Corrigendum to “Quality assessment and geographical origin authentication of extra-virgin olive oils imported into China” [J. Food Compos. Anal. 113 (2022) 104713]

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The authors regret that the printed version of the above article contained a number of errors. The correct and final version follows. The authors would like to apologise for any inconvenience caused.

Major corrigendum: The authors of article, affiliations of authors, Section 2.2 and CRediT authorship contribution statement were corrected.

Corrigendum the authors of article: The authors of article were changed.

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Corrigendum Section 2.2: The first sentence was changed into “A total of 85 commercial EVOO samples were collected from Chinese ecommerce platforms”.

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Xue Li: Methodology, Formal analysis, Investigation, Writing – original draft, Funding acquisition. Yu Zhang: Data curation. Zhi Liu: Supervision. Wei Wang: Conceptualization, Supervision, Funding acquisition. Sulin Sun: Validation. Junhong Wang: Resources. Zuoyi Zhu: Validation. Hua Yan: Project administration. Shenlong Zhu: Funding acquisition. Erli Niu: Funding acquisition. Romero Agusti: Supervision, Writing – review & editing

Abstract

Extra-virgin olive oil (EVOO) is an important imported commercial product in China. This study comprehensively evaluated the quality of EVOOs imported from different countries into China based on chemical parameters and sensory attributes. Multivariate statistics were used to authenticate their geographical origins. 71.8% of the oils failed to meet the current official standards at inland and abroad established for commercial EVOO category. Pyropheophytin, defect attributes, peroxide value, and K_{270} were above the limits in 32%, 24%, 23%, and 5% of the failed samples respectively, while free acidity, K_{232} and delta K were above the limits in 4%. Fatty acids and delta ECN42 were beyond the limits in 2% of the failed samples. Linear discriminant analysis (LDA) and orthogonal partial least squares discriminant analysis (OPLS-DA) based on fatty acids and triacyglycerols succeeded in identifying the origin of olive oils. 3.5% of the total might be mislabelled the geographical origin. The main quality problems of the analyzed oils include oxidation, counterfeiting with refined oils and origin fraud. Considering the results and the fact that most EVOOs in China are imported from other countries, chemometrics combined with critical quality attributes should be an ideal way of EVOO quality control, protecting consumers from frauds.

Keywords: EVOOs; Fatty acids; Triacyglycerols; Food quality; Origin authenticity; Chemometrics

1. Introduction

In recent years, demand for quality assurance of extra virgin olive oil (EVOO) has grown dramatically all over the world. In China, this demand is especially strong, since EVOO is an important import commodity in the domestic market. According to data released by the General Administration of Customs of China, the country imported approximately 45000 tons of olive oil in 2016, and the import volume increased rapidly by 60% to 70% over the following two years (Deng, 2018; Wang, Zhang, Farooqi, Ma, Yu, & Jia, 2017). Increasing interest in EVOO is not only due to its pleasant flavour, but also attributed to its high nutritional value and health benefits. Over the years, EVOO has been explored against cardiovascular disease, cancers, diabetes mellitus, and liver damage (Foscolou, Critselis, & Panagiotakos, 2018; Soto-Alarcon, Valenzuela, Valenzuela, & Videla, 2018). The health benefits of EVOO are mainly correlated to its chemical composition, particularly, to the presence of monounsaturated fatty acids, mainly oleic acid (55-83%), polyunsaturated fatty acids, and several minor components (1-2%), represented by squalene, triterpenes, sterols, tocopherols, pigments and phenolic compounds (Criado-Navarro, Ledesma-Escobar, Parrado-Martínez, Marchal-López, Olmo-Peinado, Espejo-Calvo, et al., 2022; Soto-Alarcon, Valenzuela, Valenzuela, & Videla, 2018).

Commercial olive oil can be classified as EVOO, virgin olive oil, and lampante olive oil according to physical, chemical and sensory properties (Gerhardt, Schwolow, Rohn, Pérez-Cacho, Galán-Soldevilla, Arce, et al., 2019). Generally, EVOO costs more due to its great nutritional value and limited production. Thus, it is the higher price that makes it difficult to root out intentional adulteration of EVOO. According to the data

from International Food Standard in 2020, olive oil was one of the main culprits for mislabeling in the edible oils market around the world (https://mp.weixin.qq.com/s/TkeMjLAReHDcBMcjMH2IfQ). The booming import trade leads to EVOOs from numerous origins flooding into the Chinese domestic market. Possible fraud and adulteration associated with EVOOs have raised increasing concern among Chinese consumers. Therefore, quality control of the commercial imported EVOOs is critical to combat the illegal attempts of commercial fraud and protect the legitimate interests of consumers. It must be considered, as well, those EVOOs that lose their category by hydrolytic degradation and lipid oxidation due to the inadequate storage conditions. Several studies have proved that improper temperature, light, or oxygen conditions could influence the quality of the oil and change the compounds profiles responsible for its flavor (Caipo, Sandoval, Sepúlveda, Fuentes, Valenzuela, Metherel, et al., 2021; El Yamani, Sakar, Boussakouran, & Rharrabti, 2022; Zaroual, Chèné, Mestafa El Hadrami, & Karoui, 2022). However, by now a study on evaluating the quality of commercial imported EVOOs in the Chinese market is still missing.

It is tricky to develop a reliable means of examining the characterization of olive oil since the products are variational depending on the cultivar, climatic conditions, geographic origins, olive processing conditions, and storage conditions (Bajoub, Medina-Rodríguez, Gómez-Romero, Ajal, Bagur-González, Fernández-Gutiérrez, et al., 2017; Li, Zhu, Shoemaker, & Wang, 2014; Lukić, Žanetić, Jukić Špika, Lukić, Koprivnjak, & Brkić Bubola, 2017; S. Portarena, Farinelli, Lauteri, Famiani, Esti, &

71 Brugnoli, 2015). Presently, the classification of olive oil is distinguished on the basis of
72 the limit values of some quality indices (chemical parameters and sensory attributes)
73 proposed by European Community Regulation (EC Regulation 2568/91), the trade
74 standard of International Olive Council (COI/T. 15/NC No 3/Rev. 15), and the National
75 Standard of People's Republic of China (GB/T 23347-2009). In most cases, however,
76 it is difficult to process the data or capture the subtle differences between authentic and
77 adulterated samples (Gómez-Caravaca, Maggio, & Cerretani, 2016). Meanwhile, these
78 measures alone are obviously not enough to support the certification and are unable to
79 reveal whether the product is mislabeled (ensure its quality and genuineness). In this
80 context, the current inspection procedures for EVOO need to be improved and
81 supported by novel methods.

82 Recently, geographic origin authentication has become an promising tool to ensure
83 olive oil quality due to the fact that the growing area has a significant effect on the
84 unique characteristics of olive oil (Al Riza, Kondo, Rotich, Perone, & Giametta, 2021).
85 Therefore, despite existing routine methods for quality control (IOC, 2019),
86 geographical characterization of EVOO remains a great attraction. The methodology is
87 mainly based on analysis of the chemical components that might be used for
88 discrimination of olive oils from different geographical regions (Cecchi, et al., 2020).
89 In particular, fatty acid composition and triacylglycerols profile have been successfully
90 used in analysis of geographical origins (Ollivier, Artaud, Pinatel, Durbec, & Guérère,
91 2006; Peršurić, Saftić, Mašek, & Kraljević Pavelić, 2018). Meanwhile, chemometrics
92 coupled with different analytical instruments is the most widely used approach to

93 verifying the geographical origins of olive oil by determining the chemical information
94 (Al Riza, Kondo, Rotich, Perone, & Giametta, 2021; Peršurić, Saftić, Mašek, &
95 Kraljević Pavelić, 2018; Ruisánchez, Jiménez-Carvelo, & Callao, 2021). The main
96 advantage of chemometrics is that it makes possible coping with vast amounts of data
97 and guarantees authenticity by detecting the intrinsic quality parameters of olive oil.
98 Chemometrics, as a well-known analytic tool based on proper statistical and
99 mathematical approaches, mainly consists of the application of unsupervised and
100 supervised pattern recognition techniques (Pérez-Castaño, Medina-Rodríguez, &
101 Bagur-González, 2019). Thus, a chemometrics-based approach, combined with the
102 main quality indices, may provide information about the quality and authenticity of
103 EVOOs. Nonetheless, chemometrics has not yet been accepted as a reliable tool by
104 national and international laws and regulations for EVOO data treatment and quality
105 assessment. Under such circumstances, the significant effect of chemometrics on olive
106 oil quality control should be highlighted.

