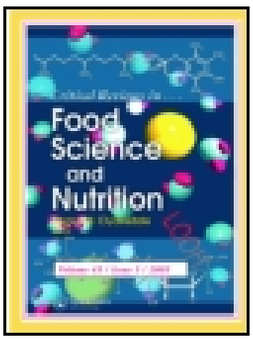

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REVIEW

Deriving valorization of phenolic compounds from olive oil by-products for food applications through microencapsulation approaches: a comprehensive review

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ABSTRACT

Nowadays, olive oil consumption is correlated to many health benefits, essentially due to the presence of antioxidants, especially phenolic compounds, which fostered its intensive production worldwide. During olive oil extraction, through continuous or discontinuous processes, many olive oil by-products are generated. These by-products constitute an environmental problem regarding its management and disposal. They are phytotoxic and biotoxic due to their high content of phenolic compounds, presenting contrastingly relevant health benefits due to their potent radical scavenging activities. In the framework of the disposal and management of olive oil by-products, treatment, and valorization approaches are found. As currently, the majority of the valorization techniques applied have a null market value, alternative strategies for the obtainment of innovative products as fortified foods are being investigated. The recovery and valorization strategies of olive oil by-products may comprise extraction and further encapsulation of bioactive compounds, as an innovative valorization blueprint of phenolic compounds present in these by-products. The majority of phenolic compounds present in olive oil by-products possess limited application on the food industry since they are promptly amended by environmental factors like temperature, pH, and light. Consequently, they must be protected previously ending in the final formulation. Prior to foods fortification with phenolic-rich extracts obtained from olive oil by-products, they should be protected through microencapsulation approaches, allowing a sustained release of phenolic compounds in the fortified foods, without losing their physicochemical properties. The combined strategies of extraction and microencapsulation will contribute to promoting the sustainability of the olive oil sector and aid the food industry to obtain reinvented added-value products.

KEYWORDS

Olive oil by-products; olive pomace; olive oil wastewaters; olive leaves; microencapsulation; phenolic compounds; extraction; valorization

Introduction

The Mediterranean Diet is a cornerstone on the prevention of cardiovascular diseases and some types of cancer, the lowering of the blood pressure and the positive effects on inflammatory processes as well as in the treatment of endothelial dysfunction (Pelucchi et al. 2011; Psaltopoulou et al. 2011; Buckland and Gonzalez 2015; D'Amore et al. 2016; López-Miranda et al. 2010; Violi et al. 2015). These positive health benefits associated with the Mediterranean dietary pattern are, in part related to the consumption of a specific source of fat – the olive oil (Psaltopoulou et al. 2011; Pelucchi et al. 2011; D'Amore et al. 2016; Violi et al. 2015; Buckland and Gonzalez 2015; López-Miranda et al. 2010).

The worldwide acknowledgment of the health benefits associated with the consumption of olive oil integrated into the context of the Mediterranean Diet has been stimulated an intensive production of olive oil, fostering the economic importance of the olive oil sector, especially among the producing countries (Stillitano et al. 2017; Patumi et al. 2002).

However, the olive oil sector generates a large amount of undesired olive oil by-products (OOBP) as olive leaves, olive stones, olive oil pomace (OOP), and olive oil wastewaters (OOWW). In the spectrum of OOBP, the olive leaves are the first olive oil by-product obtained during the pretreatment of olives. Considering the other typical OOBP, the olive pomace corresponds to the solid residue obtained from olive oil production. Contrastingly, olive oil wastewaters are the effluents obtained during the extraction of olive oil from olive drupes (Gogus and Maskan 2006; Rodríguez et al. 2008).

The majority of the compounds present in olives drupes are phenolic compounds which the octanol/water partition coefficient ranges between 6×10^{-4} and 1.5, and therefore they can present more affinity to aqueous systems rather than oily phases. A considerable amount of phenolic compounds–around 98%–are retained in olive oil by-products, and only 2% of these bioactive compounds are transferred to olive oil (El-Abbassi et al. 2014).

The high loading of phenolic compounds in OOBP has been rising attention of the scientific community due to

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their pollutant load. Moreover, olive oil by-products retain potent phytotoxic and biotoxic compounds effectively resistant to degradation, which constitutes a problem regarding by-products management and wastes disposal (Dermeche et al. 2013; Ghanbari et al. 2012; Rodrigues, Pimentel, and Oliveira 2015).

Considering the polluting load of phenolic compounds and simultaneously the outstanding protective effect of these compounds for human health mainly due to their potent radical scavenging activities, alternative and innovative valorization approaches of OOBP are currently being considered. One alternative that started to be explored is the extraction of these valuable compounds followed by its microencapsulation for further incorporation in complex matrices as foods considering the achievement of two leading goals: (i) the valorization of phenolic compounds present in olive oil by-products, and (ii) the address of the demand of added value products for food industry.

The scope of the present review is focused on (i) the compilation of data regarding OOPB, (ii) the presentation of the most commonly used extraction processes of olive oil from olives, (iii) the explanation of the environmental impact of olive oil by-products. Moreover, (iv) it is intended to introduce relevant data regarding valorization approaches of phenolic compounds present in olive oil by-products, (v) the display of relevant information regarding valorization of phenolic compounds present in olive oil by-products by extraction followed by the encapsulation of phenolic-rich extracts obtained, (vi) the critical exhibition of scientific studies regarding the phenolic content of OOBP and (vii) the review of studies regarding the valorization of OOBP extracts using encapsulation techniques exploring its main advantages.

Olive oil by-products composition

There are available different types of techniques that can be used to extract the olive oil from the olives, such as: (i) the traditional pressing mill process mostly used by small producers, (ii) the two-phase process and (iii) the three-phase process (Azbar et al. 2004; Dermeche et al. 2013; Obied et al. 2005; Qdais and Alshraideh 2016; Roig, Cayuela, and Sánchez-Monedero 2006; Sampedro et al. 2004). The olive oil extraction can be accomplished in traditional or modernized mills through discontinuous (pressing) or continuous (centrifuging) processes (Dermeche et al. 2013; Roig, Cayuela, and Sánchez-Monedero 2006).

Several steps employed to extract olive oil from the olives are common to the three extraction processes, as the harvesting of the olives, followed by its subsequently washing, crushing of the olives, and the malaxation of the obtained paste. As olive leaves are firstly separated during the harvesting process, they may be considered the first obtained olive oil by-product (Dermeche et al. 2013; Roig, Cayuela, and Sánchez-Monedero 2006).

The oldest and most outspread technique used to obtain olive oil from olives is the discontinuous pressing process (Roig, Cayuela, and Sánchez-Monedero 2006). The obtained malaxated olive paste is spread in the top fiber disks. These fiber disks containing the olive paste are placed at the top of the previous one. Then, pressure is applied in the fiber disks to compact the solid phase of the olive paste, allowing the percolation of the liquid phases (oil and water). In this step, a small amount of water is added to aid the separation of the oily phase from the other phases (aqueous and solid paste) (Azbar et al. 2004). Then after the press subprocess, a solid fraction is obtained. This new-obtained solid product is commonly known as olive oil pomace or olive cake and is made of olive skins, pulp, stones, and water. The liquid phases are then separated by decantation, resulting in the final of the extraction process, the olive oil, and olive oil wastewater. Some water may be added in the last step of the extraction process to promote the separation phase between olive oil wastewater and olive oil during decantation (Dermeche et al. 2013).

Until approximately the 1980s, the discontinuous pressing extraction technique was the most popular and employed technique to extract olive oil from olives. However, during the 1970s, this process was gradually replaced by innovative approaches (the two-phase process and the three-phase process) in order to reduce the labor force intense and dependence. The continuous processes (the two-phase process and the three-phase process) share a characteristic feature: all the phases (aqueous, oily, and solid) are separated by centrifugation using industrial decanters. Some decanters can be operated alternately either for the three-phase or the two-phase processes (Pedro et al. 2006).

The three-phase continuous centrifugation process requires the addition of warm water in the centrifugation step. This consequently leads to a higher production of OOWW (around 80–120 L per 100 kg of olives). The designation of the process as the three-phase is due to the fact that at the end of the continuous centrifugation process three different phases are obtained as a liquid oily phase (the olive oil), an aqueous phase (OOWW) and a solid residue, the olive oil residue (OOR) also known as olive oil pomace (Roig, Cayuela, and Sánchez-Monedero 2006). Even though its requirement of high-water consumption, the three-phase process is still employed in some countries as Portugal, Greece, and Italy to obtain olive oil from olives, especially in mills where is mandatory the production of olive oil in a short period of time (Dermeche et al. 2013). Nevertheless, the three-phase process has been replaced by the two-phase process in leading olive oil-producing countries like Spain–where around 90% of olive-mills operate using the two-phase process–as it allows to reduce the energy and water consumption (Agriculture and Food Organization of the United Nations 2019; Ruiz et al. 2017; Alburquerque et al. 2004).

In this context, the two-phase process was developed in the 1990s to minimize the water consumption that was used with the three-phase process. Using this technology, only two phases are produced in the end of the centrifugal process such as an oily phase (the olive oil) and a semi-solid phase–the wet pomace also known as the two-phase olive oil waste (TPOOW) also known as “alperujo” or even as “alpeorujo” (Roig, Cayuela, and Sánchez-Monedero 2006; Azbar et al. 2004; Benitez et al. 1997; Tortosa et al. 2012).

This semi-solid by-product obtained can be described as a combination of olive husk and olive oil wastewater. The TPOOW is usually further re-processed through second centrifugation to increase the olive oil extraction yield (Roig, Cayuela, and Sánchez-Monedero 2006; Alburquerque et al. 2004).

Alternative extraction techniques to obtain olive oil from olives are reported in the literature, such as the continuous combined percolation-centrifugation process, also known as Sinoela process. In this process, against the three and the two-phase processes in which are employed, a combination of pressure and centrifugation, in this process, is used a combination of filtration and centrifugation to obtain olive oil from the olive paste. The extraction technique is based on the difference of interfacial tension between the oil and the water that gets in contact with the steel plate. When the steel plate gets in contact with the olive paste, it is coated with oil due to the lower oil interfacial tension over water interfacial tension. However, the extraction efficiencies of this process are relatively low compared to the other ones, and additionally, it requires high operational and maintenance costs and high energy input. Generally, the olive paste obtained with the Sinolea process is re-processed using the traditional centrifugal apparatus in order to increase the extraction efficiency (Dermeche et al. 2013; Kapellakis, Tsagarakis, and Crowther 2008).

During the olive oil production, and regardless, the olive oil production process (continuous or discontinuous), two types of residues are produced: (i) a liquid one known as olive oil wastewater also known as black water in the case of the continuous three-phase process and the TPOOW in the case of the two-phase process and (ii) a solid residue, commonly known as olive oil pomace or even as olive oil residues or "prina" (Dermeche et al. 2013; Mirabella, Castellani, and Sala 2014; Naziri et al. 2014). Additionally, olives leaves can be considered the first residue that is obtained during olive oil production during the pretreatment phase, common to all olive oil extraction processes.

