origin. In a study, Mallamace et al. (2017) applied DSC-PCA analysis to discriminate between 39 organic OOs obtained from different regions of the world. Authors investigated the oil melting parameters (temperature and enthalpies of characteristic DSC peaks) by using seven principal components (PCs), which covered about 98% of the total variance using a very small quantity of samples (5–10 mg) and short analysis time (20–30 min). DSC-PCA coupled with the chemometrics techniques were also successfully classified EVOOs based on their geographical origin (Italy, Tunisia, USA, Turkey, Albania, Greece, Andalusia, and Spain) (Mallamace et al., 2017).

4. Conclusions and future perspectives

Identification of the geographical origin of edible oils is of great value for legislation and consumers worldwide. This review has pointed out how a wide type of analytical tools ranging from classical spectroscopy (such ICP-MS and NMR) to less frequently used tools (such LIBS, DSC, e-nose, and e-tongue, PTR-MS, SIFT-MS, Raman, and THzS), without exclusion of the popular separation techniques (HPLC and GC) have been combined with chemometrics to achieve the objectives listed earlier. Chromatographic, ICP-MS and ICP-AES are the main approaches for oil geographical verification. Spectroscopic techniques is frontline tools rapidly substituting other approaches in many fields of food science. These trends extend to edible oil authentication, due to the multi-analyte abilities of these techniques. Multi-analyte capabilities are important aspects for food authentication researches since they give more measurements and thus, enable rapid classification. From this review, it is clear that PCA, PLS-DA and LDA are the most chemometrics approaches used while RF, SVM, LS-SVM, ANNs, KNN, Fuzzy ID3, C4.5, and SVC were scarcely used to authenticate the geographical of oil although their advantages have demonstrated and obtained good classification models. Overall, this review is an important source of information for food analysts that would like to know different analytical tools used for oil geographical authentication.

The future of geographical authentication of edible oil critically depends on combining multivariate tools in processing and interpreting the data acquired from various analytical techniques. Outcomes of this review suggest that in the future, the relative scientific group will build a fingerprint (e.g. spectrum, chromatogram, multi-element) database to harmonize analytical tools and to develop a worldwide system of the geographical declaration of oils should carry out collaborative research work. Such types of research collaboration could help the inclusion of chemometric combinations in official regulations and scaled up to meet routine analytical requirements.

Declaration of Competing Interest

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

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