107 Based on these premises, the aim of this study was to evaluate the quality of the
108 imported EVOOs marketed in China by using chemical parameters and sensory
109 attributes, and to verify the geographical origins of oils samples by chemometrics. For
110 this purpose, we collected commercial imported products from a wide range of
111 geographical origins including the main worldwide producers (Spain, Greece, Italy,
112 Tunisia, Turkey and Australia). And then, a wider range of olive oil quality parameters
113 were determined, such as sensory attributes, conventional commercial quality
114 parameters (acidity, peroxide value, Pyropheophytins, and UV coefficients), the main

chemical compounds (fatty acid composition and triacyglycerols) and delta ECN42. Furthermore, exploratory data analysis was performed on a dataset containing chemical compositions by using two unsupervised pattern recognitions, PCA and a cluster heat-map, and two supervised pattern recognitions, LDA and OPLS-DA. To the best of our knowledge, this is by far the first study that employed quality parameters combined with multiple pattern recognitions for the evaluation of the quality and traceability of the imported EVOOs in the Chinese market.

2. Materials and methods

2.1 Materials and reagents

19 fatty acid methyl ester (FAME) standards and triolein (purity≥99%) were purchased from NU-CHEK (MN, USA). Propan-2-ol, potassium hydroxide, phenolphthalein, isooctane, glacial acetic acid, potassium iodide, sodium thiosulfate, starch and cyclohexane were all analytical grade. Acetone, acetonitrile, methanol, hexane, and heptane were HPLC grade (Merck, Germany). Water (resistivity above 18MΩ • cm) was obtained from a FDY1001-UV-P water system (Fulham Technology Co., Ltd, Qingdao, China). Silica gel cartridges (1 g, 6 mL) were from Agela (Tianjin, China).

2.2 Sampling

A total of 85 commercial EVOO samples were provided by Qianjiang customs of the People's Republic of China (Hangzhou, China) between March and May in 2018. According to the description on the label, the olive oil samples, which were limited the best before date to 24 months after bottling, were collected between 2016 and 2018

crop seasons and their production dates were from June 2016 to March 2018. The samples were from Spain (‘S’, 18), Greece (‘G’, 16), Italy (‘I’, 13), Turkey (‘Tr’, 14), Tunisia (‘Tn’, 13), and Australia (‘A’, 11). Of these samples, 18 samples were labeled as products of protected designation of origin and 9 samples were organic products. At least three brands were selected from each country, and at least two bottles of olive oil were purchased for each brand. Meanwhile, some samples from the same brand but different production years and batches were also collected. The detailed information of oil samples was shown in Table 2. All samples were stored at below 4℃ and away from light. Before analysis, the samples were brought to room temperature, which was kept at 20℃. All the parameters of the samples were analyzed within their shelf life.

2.3 Chemical quality analysis

Classical quality indices such as free acidity (FA), peroxide value (PV) and specific UV extinction coefficients (K_{232}, K_{270}, and delta K) were determined according to ISO and IOC methods (AOCS, 2009; IOC, 2015a; ISO, 2017).

Triacyglycerols (TAGs) were analyzed according to IOC method (IOC, 2010). TAGs in olive oils were separated according to equivalent carbon number (ECN), defined as CN-2n, where CN is the total acyl carbon number and n is the number of double bonds of fatty acids (Ollivier, Artaud, Pinatel, Durbec, & Guérère, 2006). IOC method was used to analyze the absolute difference between the experimental values of triacyglycerols (TAGs) and the theoretical values of TAGs (delta ECN42) (IOC, 2010).

Pyropheophytin (PPP) was determined according to ISO 29841-2009 (ISO, 2009). The results were expressed as the ratio (%) of pyropheophytin *a* to the sum of

159 pheophytin *a*, *a'* and pyropheophytin *a*.

160 For fatty acid composition (FAC), IOC method (IOC, 2015b) was used with some
161 modifications. Fatty acid methyl esters were obtained through the reaction of 50 mg of
162 oil dissolved in heptane (2 mL) with 2N potassium hydroxide in methanol (0.2 mL) and
163 then were analysed by gas chromatography coupled with flame ionization detection
164 (GC-FID, Thermo, USA). The column was DB-FFAP (30 m×0.32 mm×0.5μm i.d.,
165 Agilent). The GC oven temperature program was set at 170℃ for 1 min, and then
166 increased at a rate of 10℃/min to 220℃. Nitrogen was supplied as the carrier gas and
167 the flow was set at 2.0 L/min. The injection and detector temperatures were set at 270℃.
168 The injection mode was split (1:50) and injection volume was 0.5 μL.

169 **2.4 Sensory analysis**

170 Organoleptic analysis used for classification of samples was performed by a panel
171 in accordance to COI/T.20/Doc.No15. Each taster evaluated the odour and taste
172 attributes, quantifying the intensity of each negative and positive attribute on the 10 cm
173 scale from 0 (no perception) to 10 (the highest intensity). The oil was graded by
174 comparing the median value of the defects and the median for the fruity attribute
175 (EVOO, the median of the defects is 0 and the median for "fruity" is > 0).

176 **2.5 Statistical analysis**

177 One-way analysis of variance (ANOVA) was performed by GraphPad prism (ver.
178 5, GraphPad Software®, USA) to find whether the difference of each quality parameter
179 was significant at 95% confidence level ($p < 0.05$). Then, multi-variate statistic method
180 was applied to further interpret the difference of chemical composition of samples of

different classes (origins). Cluster heat-map was performed using MeV (ver. 4.0, TIGR®, USA). Linear discriminant analysis (LDA) was performed using SPSS (ver. 18.0, IBM®, USA). Principal component analysis (PCA) and orthogonal partial least squares discrimination analysis (OPLS-DA) were run on SIMCA-p (ver. 13.0, Umetrics®, Sweden).

3. Results and discussion

3.1 Quality of the EVOO samples

The European Commission Implementing Regulation (EUReg.No1348/2013), International Olive Council (COI/T.15/NC No 3/ Rev.15) and Chinese standard GB/T 23347-2009 defined a decision tree for verifying whether an extra virgin olive oil is consistent with the category declared, and the quality criteria that must be checked by analysts are: FA, PV, specific extinctions in UV, etc. (Grossi, Palagano, Bendini, Riccò, Servili, García-González, et al., 2019). Table 1 presents the values obtained for the main parameters, their tolerance limits by the domestic and international standards for commercial EVOO, and the number of samples that exceeded the limits.

3.1.1 EVOO quality parameters

As the results shown, FA detected in these samples ranged from 0.14 % to 1.26 %. In particular, 5 samples from Turkey exceeded the maximum limit of 0.8 % for extra virgin category. Meanwhile, in terms of the value of FA, 26 samples were in disagreement with the descriptions on their respective labels. As for PV, the range was 4.23 meq O_2/kg oil to 16.31 meq O_2/kg oil. 30 of all the samples were above the limit of 10.0 meq O_2/kg which was the maximum value set by Chinese standard GB/T 23347-

2009 for olive oil classified as EVOO. Howerer, if UE and IOC standards were adopted, no sample exceeded the limit value of 20 meq O_2/kg.