Examples of studies performed regarding the quantification of the total phenolic content and the antioxidant capacity of extracts obtained from alternative types of olive oil by-products (olive leaves, OOP, and OOWW) are presented in Table 1.

Olive leaves

During the pretreatment of olives, olive leaves are the first OOBP obtained. The chemical composition of olive leaves is widely dependent on several factors such as the geographical origin, the proportion of branches in the olive tree, the climatic conditions, storage conditions, moisture content and degree of contamination with soil and oils (Delgado-Pertíñez, Gómez-Cabrera, and Garrido 2000; Sabry 2014). The olive leaves, even though they account only 5% of olive weight, own many potential health benefits (Xynos et al. 2012).

For centuries, the leaves of olives and their extracts have been widely used in folk medicine for the treatment of diabetes, hyperglycemia, and infectious diseases (Komaki et al. 2003). Animal and in vitro studies pointed out that leaves and its extracts may have positive health effects on a wide range of diseases such as hypercholesteremia, hypertension, also possessing a vasodilator and antiallergic properties (Komaki et al. 2003; Gonzalez et al. 1992; Zarzuelo et al. 1991; de Bock et al. 2013).

The authors Benavente-García et al. (2000) (Table 1) determined the total antioxidant capacity of extracts obtained from olive leaves through the ability to scavenge the radical cation 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid (ABTS•+) (1.58 ± 0.06 mM of Trolox Equivalents). They also assessed the presence of specific phenolic compounds and their abundance. They found out that the major constituent of the olive leaf extracts obtained was oleuropein (24.54%), followed by hydroxytyrosol (1.46%), luteolin-7-glucoside (1.38%), apigenin-7-glucoside (1.37%), verbascoside (1.11%) and tyrosol (0.71%) Residual amounts of other phenolic compounds were also found (vanillic acid, dosmetin-7-glucoside, caffeic acid, luteolin, rutin, diosmetin, vanillin, and catechin; for all, the relative abundance was lower than 1%). Nevertheless, the authors' findings are significative: they identified that the relative amount of each phenolic compound was not directly related to its antioxidant capacity. Even though oleuropein was the compound mostly abundant in the obtained olive leaf extracts, it demonstrated a slight ability to scavenge radicals (total antioxidant capacity of 0.88 ± 0.09 mM of Trolox Equivalents). Alternatively, rutin which relative abundance was determined to be 0.05%, demonstrated the highest scavenging capacity (2.75 ± 0.05 mM of Trolox Equivalents).

Nevertheless, it is generally recognized that the main health properties of olive leaves and their extracts are associated to their high content of phenolic compounds, particularly, oleuropein and hydroxytyrosol as these two phenolic compounds demonstrate a strong radical scavenging capacity (Souilem et al. 2017; El and Karakaya 2009). Hydroxytyrosol is considered a novel safe food pursuant according to the last report of the European Food Safety Authority (EFSA) published in 2017.

Moreover, it is believed that the antioxidant capacity of extracts obtained from olive leaves is strongly affected by several factors as (i) the maturation status of olive leaves--mature olive leaves present more antioxidant capacity than young ones, (ii) the olive variety, (iii) the geographical localization of olives cultivation, (iv) the cultivation conditions, (v) the olive oil extraction processes–the content of water used for the extraction of the olive oil sets the antioxidants recovered and consequently the antioxidant activity of extracts as during the extraction many high molecular weight antioxidants are converted to low molecular weight antioxidants through hydrolysis–among others (Şahin and Bilgin 2018; De Marco et al. 2007).

Olive pomace

The olive pomace, also known as olive oil pomace (OOP), is semi-solid to a semi-liquid residue obtained through the olive oil extraction from olives. It presents low pH, high salinity, high organic loads, and high phenolic content. Due to its high phenolic content, olive pomaces are phytotoxic and

Table 1. Examples of studies performed regarding the quantification of the total antioxidant and phenolic content of different olive oil by-products.

Olive Oil By-Product	Study	Extract origin	Extracts Type/Origin	TAC	TPC	References
Olive Leaves	-Identification of phenolic compounds present OLE -Determination of the TAC of each compound and the extract	Alcantarilla, Spain	OLE	1.58 ± 0.06 [a]	N.F.	Benavente-García et al. (2000)
Olive Oil Pomace	-Study the effects of the 3-PhP and the 2-PhP on the phenolic composition of olive pomaces -Quantification of simple phenolic compounds regarding their antioxidant potentiality	Alpes-Côte-d'Azur, France	OOP (3-PhP) OOP (2-PhP) Extract (3-PhP) Extract (2-PhP)	N.F. 5.27 [b] 2.61 [b]	24.0 [c] 20.4 [c] 28.8 [g] 42.2 [g]	Lesage-Meessen et al. (2001)
	-Quantification of the TAC and the TPC of the OOP extracts obtained from different cultivars -Evaluation of the antimicrobial activity of the obtained extracts	Wagga Wagga, Australia	Mission OOP extract Frantoio OOP extract	60.6 ± 2.3 [c] 79.9 ± 2.0 [c]	17.7 ± 0.9 [d] 21.1 ± 0.9 [d]	Obied et al. (2007)
	-Evaluation of TAC of hydroalcoholic extracts in cell-free assays and in cell systems exposed to free radical attack	Northern Barese, Italy	Coratina OOP	26.96 ± 1.56 [d]	19.7 [c]	
Olive Oil Wastewater	-Evaluation of different extraction methods for the recovery of bioactive compounds for OOWWs -Evaluation of the in vitro TAC and anti-inflammatory activities of the obtained extracts	Italy, Spain, and France	Extract obtained by Lio-OOWW fractionation Extract obtained by LLE Extract obtained LLE fractioned in a Sephadex LH-20 column	9.42 [d] 3.12 [d] 1.83 [d]	N.F.	Visioli et al. (1999)
	-Determination of the TPC, flavonoids, flavanols, and proanthocyanidins of two different OOWWs -Evaluation of TAC and radical scavenging activity of phenolic extracts and microfiltered samples using different tests (iron(II) chelating activity, total antioxidant capacity, DPPH assays, and lipid peroxidation test).	Marrakech, Marroco	OOWW 1 Semi-modern 3-PhP OOWW 2 Modern 3-PhP	263 ± 2.5 [e] 169 ± 5.2 [e]	9.82 ± 0.53 [h] 6.11 ± 0.2 [h]	El-Abbassi, Kiai, and Hafidi (2012)
	-Study the influence of LLE in the obtainment of phenolic-rich extracts -Fractionation of OOWW phenolic extracts by reversed phase solid phase extraction	Benevento, Italy	Extract (acidified OOWW) Extract (crude OOWW)	N.F. 55.8 [f]	N.F. 25.2 [i]	De Marco et al. (2007)

2-PhP – Two-Phase Process; 3-PhP – Three-Phase Process; ABTS – 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid) assay; DPPH- 2,2-diphenyl-2-picrylhydrazyl radical scavenging test; Lio-OOWW – Lyophilizate of olive oil wastewater; LLE – Liquid-liquid extraction; N.F. – Not Found; OLE – Olive Leaf Extract; OOP – Olive Oil Pomace; OOW – Olive Oil Waste; OOWW – Olive Oil Wastewater; TAC – Total Antioxidant Capacity; TPC – Total Phenolic Content;
[a]Expressed in Trolox Equivalent Antioxidant Capacity (mM of Trolox Equivalents); Data is expressed as mean ± standard deviation;
[b]IC_{50} values; Antiradical activity was defined as the concentration of sample (EC_{50}) necessary to decrease the initial DPPH radical concentration by 50%; Data expressed in mg of gallic acid equivalents/L of reaction medium;
[c]% (w/w) dry basis; Determined as gallic acid equivalents by Folin–Ciocalteu method;
[d]mg/g of dry matter; EC_{50} (ppm);
[e]IC_{50} values; Determined for 0.1 g/L of DPPH initial concentration; Expressed in μg/mL;
[f]mmol of Trolox equivalents/L OOWW;
[g]% (w/w) of the total phenols present in the dry residue;
[h]g of tyrosol equivalent/L;
[i]mmol tyrosol/L OOW.

biotoxic, nevertheless they present antioxidant and antimicrobial properties (Bertin et al. 2011; Bleve et al. 2011; Sampedro et al. 2009; Khoufi, Feki, and Sayadi 2007; José Antonio Alburquerque et al. 2006; Linares et al. 2003; Piotrowska et al. 2011; Morillo et al. 2009).

Depending on the olive oil extraction process used, approximately 35–40 kg of OOP is produced for 100 kg of olives processed. It consists of a complex matrix of pits (42–54% w/w), pulp fragments (21–33% w/w), and olive skins (10–11% w/w) (Akay et al. 2015; Chanioti and Tzia 2019).

Subsequently, the OOP can be refined, submitting to a centrifugation process to extract residual olive oil and the resulting by-product can be dried and further extracted using solvents (e.g., hexane) to obtain edible oil. The amount of edible oil obtained after refining OOP is dependent on the moisture content, however, it is pointed out that the amount of OOP obtained can reach 25–30%, 45% and 70% w/w on a dry basis, using the pressing, the three-phase, and the two-phase process, respectively (Meziane and Kadi 2008; Akar et al. 2009; Alburquerque et al. 2004; Roig, Cayuela, and Sánchez-Monedero 2006).

Even though the physicochemical composition of the olive pomace is dependent on the olive variety, geographical localization of olives cultivation, olive oil extraction processes, and cultivation conditions, the final chemical composition is quite similar among different samples of OOP. However, olive oil pomaces are rich in pectic

polysaccharides, cellulose, and hemicellulosic polymers. Regarding the mineralogical content of OOP, it is mostly constituted by potassium (P), calcium (Ca), and sodium (Na). Additionally, it contains high quantities of water (70–80%), low olive oil content (2–3%), and many phytochemicals as antioxidants (especially phenolic compounds) and peptides (Dermeche et al. 2013; Rodríguez-Gutiérrez et al. 2014).

The authors Lesage-Meessen et al. (2001) studied the effects of using the three-phase centrifugation process and the two-phase centrifugation process on the phenolic profile of olive pomaces and the antioxidant capacity of extracts obtained from the pomaces (Table 1). Regarding the total phenolic content of OOPs, they found out that the amount of phenolic compounds was higher in the OOP obtained through the three-phase system (24.0% w/w on a dry basis) rather than the olive pomace obtained through the two-phase system (20.4% w/w on a dry basis). During the three-phase process are produced pomaces with higher content in phenolic compounds when compared to the two-phase olive oil production processes.

Interestingly, when the authors submitted the olive oil pomaces to the same extraction procedure, they verified that the total amount of phenols recovered was significantly higher in the extract recovered from the pomace obtained from the two-phase process (42.2% w/w in the dry residue) rather than in the three-phase process (28.8% w/w in the dry residue). It can be concluded that the amount of phenolic compounds recovered from OOP is significantly dependent on the extraction technique–even though the pomace obtained during the three-phase system presented a higher amount of phenolic compounds, the respective extract presented a lower content on phenolic compounds. Additionally, the authors Lesage-Meessen et al. (2001) measured the antiradical activity of the extracts by the concentration of the sample (EC_{50}) required for the decrease of the initial amount of the radical 2,2-diphenyl-2-picrylhydrazyl (DPPH) concentration by 50%, being the obtained value inversely proportional to the extract antioxidant activity. According to the results obtained for the total phenolic content, the highest antiradical activity was obtained with the extract processed after the recovery of olive oil through the two-phase centrifugation process (IC_{50}=2.61 mg of gallic acid equivalents/L of reaction medium) rather than the three-phase process (IC_{50}=5.27 mg of gallic acid equivalents/L of reaction medium).

The authors, Obied et al. (2007), quantified the total antioxidant capacity and the total phenolic content of the OOP extracts obtained from different cultivars. Even though the authors do not specify the differences among the two cultivars, the highest antiradical activity of extracts was associated to the lowest phenolic compounds content (OOP extract obtained using the Mission cultivar: IC_{50} of 60.6 ± 2.3 ppm and total phenolic content of 17.7 ± 0.9 mg of gallic acid equivalents/g of dry weight; OOP extract obtained using the Frantoio cultivar: IC_{50} of 79.9 ± 2.0 ppm and total phenolic content of 21.1 mg of gallic acid equivalents/g of dry weight). These findings are in agreement with the ones obtained by Benavente-García et al. (2000) when they were determining the total antioxidant capacity of extracts obtained from olive leaves through the ability to scavenge the radical cation ABTS•+. Nevertheless, this tendency should be examined thoroughly as other researches obtained opposite results–the highest value for the total antioxidant capacity can be correlated to the highest amount of phenolic compounds present in the evaluated extract. The results obtained by Obied et al. (2007) are significant as they prove the total antioxidant capacity and the total phenolic content dependence of the olive cultivar.

Olive oil wastewater

Generically, the olive oil wastewater is an organic residue with a high content of phenolic compounds, and other valuable nutrients. This high conductivity liquid is also rich in nitrogen (N), phosphorus (P), potassium (K), and magnesium (Mg). Alongside its water high content (around 83–92%), minerals, and phenolic compounds, it also may contain sugars and organic acids (Dermeche et al. 2013; Mekki, Dhouib, and Sayadi 2007; Piotrowska et al. 2011).

The octanol/water partition coefficients of the majority of olive phenolic compounds range between 6.0×10^{-4} and 1.5, and therefore, they may present more affinity to water systems rather the oily systems. This consideration agrees with the reported data regarding the amount of phenolic compounds in olive oil (around 2%), in OOWW (approximately 53%) and OOP (around 45%). Therefore, the majority of phenolic compounds are present in the olive oil wastewaters (El-Abbassi et al. 2014).

The OOWW physicochemical composition varies according to the olive variety, cultivation features, olives storage time, and extraction process applied (Dermeche et al. 2013).

The high organic load of OOWW is associated with its high content on phenolic compounds and low pH. These physicochemical features of olive oil wastewaters contribute significantly to environmental pollution, especially in the olive oil production areas.

Even though olive oil wastewater represents a threat to the environment, it also owns relevant biological properties, and it is a potent radical-scavenging and metal-chelating matrix (Khoufi, Aloui, and Sayadi 2008).

The recovery of phenolic compounds-rich extracts from OOWW by liquid-liquid extraction using ethyl acetate as the extraction solvent was described by Khoufi, Aloui, and Sayadi (2008). The extraction was applied as a pretreatment of electrocoagulation of the exhausted fraction of wastewater to reduce its toxicity on anaerobic digestion. The analytical results of the OOWW extract by gas-chromatography mass-spectrometry (GC-MS) analysis pointed out that the major compound present in the extract was hydroxytyrosol (66.5%). Minor concentrations of tyrosol, caffeic acid, p-coumaric acid, and ferulic acid were also detected. The authors concluded that olive oil wastewater is a valuable source of natural antioxidants. Additionally, other authors concluded that the antimicrobial and phytotoxicity potential of extracts obtained from olive oil wastewater is mainly attributed to its

high content of minor phenolic compounds and phenolic acids (Casa et al. 2003; González et al. 1990; MacCi, Masciandaro, and Ceccanti 2010; Ghanbari et al. 2012).

The positive effect of extracts recovered from OOWW as an antiatherogenic matrix was highlighted by Leger, Kadiri-Hassani, and Descomps (2000), reinforcing the olive oil wastewaters potential as a matrix source of phenolic compounds-rich extracts to be incorporated into foods.

The OOWW presents not only oxidative scavenging potential but also antimicrobial activity, which makes this olive oil by-product an excellent source of valuable natural antioxidants after their extraction from the wastewater complex matrix.

The valorization of OOWW through the extraction of phenolic compounds may have a positive impact regarding the disposal and the by-product management of this complex matrix.

Regarding the studies of the total antioxidant capacity and the phenolic content of extracts obtained from OOWW (Table 1), Visioli et al. (1999) evaluated different extraction methods for the recovery of bioactive compounds for OOWW. They concluded that the extract obtained from OOWW submitted to a freeze-drying process and afterward fractioned, presented the lowest antiradical activity (EC_{50}=9.42 ppm), followed by the extract obtained by liquid-liquid extraction (EC_{50}=3.12 ppm), being the liquid-liquid extraction followed by the fractionation of the extract in a Sephadex LH-20 column, the best extraction approach for the recovery of antioxidants (obtained extracts presented an EC_{50} value of 1.84 ppm). The research presented by Visioli et al. (1999) supports the thesis of the significant influence of the extraction approach on the total antioxidant capacity of extracts obtained from the same initial sample.

The study on the dependence on the modernization degree of olive oil production bases both operating in a three-phase process of the total antioxidant capacity and the total phenolic content of extracts obtained from OOWW was performed by El-Abbassi, Kiai, and Hafidi (2012). The degree of modernization of olive oil production bases seems to affect the antiradical activity and the total phenolic content of the extract–the most modernized olive oil production plant presented the lowest antiradical activity (IC_{50} of extracts obtained of OOWW from a semi-modernized base of $263 \pm 2.5\ \mu g/mL$ versus IC_{50} of extracts obtained of OOWW from a modernized plant of $169 \pm 5.2\ \mu g/mL$). Similarly to the results obtained Obied et al. (2007) in the case of extracts obtained from olive oil pomace and Benavente-García et al. (2000) in the case of extracts obtained from olive leaf extracts, the highest recovery of phenolic compounds was associated to the lowest antiradical activity–the semi-modernized olive oil production plant presented the highest total phenolic content (9.82 ± 0.13 g of tyrosol/L) but simultaneously the lowest antiradical activity.

Accordingly, from the comprehensive complication of studies presented in Table 1, can be concluded that the type of OOPB (olive leaves, olive oil pomace or olive oil wastewater), the extraction technique applied, the olive oil production process (three-phase process versus two-phase process), the type of cultivar and the degree of modernization of the olive oil production process plant from where the olive oil by-products are obtained influence considerably the total antioxidant capacity of the extracts as well as their phenolic content.

Phytotoxicity and biotoxicity of olive oil by-products–an environmental concern

The olive oil production industry generates a large amount of undesired liquid and semi-solid by-products.

Regarding the organic load, the amount of olive oil by-products produced by the classical (discontinuous) process and by the continuous process are 1.18 and 1.68 m^3/tonne of olives processed, respectively. The chemical oxygen demand (COD) and the biological oxygen demand (BOD) of the residues obtained through the discontinuous process can be found at maximum values of 79.2 and 100 g/L, respectively. In the case when the olive oil is produced by continuous processes, the COD and BOD levels may reach values of 121.7 and 200 g/L (maximum concentrations reported) (Eroğlu et al. 2006; Ergüder, Güven, and Demirer 2000; Eroğlu et al. 2004).

The disposal of untreated olive oil residues is considered an environmental issue mainly due to the highly toxic organic loads and also due to its low pH and the presence of sugars, nitrogenous compounds, volatile acids, polyalcohols, pectins, fats and most importantly, polyphenols (Dermeche et al. 2013; Ghanbari et al. 2012). Regarding the disposal and management of olive oil by-products, the high loading of phenolic compounds in these by-products has a relevant and negative effect on the environment.

The main effluents of olive oil production are OOWW. It is estimated that the worldwide OOWW generation accounts approximately between 10 to more than 30 millions m^3, corresponding approximately to 0.55–2 L of OOWW/kg of olives processed (El-Abbassi, Kiai, and Hafidi 2012; Ergüder, Güven, and Demirer 2000; Eroğlu et al. 2006, 2004).

Even though the amount of residues produced is much lower than compared to residues from other agro-food industries, its impact on the environment is considered to be significant. As regards environmental pollution, is reported that 1 m^3 of OOWW is equivalent to 200 m^3 of domestic sewage (Shilev and Naydenov 2007; El-Abbassi, Kiai, and Hafidi 2012).

During the production of olive oil through the two-phase process, the recycling of olive oil wastewater can be performed during the processing of the olives in order to reduce the wastewater effluent and environmental disposal problems (Lesage-Meessen et al. 2001; La Casa and Castro 2014). Considering the OOWW disposal, a pretreatment is essential as its disposal in water systems (surface water, groundwater systems, seashores, and sea) without previous pretreatment leads to a significant impact on the ecosystems. Many studies regarding the impact of olive oil wastewater as soil, and water bodies polluter as well as a cause of undesired underground seepage and odors, are available

(Paraskeva and Diamadopoulos 2006; Rinaldi, Rana, and Introna 2003).

The OOWW are also a toxic waste for some microorganisms and plants as their total phenolic content, organic load, total tannins content are considerably high (El-Abbassi, Kiai, and Hafidi 2012; Rinaldi, Rana, and Introna 2003). Moreover, the presence of long-chain fatty acids increases olive oil wastewater pollutant load (Boari et al. 1984; Paraskeva and Diamadopoulos 2006).

Although different authors report alternative chemical explanations for the OOWW environmental impact, it is generally recognized that the phytotoxicity and biotoxicity of these wastewaters are mainly due to the high phenolic content of this organic by-product.

Notwithstanding, contradictory studies are found in the literature. Some authors claim that both soils and plants may beneficiate from the application of OOWW and even olive oil pomaces as fertilizers, as others report that it may have a phytotoxicity effect as it may inhibit seeds germination and plants growth (Saviozzi et al. 2001; Paraskeva and Diamadopoulos 2006; Rinaldi, Rana, and Introna 2003).

The authors Mekki, Dhouib, and Sayadi (2007) studied the phytotoxicity of phenolic compounds in a soil amended with olive oil wastewater. They verified that the infiltration of olive oil wastewater in the soil caused a significative modification of its physicochemical characteristics. Phenolic compounds were detected at a depth of 1.2 m after four months of the final application of the olive oil wastewater in the soil. They verified the presence of a moderate phytotoxic phenolic residual fraction after one year of the olive oil wastewater application. The phenolic phytotoxicity level was comparable to a 25 times-fold diluted olive oil wastewater. The negative impact on the amended soil properties was also discussed by Mekki and Dhouib (2006) and Paredes et al. (1999).