According to the regulation (GB/T 23347-2009), the maximum values of absorption at wavelength 232 nm (K_{232}) and 270 nm (K_{270}) for commercial EVOO category are 2.5 and 0.22, respectively. The results obtained for K_{232} values ranged from 1.10 to 2.71, and 5 samples presented values higher than the set limit, suggesting that they started the primary oxidation process prior to peroxides formation. K_{270} values of the 85 samples ranged between 0.06 and 0.80. Among all samples, 7 presented higher results of the value than required in the regulation, indicating that they were in the propagation step of the oxidative process, during which peroxides broke to produce secondary compounds that had absorbtion at 270 nm. The delta K values of all oil samples ranged between 0 and 0.09, and 5 samples exceeded the limit, namely 0.01 set by the regulation (GB/T 23347-2009).

3.1.2 Pyropheophytin A and detal ECN42

The ranges of PPP in the investigated samples were between 2.14% and 43.80%. According to the regulations of CDFA, SA, and SANS, the minimum value of PPP accepted for EVOOs is 17% (Aparicio-Ruiz, Romero, García-González, Oliver-Pozo, & Aparicio, 2017), and 41 samples had a PPP content above the limit. Therefore, we verified that 48.2% of all the samples would not be qualified as EVOOs if only PPPs were taken into account. It should be noted that the value of PPP follows a curve with time increasing gradually to reach a maximum and then began decreasing due to PPP degradation and lost its characteristics as pigment (Aparicio-Ruiz, Romero, García-

González, Oliver-Pozo, & Aparicio, 2017). Thus, high PPP value represents an aged oil whereas a low value must be interpreted together with other oxidative descriptors such as K_{270} absorbance in order to identify in which part of the PPP curve of the sample is placed.

According to the regulations of GB/T 23347-2009, the maximum value for EVOOs concerning delta ECN42 is 0.2. As shown in Table 1, delta ECN42 values ranged from 0.0003 to 0.2221. Two samples from Tunisia presented values higher than the established limit for EVOO, which indicated that the two samples could not be genuine.

3.1.3 Fatty acid profile

Fatty acid composition is a main nutritional feature for EVOO. The main fatty acid was oleic acid (C18:1n-9), a monounsaturated omega-9 fatty acid accounting for 55-83% of the total oil composition. Moreover, EVOO contains some other fatty acids, like palmitic acid (C16:0), palmitoleic acid (C16:1), stearic (C18:0), linoleic acid (C18:2n-6), and α-linolenic acid (C18:3n-3). The regulations of China and IOC have presented the ranges of fatty acids in EVOO. As shown in Table 1, two samples of Spain were beyond the limit range (3.5-21.0) for C18:2. However, it should be noted that some olive cultivars show particular fatty acid profiles out of the official limits, as is the case with 'Aguilar' from Spain that is typically poor of linoleic acid (Ruiz-Domínguez, Raigón, & Prohens, 2013); thus, with just one anomalous value, the sample needs further analysis before declaring as a counterfeit.

3.1.4 Sensory analysis

247 Sensory notes of olive oil are considered essential to consumers' approval. The
248 olfactory test of oil samples was performed by a panel consisted of five staffs from a
249 research group from Zhejiang Academy of Agricultural Sciences, China. It must be
250 pointed out that the official regulation requires 8-12 tasters to set up a panel whereas
251 for this study only five trained tasters were available. Thus, results about sensorial
252 analysis are only estimation. However, for defective samples the median has been
253 estimated provided eight tasters would have tasted them and three of them did not detect
254 the defect (three zeros were added to the five observed results). The results from sensory
255 analysis performed on the oil samples are reported in Table 2. 31 samples did not result
256 to comply with the quality level requested for EVOO due to a higher median than 0 for
257 negative attribute. Rancid and fusty odors were the main defects of these samples, while
258 some others had musty or vinegary attributes. It is worth noting that 28 samples should
259 have defects median value over zero even if three blank tasters were added to the matrix.

260 **3.1.5 The unconformity**

261 According to the Chinese standard GB/T 23347-2009, the total, qualified and
262 failed numbers of EVOO samples are shown in Fig 1(A). For the 18 samples labeled
263 from Spain, 3 samples failed to comply with the standards due to PV and C18:2. For
264 the 16 Greek samples, 5 samples failed due to high PV and specific extinction (K_{270}).
265 All the Turkish 14 samples failed due to high FA, PV, and specific extinction (K_{232}, K_{270},
266 delta K). 11 out of the 13 Tunisian samples could not be defined as EVOO due to higher
267 values of PV and delta ECN42 than the established limits. For the 13 Italian samples, 6
268 samples failed due to high PV, and specific extinction (K_{232}, K_{270}). 1 of the 11 Australian

samples could not be labeled as EVOO due to high PV. Therefore, of the 85 oil samples investigated in the study, only 45 samples, 52.94% of the total, conformed to the standards for commercial EVOO. If only the IOC regulation limits are taken into consideration (with PV limit lower than 20 meq O_2/kg and sensory analysis) a total of 32 samples failed (IOC, 2019). If PPP limit, the IOC regulation limits and the Chinese standard serve together as the benchmark, the number of the sub-quality samples stood at 61 (71.8%). It also should be noted that the oil samples with different production dates have different oxidation levels. In this study, the number of samples in 2016, 2017 and 2018 was 14, 60 and 11 respectively, while the number of samples failed to comply with the domestic and international standards were 12 (85.7%), 48 (80.0%) and 3 (27.3%) respectively, due to high PPP and PV. The results were listed in Table 1. The closer to the expiration date, the higher oxidation degree and percentage not conforming to standards of the oil sample were obtained..

Fig 1(B) shows the contribution percentage of each parameter to the unconformity among all the investigated samples. PPP was the largest amount factor of unconformity to oil samples at 32%, followed by sensory results at 24%, PV at 23%, K_{270} at 5%, FA, K_{232} and delta K at 4% and the other parameters were at 2%.

In this study, among the 41 samples PPP exceeded the limit, 27 samples had negative attributes, and PV values of 15 samples were exceeded the limit of 10.0 meq O_2/kg. PPP is a natural compound resulting from the degradation of pheophytins by acid condition and heat treatments, with its extent of formation depending on the intensity of the treatment (Li, Zhu, Shoemaker, & Wang, 2014). Sensory attributes of

olive oil include flavours and off-flavours which originate by different mechanisms: positive odours (fruity, green or ripe etc.) are mainly due to a combination of volatile compounds that are produced enzymatically by the lipoxygenase pathway, the main defects may be caused by sugar fermentation (winey-vinegary), amino acid conversion (fusty), enzymatic activities of moulds (musty), and to auto-oxidative processes (rancid) (Barbieri, Aparicio-Ruiz, Brkic Bubola, Bucar-Miklavcic, Lacoste, Tibet, et al., 2021). Both of these two parameters were good indicators for overall olive oil quality and freshness as well as storage history. Peroxide value is a major indicator of olive oil quality. It indicates early stages of oxidation, which is related to storage conditions (oxygen, light exposure and temperature) of olive oil after production (Grossi, Di Lecce, Arru, Gallina Toschi, & Riccò, 2015). A high peroxide value indicates preservation issues, though inaccurate extraction in the mill or low quality olives can produce high PV as well (Kamikata, Vicente, Arisseto-Bragotto, Miguel, Milani, & Tfouni, 2019). The high PPP, median of defects and PV means ageing and oxidation are the main problems of these samples.

In addition, it should be note that 5 samples had very high values of K_{270} (0.73 ~ 0.80), delta K (0.08 ~ 0.09) and K_{232} (2.42 ~2.71). The values of PPP and MeD (rancid) of these samples also exceeded the limits. This particular combination of UV values suggests a counterfeit with refined oils that can produce conjugated diene and trienes as well as high delta K values. Maximum absorptivity in specific extinction (K_{232} and K_{270}) can be due to improper storage or an energetic refining process (Aued-Pimentel, Takemoto, Kumagai, & Cano, 2008; Jabeur, Zribi, & Bouaziz, 2016). K_{232} value

indicates primary oil oxidation rate, which is related to the formation of unsaturated fatty acid hydroperoxides, conjugated dienes and carboxylic compounds (da Silveira, Vágula, de Lima Figueiredo, Claus, Galuch, Santos Junior, et al., 2017). K_{270} value is a marker for the secondary oil oxidation step that releases breaked compounds from the hydroperoxides that absorbs at 270 nm; in addition, it is associated with the formation of conjugated trienes during the refining process (da Silveira, et al., 2017). Delta-K (in absolute value) higher than legal limit suggests illegal blending with refined oils, since the refining process can produce conjugated dienes and trienes that are very active within the 268 to 272 nm, which lead to significant drift for delta-K.

According to the above results, these imported olive oil samples investigated in this study mainly have the following quality problems: different degrees of oxidation and counterfeit with refined oils. The oxidation of oils is initiated by the formation of unstable hydroperoxides through triacylglycerol fatty acid reactions with molecular oxygen, and stimulated by free fatty acids, mono and diacylglycerols and thermally-oxidized compounds. The unstable hydroperoxides degradation allowed generation of volatile and non-volatile substances that are responsible for VOO defect or its oxidative rancidity (Cecchi, Migliorini, Giambanelli, Rossetti, Cane, & Mulinacci, 2019). They also can cause the values of quality indices of olive oil (FFA, PV, specific extinction coefficients, and PPP, etc.) to rise. These reactions are catalyzed by metal traces, exposure to light and temperature increase during storage after production.

3.2 Discrimination of olive oil for origin authenticity

Over the last decades, consumers are increasingly concerned about the olive oil's

origin. However, the former can not verified the products' origins due to the asymmetric information about products arising between producers and consumers. Therefore, in all European countries, a mandatory labeling information requiring producers to indicate on the label the nature of origin of the EVOO has been introduced by Regulation No 29/2012 and Regulation (EU) No. 1151/2012 (Bimbo, Roselli, Carlucci, & Gennaro, 2020; Calò, Girelli, Wang, & Fanizzi, 2022). Despite mandatory geographical origin labelling of EVOOs in Europe, the indications for this issue are different in China. According to the Chinese standards GB 7718-2011 (China, 2011), the label must include information about the country of origin, but some Chinese importers may indicate country of origin as the location of blending and bottling rather than that pressing or growing. Therefore, the products labeled as 'Product of Italy' may be bottled in Italy, but the olive oils from different geographic origins such as Spain. For this reason, identification of the geographical origin of olive oil is critical for tracing the production and supply chain, which can avoid counterfeit and adulterated conducts of high-price EVOO products in its complex industrial chain and protect its commercial brand value. FAC and TAGs were considered to be useful for identifying the geographical origin of olive oil (Fuentes, Paucar, Tapia, Ortiz, Jimenez, & Romero, 2018; Peršurić, Saftić, Mašek, & Kraljević Pavelić, 2018; Silvia Portarena, Leonardi, Scartazza, Lauteri, Baldacchini, Farinelli, et al., 2019). Therefore, multivariate statistical methods based on the descriptive variables (FAC and TAGs) were applied according to their origins for samples classification. The classification of oil samples in this work was based on their declared origin on the labels.

3.2.1 PCA and cluster heat map

Two unsupervised classification methods, i.e., PCA and cluster heat map, were performed to identify the intrinsic tendency for grouping and to obtain the similarities among the samples from six countries (Spain, Greece, Italy, Turkey, Tunisia and Australia). Data matrix X used for modeling included the contents of fatty acids and triglycerides of 85 samples, which represent one of the main qualities of EVOO and describe the main constituents present in the sample. The first 2 components (PC1 and PC2) accounted for 85.6% of the total variability of data matrix X between groups, while the first 8 components (PC1 to PC8) capture 98.6% of all the discriminatory information. Fig. 2 (A) shows the score plot by using the first 2 principal components. Five natural groups representing EVOOs from Spain/Italy, Greece/Italy, Turkey/Italy/Australia, Tunisia and Australia can be observed. As the results show, most of the oil samples could be well defined by separate clusters based on their regions; however, the samples from Italy and Australia partly overlapped with the other groups. The PCA analysis also indicated that the data differed most on the profiles of FAC and TAGs, which was worth exploring further in the following pattern recognition analysis. The plot of DModX indicated that no outlier was obtained in each group according to a confidence interval test (α=0.05 level) (Fig. 2 (B)). Principal components were extracted to represent patterns encoding the highest variance in the dataset and not to maximize the separation between groups directly, while hierarchical clustering analysis (HCA) could be used to verify the PCA results (Gerhardt, et al., 2019).

HCA could divide the samples into uniform groups, and hence the inter-group

similarities are smaller than the intra-group similarities. Heatmap merged with hierarchical cluster analysis was performed for the measured fatty acid composition and triglycerides in the samples from Spain, Greece, Turkey, Tunisia, Italy and Australia. Euclidean distance was used to calculate the distance between two clusters and an average linkage algorithm was applied to draw a dendrogram (Nami, Panahi, Mohammadzadeh Jalaly, Vaseghi Bakhshayesh, & Hejazi, 2020). Fig. 3 shows the hierarchically organized cluster heat-map based on the dendrogram of the samples and the variables. The row represents the variables and the columns represent the olive oil samples. As shown in this graph, the 85 samples could be partitioned into five groups on the horizontal axes. The first group only consists of Greek samples. The second group includes Spanish samples with a fraction of Italian samples. The third cluster comprises the Turkish samples. The fourth class is composed of the total samples of Tunisia with a small part of Italian and Australian samples. The fifth cluster contains both Italian and Australian samples. Table S1 shows the detailed information of the group composition. Overall, most of the samples were able to cluster according to geographical origins (except samples from Italy), indicating its potential for further classificatory analysis.

Results of PCA and HCA prove that the investigated olive oil samples have specific fatty acid and triglycerides profiles and that these compounds can be used to distinguish the different regions of olive oil. However, it should be noted that these two methods, PCA and HCA, are still not sufficient to satisfactorily group the olive oil samples according to their origins. In this context, it is very important to take the known

401 sample class into consideration for more accurate discrimination.

402 **3.2.2 LDA**

403 Compared with the unsupervised PCA and HCA, supervised LDA based on the
404 prior knowledge of sample class has a stronger discriminating capacity, so LDA method
405 was attempted to conduct geographical classification of the olive oil samples. Within
406 the origin group of olive oil, LDA identified five discriminant functions. Wilks's λ
407 demonstrated significant effects of origin on the entire model (Table S2). The first two
408 discriminant functions (Y_1 and Y_2) together explained 84.0% variance with a large
409 effect size (the canonical correlations > 0.9) (Stamenković, Steinwall, Nilsson, & Wulff,
410 2020).