The toxicological effect of OOWW and its phenolic content against seeds, shellfishes, and freshwater animals was exhaustively studied by Aggelis et al. (2003). They found that some specific organisms can minimize the phenolic toxicity of olive mill wastewaters in bioreactor cultures. After the biotreatment, the OMWW were used as water for the irrigation of lettuces and tomatoes. Even though the biotreated olive oil wastewaters did not affect the uptake the majority of the cultivated plants' nutrients, it resulted in a decrease in the plant yields. The decrease in the plant yields was minimized when diluted biotreated olive oil wastewater was used for irrigation purposes. The authors acknowledged that high olive oil wastewater dilutions after biotreatment are required and/or additional treatments should be considered before the use of OOWW for water irrigation. Moreover, the authors clarified that research in this field should be transferred to real/industrial conditions.

The possibility of using OOWW for the irrigation of plants and its phytotoxicity was evaluated by Sassi et al. (2006). The authors found out that the use of non-diluted olive oil wastewater blocked the germination of the *Ordeum vulgare* seeds. The germination of the seeds was only possible after dilution of 1:16 of olive oil wastewater. Additionally, this research paper describes that the high concentration of heavy metals and phenolic compounds may be the explanatory reason for the toxicity of olive oil wastewater.

Regarding the biotoxicity effects of OOWW, it is reported that the river fish *Gambusia affinis* and the crustacean *Daphnia magnaare* were intoxicated when exposed to phenolic compounds derivates at a concentration of 40 mg/L for only 15 min (Angus 1983). Immediate toxic effects were verified on the fish and the ray-finned fish *Cyprinus carpio* and *Chondrostoma polylepis* at concentrations of 6.8% v/v and 8.8% v/v and on the goldfish *Carassius auratusat* at a concentration level of 10% v/v (Mekki, Dhouib, and Sayadi 2007)

Further to the environmental impact of OOWW, the olive oil pomace is also in the highlight of recent studies of its phytotoxicity and biotoxicity (Salomone and Ioppolo 2012). Due to its high moisture content, several drawbacks are faced during its disposal and by-product management as it is required high energy input for the drying process, which is inevitably associated with disposal high costs (Salomone and Ioppolo 2012). Nevertheless, it was verified a lack of studies regarding the phytotoxicity and biotoxicity of olive oil pomaces.

Alongside olive oil wastewaters and pomaces, olive stones are becoming in the highlight of environmental concerns due to problems of its disposal. Even though olive stones do not present significant phytotoxicity and biotoxicity as no large amounts of this olive residue are generated, its disposal has been studied to diminish its environmental impact. Most of the olive stones are used as biomass for fuel and active carbon production. However, a significant impact in air pollution may arise from the direct combustion of this olive oil residue (Nieto et al. 2010).

Ultimately, olive oil wastewater, the major effluent of olive oil production, is under intensive research as it has been demonstrated to be phytotoxic and biotoxic, mainly due to presence in high quantities of phenolic compounds. Even though biotreatments can be applied to reduce the polluting load of OOWW, in the majority of the studies, it only minimized the phytotoxicity after several OOWW dilutions. Therefore, the disposal and application of OOWW are considered to be a relevant environmental concern. The olive oil pomace and olive stones are also in the highlight regarding the phytotoxicity and the biotoxicity in the context of the disposal and management of olive oil by-products. Specifically, in the case of OOP, its disposal and by-product management are still challenging ascribable to the high content of phenolic compounds (Dermeche et al. 2013).

Deriving valorization from olive oil by-products

In the framework of the disposal and management of olive oil by-products, two complementary approaches are found: (i) its treatment and (ii) its valorization.

Olive oil production generates a large amount of undesired by-products. Therefore, a balanced address of both technologies (treatment and valorization) has been intensively investigated as a direct result of the problematics

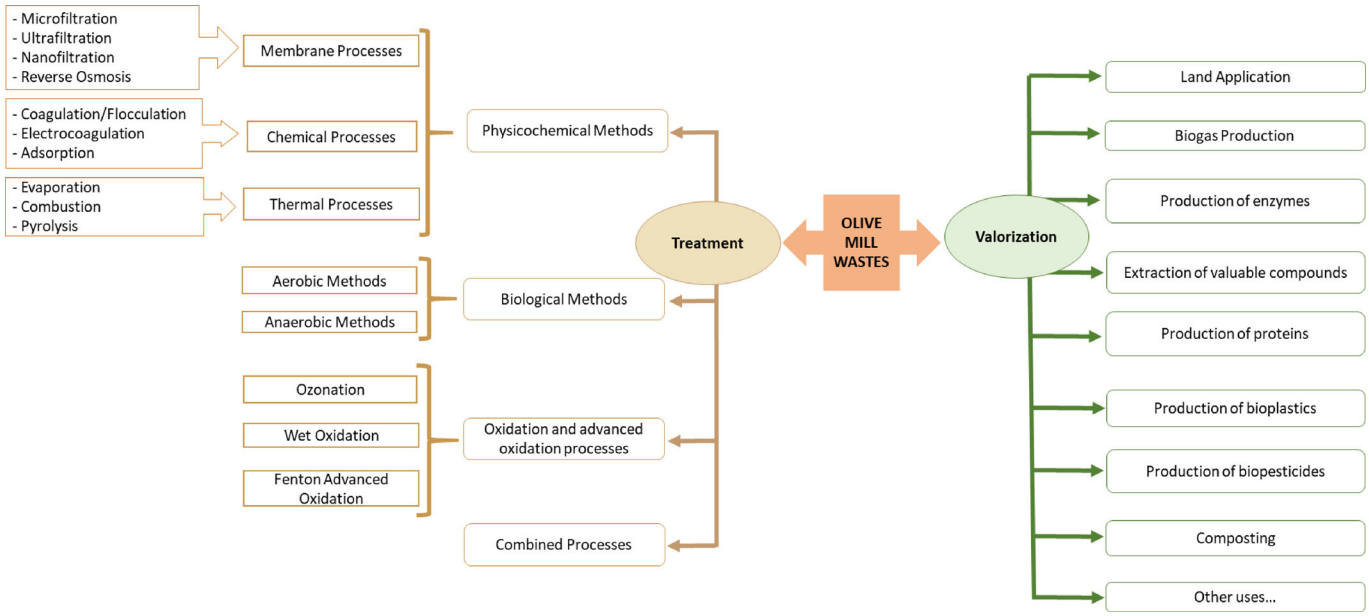

Figure 1. Olive oil by-products valorization and treatment approaches.

regarding the olive oil residues disposal and management and the phytotoxicity and biotoxicity of these residues.

The most commonly applied OOR treatments are based on physicochemical and biological methods. Moreover, olive oil residues can be treated using oxidation and advanced oxidation processes or even combined processes.

The physicochemical processes can be distinguished in chemical processes (e.g., coagulation/flocculation, electro flocculation, adsorption), thermal processes (e.g., evaporation, combustion, pyrolysis) and membrane processes (microfiltration, nanofiltration, ultrafiltration, reverse osmosis). The biological processes can be from anaerobic or aerobic nature. The oxidation and advanced oxidation processes include ozonation, Fenton advanced oxidation, and wet oxidation (Rharrabti and Yamani 2018). The authors Paraskeva and Diamadopoulos (2006) reported that the adsorption chemical process for the treatment of olive oil wastewater is effective in the removal of the total phenolic content by a maximum of 81% using activated clay as an adsorbent.

Alternatively, phenolic compounds present in olive oil residues can be valorized through the obtainment of phenolic compounds rich extracts instead of being treated through their passage from OOBP to adsorbents (e.g., activated clay). When phenolic compounds are treated by an adsorption process, a mass transference is observed, and phenols present in the activated clay should be further treated–and the problem persists.

The valorization of olive oil residues includes (i) land application, (ii) biogas production, (iii) composting, (iv) extraction of valuable compounds, and (v) alternative and innovative uses as enzymes, bioplastics and even proteins production (Rharrabti and Yamani 2018). A schematic representation of the most relevant processes and methods for olive oil by-products valorization is shown in Figure 1.

Regarding the potentially harmful impact of OOBP, combustion processes for fuel production have been applied for the withdrawal of the phytotoxicity and biotoxicity due to olive oil by-products high content in phenolic compounds and simultaneously as volume waste-reducing strategy (Rubio-Senent et al. 2015; Miranda et al. 2012).

Alternatively, olive oil by-products management can be performed through the controlled disposal in soils. However, only a minor fraction of OOBP is used as a natural fertilizer or as an additive in animal food feeding. None of these olive oil by-products management strategies is surprisingly considered to have a significant economic impact (Akar et al. 2009).

Therefore, considering the null market value of olive oil by-products controlled disposal on soils and its use as a natural fertilizer or even as an additive in animal food feeding, research in this scientific field has been focused on the finding of innovative strategies to olive oil by-products valorization, especially, its phenolic compounds as they the leading cause of olive oil by-products phytotoxicity and biotoxicity. These innovative valorization strategies of phenolic compounds from OOPB, generally encompass extraction techniques as the ultrasound-assisted, microwave-assisted, pressurized liquid, liquid-solid extractions, and extraction through the application of pulsed electric fields high voltage electrical discharges (Dermeche et al. 2013). These extraction techniques allow not only to recover phenolic compounds but even proteins from OOBP. Supercritical fluid extraction has been also studied as valorization technique of olive oil by-products, aiming the recovery of tocopherols and squalene (Putnik et al. 2017; Koubaa et al. 2017; Galanakis, Tsatalas, and Galanakis 2018; Roselló-Soto et al. 2015; Barba et al. 2015; Puértolas and Barba 2016; Şahin et al. 2017). Therefore, OOBP can be a natural source of antioxidants, especially of phenolic compounds that may be used by several industries as food, cosmetic, pharmaceutical, and nutraceutical industries.

The research in field of bio adsorbents and dye removals have making use of OOBP as adsorbent of heavy metals and dye removals (Konstantinou, Kolokassidou, and Pashalidis

2007; Doyurum and Çelik 2006; Martín-Lara et al. 2008; Malkoc, Nuhoglu, and Dundar 2006; Al-Anber and Matouq 2008; Vegliò, Beolchini, and Prisciandaro 2003; Banat et al. 2007; Akar et al. 2009).

Even though some signs of progress have been seen regarding olive oil by-products management and their valorization, there is no EU regulation regarding the disposal of olive oil by-products, so far. Therefore, each country is responsible for its disposal in nearby lands, lakes, seas, and storage or evaporation in lagoons (Paraskeva and Diamadopoulos 2006).

Hence, at the olive oil production sector, a set of inclusive practices, considering the negative polluting load of olive oil by-products, the disposal, management, and valorization of these by-products, should be implemented.

The obtainment of extracts rich in phenolic compounds that may have protective effects on human health for further incorporation in complex matrices as foods is a research field area that must be explored in the framework of valorization, disposal, and management of olive oil by-products.