411 Fig. 4 shows the discrimination of oil samples from different geographical origins.
412 For each group involved, filled marks represent the centroids, and the data points are
413 plotted in their individual coordinates and connected by a line to the respective group
414 centroid. As can be seen from Fig. 4, a clearer discrimination of the origin for the
415 samples was obtained. Tunisia was clearly separated from the others, while Spain,
416 Greece, Turkey, Italy, and Australia were distributed in areas that were close to each
417 other, with the groups of Spain and Italy partly overlapping. The dispersion of data
418 points within a given group is a key parameter for assessing the quality of the
419 differentiation achieved through LDA (de Toledo, de Melo, Pezza, Toci, Pezza, & Silva,
420 2017). In this respect, it is worth noting the Italian category had the highest dispersion.
421 The dispersion of Italy approximated that of Spanish and Greek classes. In this sense,
422 from the perspective of LDA, olive oil samples from this region seem to have

intermediate compositions of the discriminant compounds compared to the other two groups. According to the classification results shown in Table S3, EVOOs from Spain and Italy are more prone to be misclassified, while the samples from other countries were classified correctly and completely. The LDA model correctly classified 95.3% of EVOO samples. In particular, correct predictions were obtained for five regions (94.4%~100%), while a lower prediction percentage was obtained for Italian samples (76.9%). The results suggest that the origins of the most olive oils investigated were in agreement with the descriptions on their respective labels. However, 1 Spanish and 3 Italian oils, accounting for 4.7% of the total, were misclassified, which might be due to false declarations for the geographical origin.

3.3.3 OPLS-DA

For validating the results of LDA, a model between six classes was established based on supervised orthogonal partial least-squares pattern recognition methods combined with discriminant analysis (OPLS-DA). To check model reliability, two performance parameters were calculated: the cross-validation (n=7) and response permutation test (n = 200). Multivariate data were fitted into five components, showing $R^2X = 0.99$, $R^2Y = 0.784$ and $Q^2 = 0.704$. The response permutation test showed that $R^2 = 0.116$ and $Q^2 = -0.323$ (see Fig. 5 (B)). The results indicated that the model gave a satisfactory fit with a good predictive power. The score plot of samples (Fig. 5 (A)) projected on the first latent variables, i.e., R^2X [1] and R^2X [2], generally divided them into 6 clustering regions, and the first two latent variables explained 81.8% variance of data matrix. According to the score plot obtained by OPLS-DA, 3 Italian samples still

445 overlapped with Spain samples, but the other classes were completely separated from
446 each other. This result was basically consistent with that of LDA. Therefore, we could
447 conclude that, concerning the origin of the oils, the mislabeling rate of these olive oils
448 samples was 3.5%. Chemometrics with fatty acids and triglycerides were useful for
449 EVOO origin classification.

450 Moreover, the quality parameters of the 3 Italian samples were within the limits
451 defined for EVOO category designated by different regulations except PPP values of 2
452 samples. Therefore, it is not possible to reveal whether the product is origin fraud just
453 by the limits for EVOO category according the current regulations. Thus, as an
454 important aspect of assessing the quality of EVOO, traceability based on chemometrics
455 also should be highlighted.

456 ## 4 Conclusions

457 The quality and the geographical origin of EVOO are two of the most relevant
458 factors to determine their commercial value. As shown in Fig.6 , our study evaluated
459 the potential of traditional quality parameters and multi-chemical fingerprints using
460 fatty acid composition and triacylglycerols data to differentiate the quality and
461 geographical origin authenticity of the imported EVOOs (Spain, Greece, Italy, Turkey,
462 Tunisia and Australia) marketed in China . Based on the limit value of each parameter
463 for commercial category of EVOO, 61 samples failed to conform to the current official
464 standards at home and abroad, representing 71.8% of the total samples. In these samples,
465 the main quality problems were ageing, oxidation and counterfeit with refined oils.
466 Chemometric classification models coupled with fatty acid composition and

triacylglycerols data were successfully used to identify the geographical origin of olive oil, with a mislabeling rate of 3.5%. In light of the high proportion of failed samples, closer attention should be paid to adequate production and storage and shipping technologies of olive oil as well as an effective quality control of the olive oil market. Overall, the research provides a comprehensive evaluation on the intrinsic quality of the commercial imported EVOOs in the Chinese market. However, the limitation of the dataset in this study is that it is constituted by no more than 20 EVOOs from each country. In addition, the classification model was only internally validated using a training-set of 85 samples. Therefore, analysis on more samples and more in-depth research need to be carried out in the future.

CrediT authorship contribution statement

Xue Li: Methodology, formal analysis, investigation, writing-Original Draft, Funding acquisition. **Yu Zhang:** Data curation. **Zhi Liu:** Supervision. **Wei Wang:** Conceptualization, Supervision, Funding acquisition. **Sulin Sun:** Validation. **Junhong Wang:** Resources. **Zuoyi Zhu:** Validation. **Jun Liu:** Resources. **Hua Yan:** Project administration. **Shenlong Zhu:** Funding acquisition. **Erli Niu**: Funding acquisition. **Romero Agusti:** Supervision, Writing-Reviewing and Editing.

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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Figure captions

Fig. 1. (A)The total, passed and failed numbers of oil samples; (B) The contribution percentage of each parameter to the unconformity of all investigated samples, PPP: Pyropheophytin.

Fig. 2. The PCA score plot showing clustering of olive oil from Spain, Greece, Turkey, Tunisia, Italy, and Australia (A); and DModX plot (B).

Fig. 3. Heat map analysis of different compositions in six groups (Spain (blue), Greece (pink), Italy (gray), Turkey (yellow), Tunisia (green), and Australia (purple)). Colors are based on relative levels and changes in compounds, where red represents high level and dark blue represents low level.

Fig. 4. LDA score plot of oil samples from different countries.

Fig. 5. The OPLS-DA scores plots showing clustering of olive oil from different countries (A); and validation plot of the model obtained from 200 permutation tests (B).

Fig. 6. The analysis process and main results of the study.

Tables

Table 1. Parameters analysis for EVOO (n=85).

Parameters	Range detected	Means±SD	Limits	Samples exceeding limits	Samples in 2016, 2017, and 2018 exceeding limits
Free acidity (% oleic acid)	0.14-1.26	0.45±0.24	≤0.8	26[a]/5[b]	0/5/0
Peroxide value (meq O_2/kg oil)	4.23-16.31	9.39±2.72	≤10 or 20	30[c]/0[d]	5/24/1
Specific extraction					
K_{232}	1.10-2.71	2.03±0.29	≤2.5	5	0/4/1
K_{270}	0.06-0.80	0.19±0.15	≤0.22	7	2/4/1
Delta K	0.00-0.09	0.01±0.02	≤0.01	5	1/4/0
Delta ECN42	0.0003-0.2221	0.06±0.05	≤0.2	2	1/1/0

Fatty acids composition (%)				
C14:0	0.00	0.00±0.00	⩽0.05	0	0/0/0
C16:0	9.70-18.08	12.43± 2.28	7.5-20.0	0	0/0/0
C16:1	0.66-2.32	1.09±0.44	0.3-3.5	0	0/0/0
C18:0	1.94-3.59	2.75±0.41	0.5-5.0	0	0/0/0
C18:1	57.00-80.95	73.41± 6.60	55.0-83.0	0	0/0/0
C18:2	3.40-18.58	8.68±4.26	3.5-21.0	2	0/2/0
C18:3	0.55-1.00	0.68±0.09	⩽1.0	0	0/0/0
C20:0	0.35-0.49	0.44±0.04	⩽0.6	0	0/0/0
C20:1	0.23-0.40	0.30±0.05	⩽0.4	0	0/0/0
C22:0	0.10-0.18	0.14±0.02	⩽0.2	0	0/0/0
C24:0	0.04-0.08	0.06±0.01	⩽0.2	0	0/0/0
Pyropheophytin A (%)	2.14-43.80	16.44± 9.47	⩽17	41	11/30/0
Total				56[a]/40[b]	14[e] (12)[f]/ 60 [e] (48) [f]/ 11 [e] (3) [f]

a, the number of samples inconsistent with the label; b, the number of samples in exceed the limit of the Chinese standard; c, the number of samples in exceed the limit of Chinese standard (⩽10 meq O_2/kg); d, the number of samples in exceed the limit of IOC standard (⩽20 meq O_2/kg); e, the number of samples producting in 2016, 2017 and 2018, respectively; f, the number of samples failed to comply with the domestic and foreign standards. .