Phenolic compounds are widely distributed in the plant kingdom in the form of by-products produced during plants metabolism (Renes and Arci 2004). However, the majority of these compounds have a restricted application in the food industry, as they may be easily modified by external/environmental factors like pH, temperature, and light. After the attainment of the phenolic-rich extracts from OOBP, in order to shield the integrity of extracts, they must be protected from the external environment through coating/encapsulation approaches, finishing in the final formulation (e.g., food matrix) with the capacity of a sustained release of phenolic compounds, without losing their physicochemical properties (Munin and Edwards-Lévy 2011; Fang and Bhandari 2010).

From this perspective, the valorization of OOBP should address the challenge regarding the obtainment of valorized, innovative, and even reinvented products through extraction and microencapsulation approaches.

Valorization of phenolic compounds present in olive oil by-products through extraction and microencapsulation approaches

Regarding the potential bio and phytotoxic impact of the presence of phenolic compounds in olive oil by-products, alternative and innovative approaches have been proposed for the recovery of these valuable compounds from OOBP. Among them, extraction and microencapsulation approaches can be combined to obtain a phenolic-rich extract that can be coated using encapsulating approaches to shield the integrity of the extracts, and be further incorporated in the final food formulation/matrix, allowing the release of the phenolic compounds without any defeat on extracts physicochemical composition, yielding the crude olive oil by-product with a low content on phenolic compounds that can be further used for water irrigation, in the case of OOWW and soil fertilizer in the case of OOP, or even for other alternative applications.

Extraction of phenolic compounds from olive oil by-products

Over the years, the research on extraction techniques allowed to select the commendable extraction approach to be applied to OOBP and optimized them. The extraction technique that should be implemented depends on the type of OOBP–pomaces, wastewaters, and olive leaves–require alternative extraction approaches to obtain phenolic compound rich extracts.

Three key considerations should be reviewed prior to the extraction: (i) the procedures regarding the sample preparation, (ii) the selection of the most appropriate solvent, and (iii) the selection of the most suitable extraction method itself.

The procedures regarding the sample preparation (i) are considered to be an utmost important step as it is envisaged physicochemical modifications on the chemical structure of phenolic compounds. After collection, OOBP samples should be conserved in a liquid nitrogen environment or freeze-dried and storage in moisture-free environments, a desiccator (De Marco et al. 2007; El-Abbassi, Kiai, and Hafidi 2012).

As an alternative, the olive oil by-product sample can be acidified until pH of 2–3 as alkaline mediums promote oxidation and polymerization reactions. Nevertheless, the low pH may promote the precipitation of proteins, which may increase the amount of free phenolic compounds in the solvent used for the extraction (De Marco et al. 2007; El-Abbassi, Kiai, and Hafidi 2012).

During the OOP sample preparation and due to the high content of water of the OOP (circa 60%), it can be submitted to a centrifugation process in order to separate the solid residue from the vegetative waters. Vegetative waters are composed of the water used for olive oil production and the water present in the olive drupes (Suárez et al. 2009).

Prior to extraction, it is typical to remove the lipidic fraction of the OOBP samples. The non-polar compounds as fatty acids, triacylglycerols, fat-soluble pigments, and vitamins, among others may hinder the quantification of phenolic compounds present in the obtained extract. Thereof, non-polar compounds are extracted using the solvent *n*-hexane or petroleum ether (Cioffi et al. 2010; El-Abbassi, Kiai, and Hafidi 2012). Nevertheless, the lipidic fraction can be extracted after the extraction of phenolic compounds. In this case, the use of the solvent mixture methanol/water is commonly applied. However, in the case of the extraction of phenolic compounds from a non-defatted olive oil by-product sample, the phenolic profile of extract may differ from the phenolic profile form a defatted olive oil by-product sample. Generally, non-defatted samples present high quantities of phenolic compounds, probably due to the occurrence of reactions between the fats present in olive oil by-product sample and phenolic compounds (Obied et al. 2007; Alu'datt et al. 2010). The obtainment of a defatted sample prior to the extraction of phenolic compounds is highly recommended.

Extraction techniques of phenolic compounds from olive oil by-products. In the literature review were found many scientific articles regarding the extraction techniques of phenolic

compounds from OOBP. As a comprehensive scientific revisions on this topic can be found on the literature (Caporaso, Formisano, and Genovese 2018; Dermeche et al. 2013; Nguyen 2017), only the more relevant subjects regarding the extraction of phenolic compounds present in OOBP are going to be briefly discussed. An exhaustive description of the extraction procedures of phenolic compounds from olive oil by-products is out of the scope of this paper.

Several authors have proposed optimized extraction procedures of phenolic compounds from OOBP. Among the extraction procedures commonly applied stands out (i) the solid–liquid extraction, (ii) the ultrasound-assisted extraction, (iii) supercritical fluid extraction, (iv) the pressurized liquid extraction and (v) the superheated liquid extraction.

The solid–liquid extraction (i) is usually performed, starting on the homogenization or alternatively maceration of the OOBP sample. The homogenization of the OOBP samples is commonly achieved through vortexing, magnetic stirring, or using high-performance liquid homogenizers as the Ultra-Turrax®. The homogenization step is followed by centrifugation or sedimentation. Then, two phases are obtained (a solid one and a liquid one), and the analytes are distributed according to their polarity (or alternatively to their affinity to the organic solvent added). Afterward, the two phases are separated, and the analytes can be recovered. A critical design step is the choice of the solvent. It must be selected according to the analytes intended to be recovered, the extraction efficiencies, and the final application desired for the extract. This is especially critical when the extract is intended to be earmarked for food applications. An unyielding control of the contamination of extract with solvent is required. Generally, solvent evaporation is performed, and the trace solvent may be removed by freeze-drying, and the control of the presence of trace solvent can be performed through thermogravimetric analysis.

For the extraction of phenolic compounds from OOBP, generally recognized as safe (GRAS) solvents are usually employed. Solvents as methanol, ethanol, or even hydroalcoholic mixtures are frequently used in this context due to their high selectivity to phenolic compounds. The most used one for the recovery of phenolic compounds from olive oil by-products is methanol (Alu'datt et al. 2010; Klen and Vodopivec 2012; Klen and Mozeti 2011). Nevertheless, its concentration in food matrices is limited by the EU regulation 2009/32/EC to a maximum concentration of 10 mg/kg (EC 2009). Still, according to the EU regulation 2009/32/EC, ethanol is considered an extraction solvent in compliance with the good manufacturing practices. Therefore, ethanol is a safer alternative to methanol regarding the extraction of phenolic compounds from OOBP.

Nevertheless, during the choice of solvent, it should be considered that ethanol is more lipophilic than methanol, and therefore, the analytes recovered with ethanol can be as well more lipophilic and structurally complex. To increase the hydrophilicity of ethanol in order to achieve higher recovery rates of phenolic compounds, ethanol can be used at higher temperatures and pressurized as performed by Lozano-Sánchez et al. (2014), Omero et al. (2009) and Peralbo-Molina, Priego-Capote, and Luque de Castro (2012).

Unexpectedly, Lafka et al. (2011) obtained similar recoveries of phenolic compounds using methanol, ethanol, ethanol/water 1/1, and propanol, when they were studying the extraction variables–solvent, pH, time and solvent concentration–on the recovery of phenolic compounds from OOBP.

Therefore, it is recommended that further studies in order to analyze the effect of choosing alternatively ethanol instead of methanol for the recovery of phenolic compounds. The meaningfully results obtained by Lafka et al. (2011) should be corroborated by further studies.

An alternative solvent that can be used for the recovery of low to medium molecular weight phenolic compounds is ethyl acetate. This solvent is also in compliance with the good manufacturing practices according to the EU regulation 2009/32/EC (EC 2009). Notwithstanding, this solvent presents a pellicular odor that can be considered unpleasant if the extract is directly incorporated in foods. To circumscribe that, encapsulation purposes may protect the extract and mask the unpleasant odor of ethyl acetate. The authors De Marco et al. (2007), El-Abbassi, Kiai, and Hafidi (2012), and Omero et al. (2009) studied the application of ethyl acetate in the recovery of phenolic compounds from OOBP.

Conclusively, the solid–liquid extraction of phenolic compounds from olive oil by-products should be accomplished, not only considering the recoveries rates but also the toxicity of the selected solvent. For food applications, the solvent chose for the extraction should be classified as GRAS and recognized as an extraction solvent in compliance with the good manufacturing practices by EU directives.

Generally, lipophilic structures or cell wall polysaccharides with a phenolic moiety are not recovered by solid–liquid extraction using a mixture of solvents (ethanol/water or methanol/water); therefore, it commonly employed hydrolysis to breaking the ester or glycosidic bonds of phenols to the olive oil by-product matrix. The breakage of these bonds in order to improve the recovery in phenols can be performed by acidic, basic, or even enzymatic hydrolysis (Watson 2019). The acidic hydrolysis allows to break both the ester and the glycosidic bonds, and it is usually achieved through the addition of a hydrochloric acid solution. In the case of basic hydrolysis, it can be added sodium or potassium hydroxide solutions. However, it only allows breaking essentially the ester bonds. In the case of enzymatic hydrolysis, specific enzymes should be used–esterases to break ester bonds and glycosidases for the breakage of glycosidic bonds (Watson 2019).

During the research regarding the optimization, characterization, and quantification of phenolic compounds present in OOBP, Alu'datt et al. (2010) recovered the free phenols by solid–liquid extract with methanol and the bounded phenols through alkaline followed by acid hydrolysis later to the extraction of free phenolic compounds. For the recovery of bounded phenols, the authors promoted the ester and glycosidic bounds breakage using firstly a sodium hydroxide solution (pH of 12) followed by acidic hydrolysis

using a hydrochloric acid solution (pH 2). The authors determined that the amount of bounded phenols corresponded from 13% to 25% of the total amount of phenols recovered.

A straightforward alternative for the recovery of phenolic compounds from OOBP is the (ii) ultra-sound assisted extraction. It comprises the application of high-frequency waves ($\geq$ 2 MHz). These high-frequency waves are responsible for the generation of a negative pressure that changes the physicochemical properties of the medium, allowing the development of small cavitation bubbles (Chen, Sharma, and Mudhoo 2012). The formation of these cavitation bubbles allows to disrupt cell membranes and therefore favor the contact between the extraction solvent and the matrix. It can be performed using an ultrasonic bath or a probe. In the case of using an ultrasonic bath, the high-frequency waves reach the sample through the wall of the sample container, while in the case of using a probe, the high-frequency waves reach the sample directly as they are immersed in the extraction solution. Beyond being a swift and easy to handle extraction method, it does not require high temperatures, which makes this method suitable for the extraction of temperature-sensitive compounds as phenolic compounds from OOBP.

The authors Klen and Mozeti (2011) performed an ultrasonic extraction of phenolic compounds from OOWW and compared with the conventional methods. The authors accomplished five alternative extraction procedures: filtration, solid-phase extraction, liquid–liquid extraction, and ultrasonic-assisted extraction of the liquid OOWW and the freeze-dried OOWW. They verified that the use of ultrasonication allowed to obtain higher phenolic recovery yields. Comparing the extraction in an organic solvent of phenolic compounds from OOWW without previously ultrasonication was less efficient than the ultrasound-assisted counterpart. However, the authors did not obtain a representative phenol chromatogram probably due to the use of ethyl acetate as the extraction solvent–ethyl acetate probably induced modifications. Contrasting, the ultrasound-assisted extraction of the freeze-dried OOWW using methanol as the extraction solvent allowed to obtain high recovery yields of phenolic compounds without significant ultrasonic-assisted extraction modifications. Additionally, as described in section "Extraction of phenolic compounds from olive oil by-products," the freeze-drying of OOWW prior to extraction, prevents significant physicochemical modifications of the sample.

A recent alternative to the reported extraction procedures is (iii) the supercritical fluid extraction with carbon dioxide. The application of the technique is based on the fact that the solvating properties of a fluid can be improved, exceeding the fluid critical pressure and temperature. When in the form a supercritical fluid, it displays a higher diffusion through the sample matrix, allowing the obtainment of higher recovery rates (Souza et al. 2013). The most commonly employed supercritical fluid is carbon dioxide. According to the EU 2009/32/EC regulation, carbon dioxide can be used as an extraction solvent in compliance with manufacturing practices, which can be used during the processing of raw materials, foodstuffs, food components, or ingredients (EC, 2009).

The supercritical fluid extraction with carbon dioxide presents many advantages as the critical temperature is considerably low (31 °C), and it is associated with low toxicity and reactivity. Nevertheless, as phenolic compounds are relatively polar, it is required the addition of a polar modifier (e.g., methanol, ethanol).

The study performed by Lafka et al. (2011) regarding the quantification of phenolic compounds and the antioxidant activity of OOWW suggests that recovery of phenolic compounds is lower when the supercritical fluid extraction with carbon dioxide is applied when compared to the traditional liquid-–liquid extraction. The amount of phenolic compounds recovered from olive oil wastewaters using the supercritical fluid extraction was only higher to the amount of phenolic compounds recovered by conventional liquid-liquid extraction using ethyl acetate as an organic solvent.

The authors Taamalli et al. (2012) investigated the use of different extraction techniques of phenolic compounds from olive leaves (liquid-solid extraction, microwave-assisted extraction, and pressurized liquid extraction) in order to identify the phenolic profile of obtained extracts and their activity against human breast cancer cells. Similarly to the results obtained by Lafka et al. (2011), the supercritical fluid extraction shown to be the extraction technique with the lowest recovery rates on phenolic compounds. Moreover, the authors verified that applying the supercritical fluid extraction allowed to obtain high recovery rates of lipophilic phenolic compounds as luteolin and apigenin.

The leading innovative extraction procedure of phenolic compounds from OOBP are (iv) the pressurized liquid extraction and the related (v) superheated liquid extraction. These extraction techniques dwell on the employment of organic solvent combined with temperature and pressure that are controlled externally and electronically selected. The pressurized/heated liquid extraction system is composed by a stainless-steel chamber or cell where the sample is placed, an oven, a pump used for the inlet of the solvent into the system and a set of valves that allows preventing temperature or pressure drops (Carabias-Martínez et al. 2005). Both techniques can operate in three alternative operating modes: (i) the dynamic mode in which fresh solvent is continuously pumped to the system/extraction medium, (ii) the static mode in which the fresh solvent is only pumped between extraction cycles and (iii) the semi-dynamic operating mode which is a combination of the dynamic and static operating modes.

The solvents used in this technique for the extraction of phenolic compounds are generally mixtures of methanol/water or ethanol/water, even though other solvents as acetonitrile, ethanol, methanol, and ethyl acetate are frequently employed in the extraction of hydrophilic compounds (Lozano-Sánchez et al. 2014; Carabias-Martínez et al. 2005; Paper 2008; Herrero et al. 2011; Omero et al. 2009). Pressure and temperature contribute differentially to the extraction efficiency. The pressure on the system does not

contribute significantly for the efficiency of extraction as it is mostly used to maintain the solvents in the liquid phase and improve the flow of the solvents, and to promote the contact between the solvent and the extract (Lozano-Sánchez et al. 2014; Taamalli et al. 2012). Contrastingly, the temperature is considered an essential factor that may significantly influence the efficiency of extraction. When water is used as a solvent or more commonly, in a solvent mixture generally, temperatures above 100 °C and below the critical temperature of water (circa 374 °C) are employed for the extraction of phenolic compounds. Temperatures below 100 °C are not recommended as water is too polar for the extraction of organic compounds. Above 100 °C, water presents a lower dielectric constant and therefore acquires a less polar character, more suitable for the extraction of organic compounds. Temperatures above the critical temperature of water are also not recommended as many compounds are degraded at this temperature. The employment of high temperatures contributes to the dissociation of the more lipophilic phenolic compounds from the matrix, allowing, therefore, to extract them. Other factors as the flowrate of solvents and time are considered to be significant for the extraction efficiency (Kronholm, Hartonen, and Riekkola 2007).

There are many advantages associated with the employment of these innovative extraction techniques (pressurized liquid extraction and superheated liquid extraction) for the recovery of phenolic compounds from OOBP intended to further be coating and inclusion on food matrices. Compared to the conventional solid–liquid and liquid–liquid extractions, the pressurized liquid and superheated liquid extractions present a higher degree of automation, require less solvent, and they are less time-consuming approaches. Moreover, it is generally possible to achieve efficient recoveries of phenolic compounds using GRAS solvents like ethanol and water.

The authors Taamalli et al. (2012) verified when they were studying the use of advanced techniques of the extraction of phenolic compounds from Tunisian olive leaves, that amount of phenolic compounds recovered was higher using the pressurized liquid extraction technique with a solvent mixture of methanol and water when compared to the employment of a supercritical fluid and microwave-assisted extractions. Nevertheless, the results obtained by Suárez et al. (2009) indicate that a higher recovery of phenolic compounds was achieved using similarly a mixture of methanol and water but applying the solid–liquid extraction.

Nowadays, are available many extraction techniques that allow the recovery of phenolic compounds from OOBP. During the selection of the extraction technique of phenolic compounds intended to be coated and included in food matrices, should be considered the recovery of phenolic compounds, if it is desired the recovery of lipophilic or/and hydrophilic phenolic compounds, if the solvent or the solvent mixture is GRAS, the maximum concentration of solvent allowed in food matrices by the EU regulation 2009/32/EC, the amount of solvent required for the extraction, the time-consumption of the extraction procedure as well as its energy consumption.

Microencapsulation of phenolic-rich extracts obtained from olive oil by-products

Considering that the majority of phenolic compounds present in olive oil by-products possess limited application on the food industry since they are promptly amended by environmental factors like temperature, pH, and light, they must be protected previously ending in the final formulation. In the specific case of foods, prior to the fortification of them with phenolic-rich extracts obtained from OOBP, they should be protected from the surrounding environment. Moreover, it is generally intended a sustained release of phenolic compounds in fortified foods, without losing their physicochemical properties. Therefore, microencapsulation arises a potential technological strategy to protect phenolic-rich extracts obtained from OOBP to be further incorporated in food matrices.

Microencapsulation is a general term that encompasses a set of technological techniques that allows the protection and coating of micro-sized substances (solid, liquid, or gases) from the surrounding external environment. Moreover, this set of techniques allows a delayed or sustained release of the incorporated bioactive compounds (Benita 2006; Ghosh 2006; Kaur and Kakkar 2014; Paulo and Santos 2017). The bioactive compound or extract is termed as core material, or encapsulated material, internal phase, or fill, and the material used for the coating purposes is termed as encapsulating material, shell, wall material or membrane (Fang and Bhandari 2010). The encapsulated material can be temporary or permanently protected from the external environment (Carvalho, Estevinho, and Santos 2016; Casanova, Estevinho, and Santos 2016). The resulting products of the encapsulation techniques are designed microparticles. Particles can be distinguished according to their internal morphology in microcapsules and microspheres. Microcapsules and microspheres are differentiated in reservoir systems (the core is surrounded by an outside layer) or matrix systems (the core is homogeneously dispersed within the encapsulating material), respectively (Jyothi et al. 2010; Paulo and Santos 2017).

The microencapsulation technology was first presented by Green and Schleicheir in the 1950s by means of a patent registration regarding the preparation of capsules containing dyes intended to be incorporated into paper for copying purposes (Ghosh 2006; Paulo and Santos 2017). Microencapsulation technological strategies have been investigated by pharmaceutical (68%), food (13%), cosmetic (8%), textile (5%), biomedical (3%), agricultural (2%) and electronic (1%) industries (Casanova and Santos 2016; Ghosh 2006; Kim et al. 2007; Paulo and Santos 2017).

The second driving force of significant signs of progress in microencapsulation is the food industry. The high-demand of fortified and functional products by many consumers is the major motivation for the intensive investigation in this research field.

Additionally, to their sensitivity to the external environment (light, pH, and temperature), phenolic compounds-rich extracts are prone to oxidation processes and gastrointestinal degradation. Moreover, they may present an unpleasant flavor. Therefore, microencapsulation may foster the resolution these issues related to the direct incorporation of phenolic-rich extracts in foods. The incorporation of phenolic-rich extract loaded microparticles and the delayed or sustained release of phenolic compounds may aid to prevent the oxidative degradation of other vital molecules present in the food matrix; therefore, phenolic-rich extracts loaded microparticles can be added to foods as a high-tech preservatives, extending foodstuff shelf-life. Moreover, the protection of phenolic compounds from the external environment may increase the bioavailability of these compounds (Chen et al. 2019).

One of the main advantages of microencapsulation approaches is the possibility to control the release of bioactive compounds. The sustained release of phenolic compounds from microparticles incorporated in foods may reduce the amount of additives added to preserve them and extend foods shelf-life (Azeredo 2005). The release can be classified as (i) delayed–when the release is intended to occur in a specific moment or place–or (ii) sustained–when is intended to control the release rate of bioactive compound(s). Generally, the actual release is a combination of both patterns (Aguiar, Estevinho, and Santos 2016).

The release can occur by mass-diffusion through water-filled pores or throughout the polymer, osmotic pumping or even through erosion processes of the coating material which do not require transport of the bioactive compound(s) contrarily to the other release mechanisms (Fredenberg et al. 2011). There are many properties of the release pattern and the external environment that may influence the actual release. The properties affecting the delivery system can be divided in five alternative categories: (i) the coating material (molecular weight of the polymer, the chemical end groups of the polymer, the crystallinity state of the polymer), (ii) the encapsulated agent (the physicochemical characteristics of the bioactive compound(s), loading, location inside of microparticles, the presence of additives as salts, surfactants or even plasticizing agents), (iii) the delivery system itself (size, porosity, density and shape), (iv) in vitro release conditions (temperature, stirring, composition of the release medium, pH and osmolality) and (v) in vivo release conditions (presence of enzymes, lipids, physicochemical characteristics of the matrix) (Fredenberg et al. 2011).

The most common release mechanism of bioactive compounds, including the case of phenolic-rich extracts is by osmotic pumping, generally by a solvent-activation approach. Nevertheless, the use of surfactants, the control of the pH, the induce of enzymatic release, and even light may trigger the release of bioactives from microparticles in a food matrix (Aguiar et al. 2017).