Table 2. Sensory results of the analyzed olive oil samples: Quality category and sensory attribute intensities according to IOC

Sample	Brand	Country of production	PDO/Organic	Crop year	Production date	Oil category assigned by panel	Median for positive attribute			Median of negative attribute (adding 3 blank tasters)	Principal defect
							Fruity	Bitter	Pungent		
S1	Ole SPANA	Spain	/	2017	October, 2017	EVOO	2.0	3.5	3.0	0	

S2	Ole SPANA	Spain	-/-	2017	October, 2017	EVOO	3.0	4.0	3.0	0	
S3	Ole SPANA	Spain	-/-	2017	October, 2017	EVOO	2.0	3.0	2.0	0	
S4	La Espanola	Spain	/	2017	March, 2017	EVOO	2.0	3.0	3.0	0	
S5	La Espanola	Spain	/	2017	March, 2017	EVOO	3.0	4.0	3.0	0	
S6	La Espanola	Spain	-/-	2017	March, 2017	EVOO	2.0	3.0	2.0	0	
S7	San. Francisco	Spain	-/-	2017	November, 2017	EVOO	2.5	3.0	3.5	0	
S8	San. Francisco	Spain	-/-	2017	November, 2017	EVOO	2.0	3.5	2.0	0	
S9	San. Francisco	Spain	-/-	2017	November, 2017	EVOO	2.5	2.0	2.0	0	
S10	BAENA	Spain	PDO/-	2017	September, 2017	EVOO	2.5	3.5	3.0	0	
S11	BAENA	Spain	PDO/-	2017	September, 2017	EVOO	3.0	3.5	3.0	0	
S12	BAENA	Spain	PDO/-	2017	September, 2017	EVOO	2.5	3.5	4.0	0	
S13	LOVINA	Spain	-/-	2017	July 3, 2017	VOO	2.0	2.0	2.0	3.0 (2.0)	Rancid
S14	LOVINA	Spain	-/-	2017	July 10, 2017	VOO	3.0	2.0	3.0	1.0 (0.0)	Rancid
S15	Ole SPANA	Spain	-/-	2018	February, 2018	EVOO	2.0	5.0	4.0	0	
S16	Ole SPANA	Spain	-/-	2018	February, 2018	EVOO	2.0	5.0	4.0	0	
S17	Ole SPANA	Spain	-/-	2018	February, 2018	EVOO	3.0	3.0	3.0	0	
S18	Ole SPANA	Spain	-/-	2018	February, 2018	EVOO	2.0	5.0	4.0	0	
G1	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	March, 2017	EVOO	2.0	0	3.0	0	

G2	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	March, 2017	EVOO	2.0	0	3.0	0	
G3	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	March, 2017	EVOO	2.0	0	3.0	0	
G4	KALLISTO	Greece	PDO, Koroneiki variety/-	2016	July, 2016	VOO	0	1.0	3.0	2.0 (0.5)	Fusty
G5	KALLISTO	Greece	PDO, Koroneiki variety/-	2016	July, 2016	VOO	1.0	2.0	2.0	2.0 (0.0)	Fusty
G6	KALLISTO	Greece	PDO, Koroneiki variety/-	2016	July, 2016	VOO	2.0	0	2.0	3.0 (1.5)	Musty/Rancid
G7	ILIAAA	Kalamata/Greece	PDO/-	2017	July, 2017	EVOO	2.0	0	3.5	0	
G8	ILIAAA	Kalamata/Greece	PDO/-	2017	July, 2017	EVOO	1.0	2.0	3.0	0	
G9	ILIAAA	Kalamata/Greece	PDO/-	2017	July, 2017	EVOO	1.0	3.0	2.0	0	
G10	IMINOS	Crete/Greece	-/-	2018	March, 2018	EVOO	3.0	2.0	2.0	0	
G11	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	March, 2017	VOO	2.0	1.0	1.0	2.0 (1.0)	Musty/Rancid
G12	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	October, 2017	VOO	1.0	1.0	1.0	2.0 (1.3)	Musty/Rancid
G13	KALLISTO	Greece	PDO, Koroneiki variety/-	2017	March, 2017	VOO	1.5	1.0	2.0	1.0 (0.5)	Musty/Rancid
G14	ILIAAA	Kalamata/Greece	PDO/-	2018	March, 2018	EVOO	2.0	1.5	1.8	0	
G15	ILIAAA	Kalamata/Greece	PDO/-	2018	March, 2018	EVOO	2.0	1.0	1.5	0	
G16	ILIAAA	Kalamata/Greece	PDO/-	2018	March, 2018	EVOO	2.0	2.0	2.0	0	
TR1	BEYZADE	Turkey	-/-	2017	May, 2017	VOO	2.0	1.0	1.0	1.0 (0.5)	Fusty

TR2	BEYZADE	Turkey	-/-	2017	May, 2017	VOO	2.5	1.0	2.0	1.0 (0.5)	Fusty
TR3	BEYZADE	Turkey	-/-	2017	May, 2017	EVOO	2.5	1.0	2.0	0	
TR4	Rioiera	Turkey	-/-	2015	August, 2016	VOO	1.0	1.0	1.0	3.5 (2.0)	Rancid
TR5	Rioiera	Turkey	-/-	2016	March, 2017	VOO	1.0	1.0	0	2.0 (0.5)	Rancid
TR6	Rioiera	Turkey	-/-	2016	March, 2017	VOO	2.0	1.0	1.0	2.0 (1.5)	Musty/Rancid
TR7	Rioiera	Turkey	-/-	2016	March, 2017	VOO	1.0	0	0	3.0 (1.8)	Musty/Rancid
TR8	Rioiera	Turkey	-/-	2016	March, 2017	VOO	1.0	0	0	2.0 (1.0)	Rancid
TR9	Poyraz	Turkey	-/-	2017	May, 2017	VOO	2.0	2.0	1.0	2.0 (0.5)	Fusty/Vinegary
TR10	Poyraz	Turkey	-/-	2017	May, 2017	VOO	2.0	1.0	2.0	2.0 (1.0)	Fusty
TR11	Poyraz	Turkey	-/-	2017	May, 2017	VOO	2.0	2.0	2.0	2.0 (0.5)	Fusty
TR12	TOMBIK	Turkey	-/-	2017	January, 2017	VOO	2.0	1.0	1.0	3.5 (2.0)	Rancid
TR13	TOMBIK	Turkey	-/-	2017	January, 2017	VOO	2.0	1.0	2.5	2.0 (1.3)	Rancid
TR14	TOMBIK	Turkey	-/-	2017	January, 2017	VOO	2.0	1.0	2.0	1.0 (0.5)	Fusty
TN1	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	1.0	2.5	3.0	0	
TN2	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	2.0	2.0	3.0	0	
TN3	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	3.0	3.0	3.0	0	
TN4	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	2.0	3.0	3.0	0	
TN5	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	1.0	3.0	2.0	0	
TN6	BORGES	Tunisia	-/Organic	2017	December, 2017	EVOO	3.0	3.0	2.0	0	
TN7	Aljazira	Tunisia	-/-	2016	June, 2016	EVOO	2.0	2.0	2.0	0	
TN8	Aljazira	Tunisia	-/-	2016	June, 2016	EVOO	2.0	2.0	3.0	0	
TN9	Aljazira	Tunisia	-/-	2016	June, 2016	VOO	1.0	2.0	2.0	3.0 (1.5)	Rancid