The design of fortified foods with microencapsulated phenolic-rich extracts must consider several constraints as the manufacturing and storage properties of foods, economic feasibility, and consumer adherence (Aguiar, Estevinho, and Santos 2016; Paulo and Santos 2017). Additionally, particle size is considered to exert a significant influence on microparticles design for food applications as the presence of particles larger than 30 μm may irk a gritty mouthfeel (Merkus and Meesters 2014).

Microencapsulation techniques for the entrapment of phenolic-rich extracts obtained from olive oil by-products. Over the years, new and innovative microencapsulation techniques have been studied and optimized considering the utmost important factors as (i) physicochemical properties of the core material (ii) physicochemical properties of the wall material and (iii) the final intended application of obtained microparticles (Poshadri and Kuna 2010).

There are available a significant variety of microencapsulation techniques; however, no single encapsulation technique is adjustable to all core materials, polymers, and applications (Wilson and Shah 2007).

Some of the most used encapsulation techniques for food applications are the spray-drying, the freeze-drying, the coacervation, the emulsification, the inclusion complexation, the liposome entrapment, and the fluid bed coating.

Mostly, the encapsulation techniques applied for the entrapment of phenolic-rich extracts from OOBP are the spray-drying, the freeze-drying, and emulsification techniques. Among the emulsification techniques/solvent evaporation approaches, the double emulsions water-in-oil-in-water and the oil-in-water-in-oil are described for the encapsulation of olive leaf extracts. The supercritical assisted atomization is an innovative alternative also described for the protection of OOP extracts. In Table 2 are presented the main advantages and disadvantages of each technique found in the literature for the microencapsulation of phenolic-rich extracts obtained from olive oil by-products for food applications.

Comprehensive reviews of microencapsulation techniques are available in the literature (Aguiar, Estevinho, and Santos 2016; Fang and Bhandari 2010; Ozkan et al. 2019). Therefore, only a brief description of the microencapsulation techniques used for the entrapment of phenolic-rich extracts obtained from OOBP is presented.

The spray-drying technique is the most outspreading technique for the encapsulation of phenolic-rich extracts obtained from olive oil by-products intended for food applications. The spray-drying can be defined as an atomization process of a liquid in a dry powder using an injector and a hot drying chamber (Rattes and Oliveira 2007). During the spray-drying, occurs the homogenization of the feed solution using a an atomizer, followed by the drying of feed solution in a chamber passing a hot carrier gas to promote the evaporation of the solvent and finally, microparticles are recovered in a cyclone or a filter (Schafroth et al. 2012; Ozkan et al. 2019). The feed solution contains the core and the wall materials in a solution, emulsion, or a suspension form (Gharsallaoui et al. 2007).

The feature characteristics of the powder obtained by the spray-drying technique depend largely on operating

Table 2. Advantages and disadvantages of microencapsulation techniques of phenolic-rich extracts obtained from olive oil by-products.

Method	Advantages	Disadvantages	References
Freeze-drying	- Capacity of retaining high amount of antioxidants - Simple process - Low operating temperature - Drying in the absence of an oxidation atmosphere (e.g., air) - Suitable technique for the coating of heat-sensitive and/or water-soluble compounds	- Multi-stage process - Long-time process - High operating costs - Presence of porous structures during the sublimation of ice - Difficult control of particle size distribution	Ozkan et al. (2019)
Spray-drying	- Scalable - Fast procedure - Allows continuous operation - Low moisture content of the final powder - Economic - Reproducible	- Not suitable for the coating of thermosensitive compounds - High energy demand and sophisticated equipment - High possibility of compounds degradation - Low yields of production due to loss of particle on the drying vessel - Wide particle size distribution - Difficult control of particle size and shape - Selection of wall materials is limited - Wall materials with a low glass transition temperature and stickiness are challenging to process	Aguiar, Estevinho, and Santos (2016) Ozkan et al. (2019)
Emulsification/Solvent Evaporation	- Includes a broad range of approaches as water-in-oil-in-water, oil-in-water-in-oil, water-in-oil, water-in-oil, and multiple emulsions. - Easy to scale up - Accessible to scale-down (e.g., production of nanoparticles) - Use of mild conditions - Lower residual solvent content - No change on the activity of the bioactive compounds coated	- Use of organic solvents - Time-consuming technique	Hwisa et al. (2013) Aguiar, Estevinho, and Santos (2016) OOzkan et al. (2019)
Supercritical Assisted Atomization	- High-performance technique - Continuous operating mode - High selectivity due to the possibility to adjust both temperature and pressure - Mild conditions operation - Solventless or amount of solvent significantly reduced - Easy to scale-up	- Generally, carbon dioxide is used as the solvent; however, it is not a suitable solvent for polar compounds - Difficult to coat bioactive compounds using polymers with a low glass transition temperature - Adjustable for the incorporation of hydrophobic compounds. Prior modifications on hydrophilic compounds are required in order to be coated (e.g., water-soluble proteins)	KKesente et al. (2017)

conditions as the feed solution flow rate, the drying temperature, the drying air flow rate, the speed on the atomizer, and the polymer carrier (type and concentration) (Schoubben et al. 2010).

The carriers used for the encapsulation by spray-drying are generally water-soluble polymers as polysaccharides (e.g., Arabic gum, maltodextrins, cyclodextrins), proteins (e.g., sodium caseinate, whey protein, soybean protein), starch, gelatin, chitosan and gellan gum (Lee and Wong 2014).

Additionally to the application for the coating and protection of phenolic-rich extracts obtained from OOBP, the spray-drying has been used for the microencapsulation of a broad range of bioactive compounds for food industry as colors, vitamins, flavors, fats, oils and antioxidant-rich extracts (Aguiar, Estevinho, and Santos 2016; Ferris, Fine, and Hyman 1984; Nedovic et al. 2011; Pillai et al. 2012).

An alternative microencapsulation technique that has been widely employed for the encapsulation of phenolic-rich extracts from OOBP is the freeze-drying technique, also termed as lyophilization. This is a multi-stage process in which the mixture of the wall and the core materials is submitted to a freezing process followed by a sublimation known as primary drying. Then, it occurs the desorption (secondary drying) and the storage of the dry powder (Laokuldilok and Kanha 2015). It is described that wall material characteristics - structure and composition - seize an impact on both adequate protection and release (Young, Sarda, and Rosenberg 1993).

The freeze-drying technique has been used for the encapsulation of several bioactive compounds for food industry as antioxidants (Wilkowska et al. 2016; Galmarini et al. 2013), food flavors (Kaushik and Roos 2008), enzymes (Liu et al. 2011; Santagapita, Mazzobre, and Buera 2011; Anjani, Kailasapathy, and Phillips 2007; Mehrnoush et al. 2011; Kawai and Suzuki 2007) and probiotics (Morgan et al. 2006; Song et al. 2008; Saarela et al. 2006; Carvalho et al. 2008).

The microencapsulation through emulsification and solvent removal processes can be generally defined as a set of procedures in which an organic solvent is removed from an emulsion dispersed in another solvent, generally water. The emulsion contains embedded the core material, and the wall material dissolved in an organic solvent (Poncelet 2005). According to this technique, firstly an emulsion, suspension or a solution of the core material and the wall material is formed. The coating polymer is previously dissolved in an organic solvent. Afterward, occurs the emulsification of the

dispersed phase (core and wall material) in a continuous phase (aqueous solution) generally by stirring, even though it can be accomplished by static mixing, dripping, or extrusion. After that, the solvent removal is promoted through evaporation and liquid extraction. The solid powder obtained after the solvent removal is recovery by filtration or centrifugation from the dispersive phase. The obtained microparticles are subsequently dried (Hwisa et al. 2013; Paulo and Santos 2018a, 2018b; 2018c).

There are many formulation and operating conditions that affect the microencapsulation through emulsification followed by solvent removal procedures as the nature of the solvent, the volume of solvent, the physicochemical properties of the polymer, the polymer concentration, the emulsification type, the concentration of emulsifiers, the rate of solvent removal, the addition of salts or buffers to the internal phase and/or external phase, the continuous to dispersive phase volume ratio, the temperature, the stirring speed among others (Tiwari and Verma 2011).

Some authors claim that there is a lack of studies regarding the impact of this set of encapsulation approaches intended for food applications, even though they have been widely investigated for pharmaceutical applications (Ozkan et al. 2019). However, according to Lu, Kelly, and Miao (2016), during their scientific review on emulsion-based encapsulation and delivery systems of phenolic compounds, there is a vast number of studies regarding the encapsulation of phenolic compounds by emulsification approaches intended to obtain microparticles to be further incorporated in foods. Nevertheless, the authors reported that they found very little information regarding the in vivo absorption, transportation, and release of these compounds enclosed in emulsions. Therefore, agreeing with Lu, Kelly, and Miao (2016), a systematic and comprehensive research on the metabolism and effects of encapsulated phenolic compounds in emulsions is imperative.

The innovative and alternative microencapsulation technique supercritical assisted atomization is based on the use as a solvent, a supercritical fluid at temperature (T_c), and pressure (P_c) above its critical point. Above its critical point, the selected solvent acquires properties between liquids and gases such as the density and the solvating capacity of liquids and the typical low viscosity, high diffusivities, and high mass transfer rates of gases. The most commonly used solvent as a supercritical fluid is carbon dioxide due to its mild critical conditions (T_c=31.1 °C and P_c=7.38 MPa). Nevertheless, examples of other solvents that can be used as supercritical fluids are water, nitrogen, and propane (Gouin 2004). The supercritical assisted-atomization processes can be classified as (i) rapid expansion of supercritical solutions and related processes, (ii) supercritical antisolvent precipitation and derived processes, and (iii) particles from gas saturated solutions and the related processes (Munin and Edwards-Lévy 2011).

In the case of a rapid expansion of supercritical solutions (i), the supercritical fluid acts as a solvent. The bioactive compound(s), other solutes, and the polymer are dissolved in the supercritical fluid. Afterward, the solution is expanded using a small nozzle into a region with a lower pressure, which allows the precipitation of the solutes (Debenedetti et al. 1993).

In the case of supercritical antisolvent precipitation, the supercritical fluid acts as anti-solvent. The technique is based on the contact between the supercritical fluid with a solution containing the organic solvent and the bioactive compound(s) and other solutes. The contact between the solution containing the bioactive compound and the supercritical fluid is promoted through the injection of the solution through a nozzle into a pressurized chamber. When the supercritical fluids contact with the solution, the solubility of bioactive compound(s) and other solutions in the atomized particles is reduced, resulting in supersaturation, nucleation, and formation of microparticles. The organic solvent is then eliminated through a continuous flow of the supercritical fluid (Visentin et al. 2012; Mattea, Martín, and Cocero 2009; Sosa et al. 2011).

In the case of particles from gas saturated solutions procedure (iii), the supercritical fluids act as a solute. The bioactive compound(s) and other solutes are saturated with the supercritical fluid. Then, occurs the expansion of the gas-saturated solution through an atomization nozzle, which allows the formation of microparticles. During the release of the supercritical fluid, a cooling effect is verified. The occurrence of this cooling effect is directly responsible for the formation of microparticles (Mattea, Martín, and Cocero 2009; Ozkan et al. 2019).