TN10	Aljazira	Tunisia	-/-	2016	June, 2016	VOO	2.0	2.0	3.0	2.0 (0.5)	Rancid
TN11	Huilerie Loued	Sahel/Tunisia	-/-	2016	August, 2017	EVOO	2.0	2.0	2.0	0	
TN12	Huilerie Loued	Sahel/Tunisia	-/-	2016	August, 2017	VOO	2.0	2.0	1.0	2.0 (0.5)	Fusty/Rancid
TN13	Huilerie Loued	Sahel/Tunisia	-/-	2016	August, 2017	EVOO	1.0	1.0	1.0	0	
I1	FILIPPO BERIO	Italy	-/-	2016	June, 2017	EVOO	2.0	3.0	3.0	0	
I2	FILIPPO BERIO	Italy	-/-	2016	June, 2017	VOO	2.0	2.0	2.0	2.0 (0.5)	Rancid
I3	FILIPPO BERIO	Italy	-/-	2016	June, 2017	VOO	2.5	2.0	4.0	2.0 (0.5)	Fusty
I4	Bellucci	100% Italy	-/Organic	2016/2017	September, 2017	EVOO	2.0	1.0	2.5	0	
I5	Bellucci	100% Italy	-/Organic	2016/2017	September, 2017	EVOO	3.0	2.0	2.8	0	
I6	Bellucci	100% Italy	-/Organic	2016/2017	September, 2017	EVOO	2.0	2.0	3.0	0	
I7	Pietro Coricelli	Italy	-/-	2016	April, 2017	EVOO	2.0	2.0	2.5	0	
I8	Pietro Coricelli	Italy	-/-	2016	April, 2017	EVOO	2.0	2.8	2.5	0	
I9	Pietro Coricelli	Italy	-/-	2016	April, 2017	EVOO	2.0	1.0	2.0	0	
I10	OLITALIA	Italy	-/-	2017	March, 2017	EVOO	2.0	3.0	3.5	0	
I11	COLAVITA	100% certified Italian	-/-	2017	March, 2018	VOO	3.0	3.0	2.0	1.0 (0.0)	Fusty
I12	COLAVITA	100% certified Italian	-/-	2017	March, 2018	EVOO	1.0	2.0	2.0	0	
I13	COLAVITA	100% certified Italian	-/-	2017	March, 2018	VOO	1.0	3.0	3.0	2.0 (0.5)	Fusty/Rancid
A1	Red Island	100% Australia	-/-	2017	June, 2017	EVOO	2.0	1.0	2.0	0	
A2	Red Island	100% Australia	-/-	2017	June, 2017	EVOO	1.0	1.5	1.5	0	

A3	DIANA	South Australia	-/-	2016	July, 2016	EVOO	2.0	1.0	1.0	0	
A4	DIANA	South Australia	-/-	2016	July, 2016	EVOO	2.0	1.8	2.6	0	
A5	DIANA	South Australia	-/-	2016	July, 2016	EVOO	2.0	1.0	2.0	0	
A6	AUPERTH	Western Australia	-/-	2016	October, 2016	VOO	1.5	1.0	1.0	2.5 (2.0)	Rancid
A7	AUPERTH	Western Australia	-/-	2016	October, 2016	VOO	1.5	1.0	2.0	2.0 (1.0)	Fusty
A8	AUPERTH	Western Australia	-/-	2016	October, 2016	VOO	2.0	1.0	1.0	2.0 (1.0)	Musty
A9	G&G	Western Australia	-/-	2017	December, 2017	EVOO	2.0	2.0	2.5	0	
A10	G&G	Western Australia	-/-	2017	December, 2017	EVOO	3.0	2.0	3.5	0	
A11	G&G	Western Australia	-/-	2017	December, 2017	EVOO	3.0	1.0	3.5	0	

682

683

Figure in color

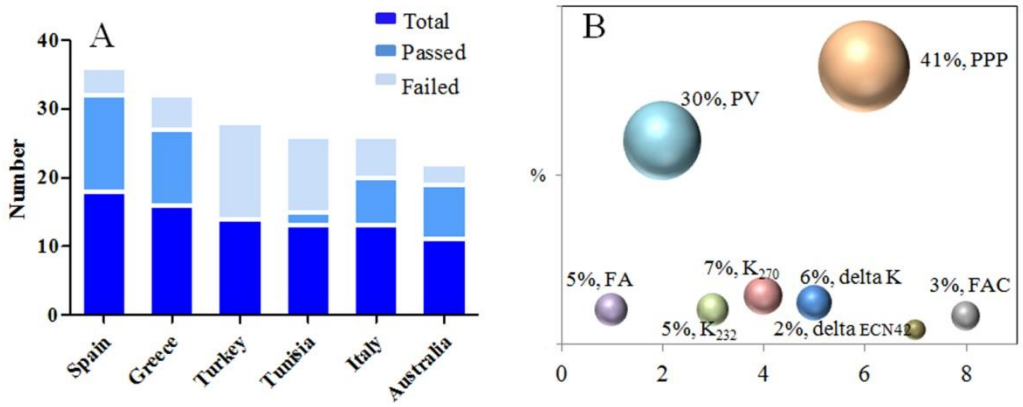

Fig. 1. (A)The total, passed and failed numbers of oil samples; (B) The contribution percentage of each parameter to the unconformity of all investigated samples.

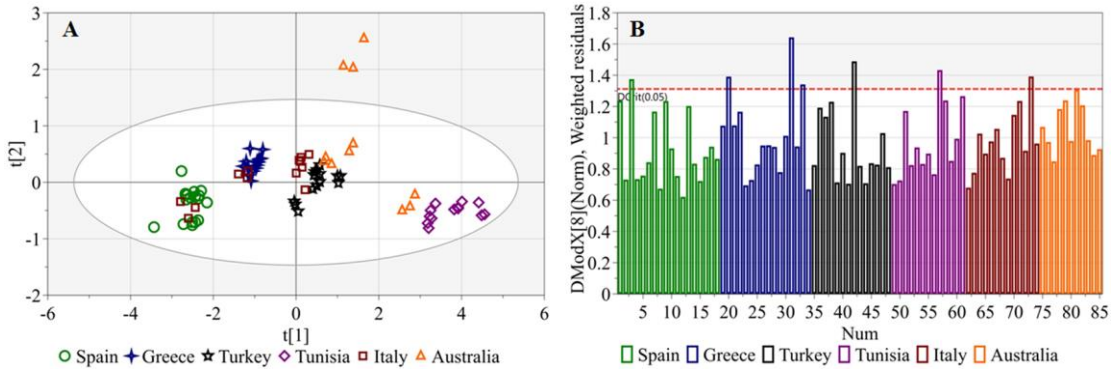

Fig. 2. The PCA score plot showing clustering of olive oil from Spain, Greece, Turkey, Tunisia, Italy, and Australia (A); and DModX plot (B).

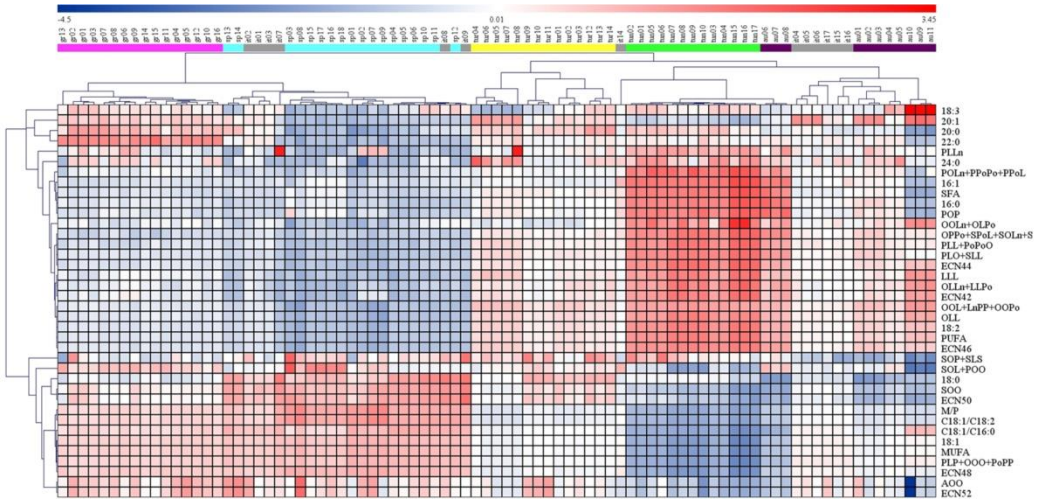

Fig. 3. Heat map analysis of different compositions in six groups (Spain (blue), Greece (pink), Italy (gray), Turkey (yellow), Tunisia (green), and Australia (purple)). Colors are based on relative levels and changes in compounds, where red represents high level and dark blue represents low level.

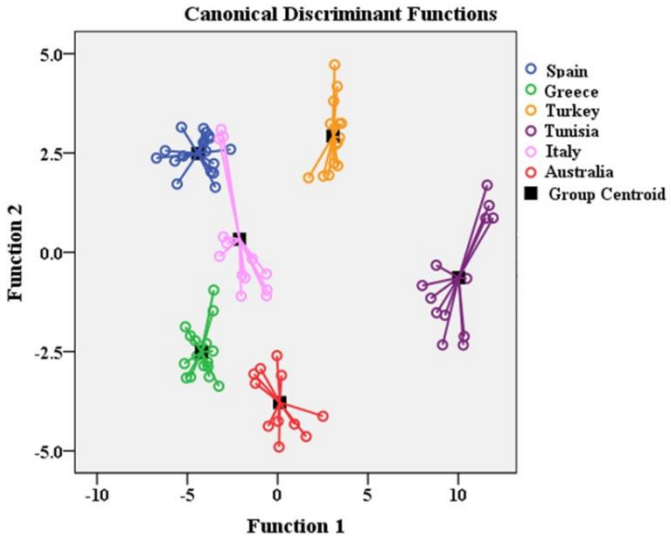

Fig. 4. LDA score plot of oil samples from different countries.

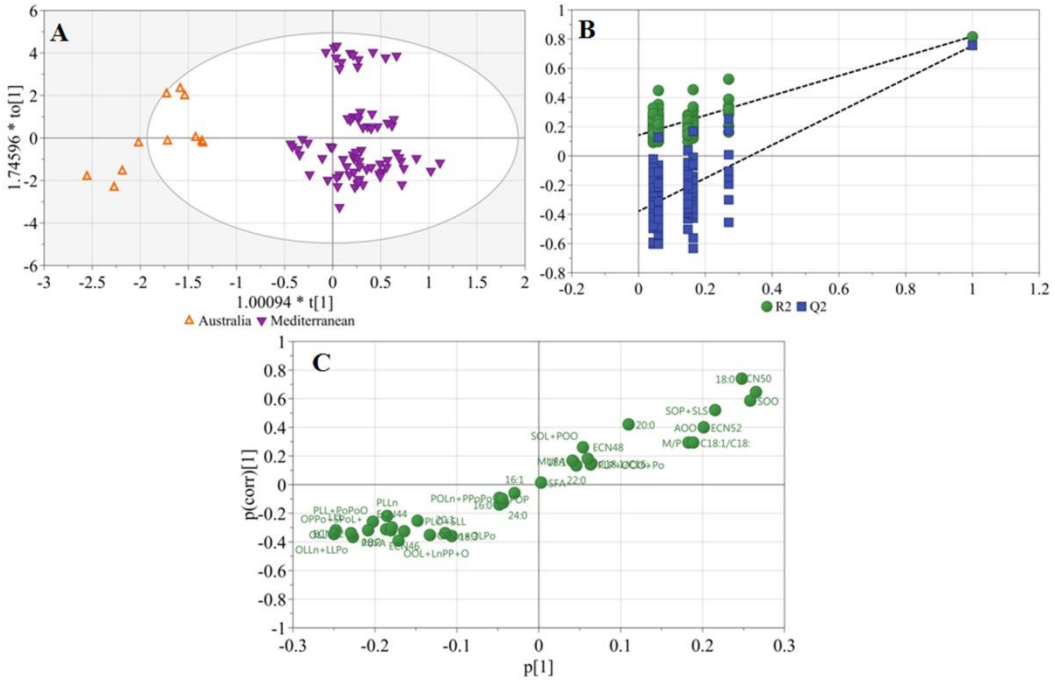

Fig. 5 OPLS-DA modeling of Australian and Mediterranean oilve oil: (A) score plot of samples; and (B) validation plot of the model obtained from 200 permutation tests; (C) S-plot

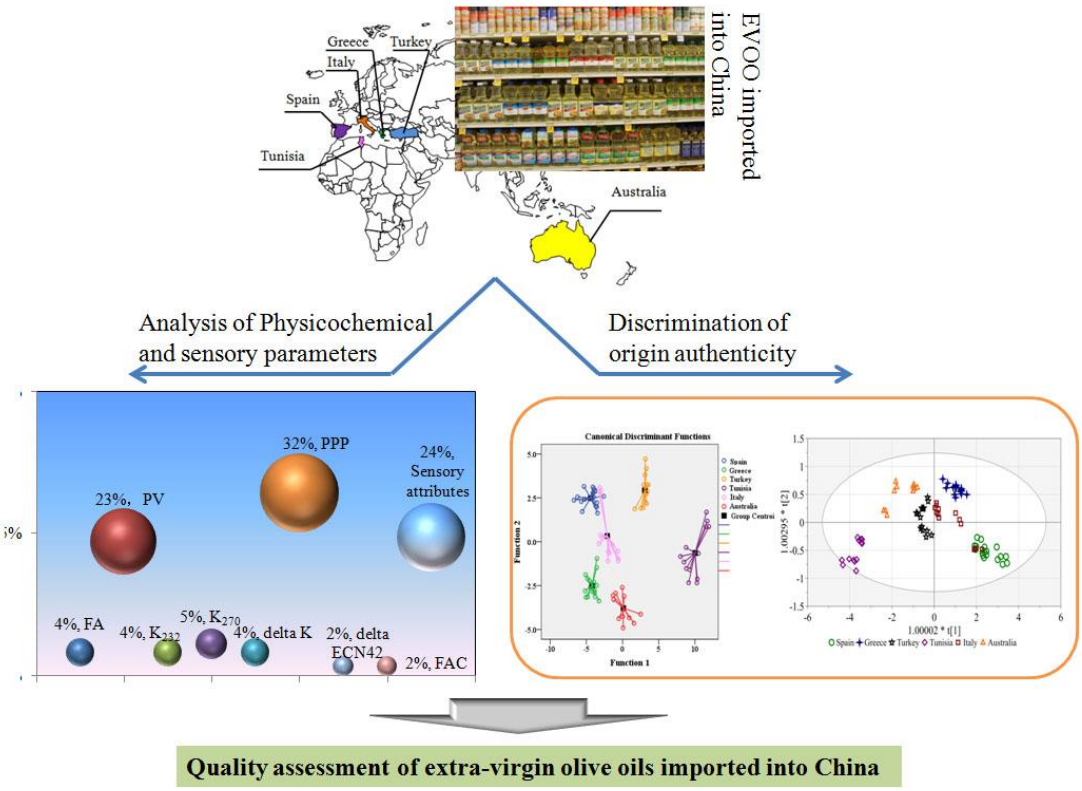

Fig. 6. The analysis process and main results of the study.