Valorization of phenolic-rich extracts from olive oil by-products through microencapsulation. To the authors best knowledge, this is the first review focused on the main studies regarding the encapsulation of olive oil by-products such as olive leaves, olive oil pomace extracts, olive oil wastewater extracts, and even olive kernel (search criteria: all databases; keywords: olive leaves, olive stones; olive kernel; olive oil wastewater; encapsulation; microencapsulation; August 2019; studies regarding the encapsulation of phenolic-rich extracts obtained from OOBP cosmetic and pharmaceutical applications were not considered for this literature review). Studies regarding the encapsulation of extracts from OOBP are presented in Table 3.

An incomparable number of studies regarding the encapsulation of olive leaves extracts were found compared to the ones found concerning the encapsulation of extracts from other olive oil residues (pomaces, wastewaters and olive kernel). A thesis that allows supporting this observation is proposed now on by the authors of this review article. This is probably related to the fact of olive leaf extracts are commercially available, while in the case of extracts from olive oil pomace, olive oil wastewater and olive kernel are always necessary the prior obtainment of the raw material for encapsulation–the extract. Moreover, some studies reported the use of a spray-dryer to obtain extract-loaded microparticles, which is straightforward in the case of extracts of lipophilic nature, as is the case of olive leaves, since the process consists in the formulation of an oil-in-water emulsion which is then dried in an atomizer chamber. Therefore,

Table 3. Studies regarding the encapsulation of olive oil by-products extracts.

Core	Shell	Method	Objectives	Results	Ref.
Olive leaf extract	β-cyclodextrin	Freeze-drying	- Study the inclusion of OLE through DSC and nuclear magnetic resonance spectroscopy Determination of OLE solubility in the presence of β-cyclodextrin by a phase solubility study	- Oxidative DSC studies indicated that the complex of OLE with β-cyclodextrin was protected against oxidation since it remained intact at temperatures where the free OLE was oxidized - Phase solubility studies demonstrated that inclusion OLE in β-cyclodextrin increased the aqueous solubility of the polyphenolic residue from the OLE by more than 150%	
	Sodium caseinate-soy lecithin	Spray-drying	- Evaluation of the retention of AA	- Maximum radical scavenging activity $\sim$ 60% (OLE concentration of 0.5 mg/mL)	
	Insulin	Spray-drying	- Optimization of encapsulation conditions using a DoE (CCD) - Determination of EE of oleuropein, a compound present in the OLE -Study the effect of microencapsulation and the frying method on the TPC and AA, fat content, and crispness, after adding MPs to starch–gluten fried matrices	- TPC of OLE: 64.3 mg of gallic acid equivalents/g of dry leaves - OLE PY: 90.1% -EE of oleuropein: 87.1% - TPC in MPs: 13.2 mg of gallic acid equivalents/g of MPs - The frying method affects the content of polyphenols - Vacuum fried matrices show high phenolic content than atmospheric fried matrices	Urzúa et al. (2017)
	Poly(lactic acid)	Nanoprecipitation	-Encapsulate OLE in poly(lactic acid) NPs - Characterization the NPs and define the experimental parameters that affect the encapsulation procedure. -Incorporation of NPs in a cosmetic formulation - Study the stability of the formulation for 3 months	-NPs exhibited anionic ζ-potential - Burst release at the first 2 hours - Increased stability of loaded NPs compared to the extract regarding viscosity, pH among others	Kesente et al. (2017)
	Complex of whey protein concentrate and pectin	Water-in-oil-in-water emulsification	- Evaluation of AA of OLE encapsulated through a nano-emulsification approach	- TPC: 206.81 $\pm$ 0.02 mg of gallic acid equivalents/g of sample - Nanoencapsulation of OLE in whey protein concentrate - pectin complex improved AA and allowed a sustained release of bioactive compounds	Mohammadi et al. (2016)
	Arabic gum and maltodextrin	Oil-in-water-in-oil emulsification	- Evaluation of the AA of OLE encapsulated by Arabic gum and maltodextrin, in soybean oil at OLE concentration of 70 mg/kg - Evaluation of storage stability of OLE-Arabic gum and maltodextrin-soybean oil for 20 days	- Slow-release of phenolic compounds: 20 to 30 mg/kg of phenolics was not released after 20 days storage at 55 °C (initial inclusion amount of 70 mg/kg) - Encapsulation can protect phenolic compounds OLE from decomposition during storage at 55 °C - The AA of encapsulated extracts was lower than the corresponding non-encapsulated contrapeneur	Taghvaei et al. (2014)
	Complex of whey protein concentrate and pectin	Water-in-oil-in-water emulsification	- Study the behavior of water-in-oil-in-water double emulsion encapsulating OLE in a complex of whey protein concentrate and pectin	- EE (day 1): In whey protein concentrate only: 93.34% In whey protein concentrate-pectin complex: 96.64% - EE (day 22): In whey protein concentrate only: 72.73% In whey protein concentrate-pectin complex: 88.81% - Total amount released (20 days, 30 °C): Using only whey protein concentrate: 22% Using whey protein concentrate-pectin complex: 8.1%	Mohammadi et al. (2016)
	Chitosan	Spray-drying	- Preparation of chitosan OLE loaded MPs - Understand the structural interactions caused by the inclusion of the extract in chitosan MPs by FTIR and DSC	- From FTIR studies was verified that occurred interaction between the hydroxyl/carboxyl/aldehyde groups of the OLE and the amine - the functionality of the chitosan molecules	Kosaraju, D'ath, and Lawrence (2006)

(*continued*)

Table 3. Continued.

Core	Shell	Method	Objectives	Results	Ref.
			- Estimation of the encapsulation loading of phenolic compounds	- From DSC studies was verified a shift in Tg from PO-MP to L-MP supporting the efficient encapsulation of polyphenolic compounds in chitosan-based matrices - Loading content of 27% in gallic acid equivalents	
	Maltodextrin	Freeze-drying	- Choose the best extraction technique comparing conventional versus alternative extraction techniques - Perform the encapsulation of the extracts - Evaluation of MPs hygroscopicity, solubility, MC, and EE	- MPs obtained by conventional extraction (1 h, 60 °C) showed the best encapsulation features: EE: 91.06 ± 1.40% MC: 0.78 ± 0.04% Water activity: 0.16 ± 0.02 (at 25 °C) Hygroscopicity: 26.59 ± 0.07 g H_2O/ 100 g of dry weight Solubility: 98.19 ± 0.12% AA: 0.61 ± 0.05 mg Trolox equivalents/g of raw material	Chanioti, Siamandoura, and Constantina (2016)
Kernel/ stones extract	Maltodextrin	Freeze-drying	- Choose the best extraction technique comparing conventional versus alternative extraction techniques - Perform the encapsulation of the extracts - Evaluation of MPs hygroscopicity, solubility, MC, and EE	- MPs obtained by conventional extraction (1 h, 60 °C) showed the best encapsulation features: EE 90.12 ± 1.87% MC: 0.99 ± 0.01% Water activity: 0.16 ± 0.00 (at 25 °C) Hygroscopicity: 22.88 ± 0.13 g H_2O/ 100 g of dry weight Solubility: 97.41 ± 0.11% AA: 1.25 ± 0.05 mg Trolox equivalents/g of raw material	Chanioti, Siamandoura, and Constantina (2016)
Olive pomace extract	Maltodextrin	Supercritical Assisted Atomization	- Study the effect of the ratio of maltodextrin content to the total solid content of the extract and drying temperature on physical characteristics, TPC and AA of the powdered product	- TPC: 105.0 ± 0.1 mg of caffeic acid equivalents/g of dry powder - AA: 98.8 ± 3.0 mg of DPPH/mL of extract - WSIs: 53.0 ± 0.7– 67.0 ± 1.0% - WAIs: 0.28–0.58 g/g of dry powder - PY: 46.0 and 65.3% -SC higher than 2.4 ± 0.0 g/g of dry powder, with the maximum SC obtained of 3.2 ± 0.0 g/g of dry powder when the extract was encapsulated using 30% of encapsulating material and treated at 75 °C - MC lower than 6% w/w	Aliakbarian et al. (2017)
	Maltodextrin	Spray-Drying	- Study different concentrations of maltodextrin as a coating material - Study the effects of inlet temperature (130 °C and 160 °C) and feed flow (5 mL/min and 10 mL/min) in the spray-dryer - Evaluation of AA and stability of MPs	- The high inlet temperature was associated with lower MC and bulk density without affecting AA - The increase of maltodextrin concentration caused lower bulk density and higher MPs sizes, while higher feed flow leads to an increased MC - Fixing the operational conditions at an inlet temperature of 130 °C, maltodextrin concentration at 100 g/L and a feed flow of 10 mL/ min resulted in: PY: 94 ± 0.4% EE: 76 ± 3.3% TPC: 39.5 ± 4.9 mg of caffeic acid equivalents/g of dry powder AA: 33.8 ± 4.3 mmol of DPPH/L of extract - MPs were stable at 5 °C in dark condition for 70 days, and only 21% were degraded, increasing storage temperature up to 25 °C. Light exposure resulted in a 66% loss in polyphenols after 48 h	Paini et al. (2015)
	Maltodextrin	Spray-Drying	- Optimize spray-drying conditions for the MPs of olive pomace extract, stabilizing its phenolic compounds. - Optimization approaches study using a RSM or ANN - Optimization variables considered were: Maltodextrin concentration: 100–500 g/L Inlet-drying temperatures: 130–160 °C Drying compressed flowrates: 20–32 m3/h	- At optimized conditions: PY: 65–82% MC: 9–14 μg/100 g of dry powder WSI: 64–65% Specific TPC: 38–52 mg of caffeic acid equivalents/g of dry powder Specific AA: 230–487 μg of Trolox equivalents/ g of dry powder EE: 85–92%	Aliakbarian et al. (2018)

(*continued*)

Table 3. Continued.

Core	Shell	Method	Objectives	Results	Ref.
Olive Oil Wastewater extract	Whey protein isolate and xanthan gum	Spray-drying	- Study the effects of OOWW extract concentration, whey protein isolate concentration, xanthan gum concentration on physical stability, viscosity, emulsion droplet size, particle size distribution primary and secondary oxidation products, under accelerated storage conditions. - Application of a RSM as an DoE for process optimization	- The effects of OOWW phenolic extracts, the concentration of both whey protein isolate and xanthan gum were statistically significant on emulsion creaming rate, viscosity among other responses studied - The performance of the RSM applied was considered accurate for creaming rate and viscosity	
	Modified starch	Freeze-drying	- Enrichment of non-encapsulated and encapsulated OOWW extracts of yogurt	- TPC: 20% w/w o - A protective effect against undesirable pH drop during yogurt storage was verified	Petrotos et al. (2012)

AA – Antioxidant Activity; ANN -Artificial Neural Network; CCD – Central Composite Design; DoE – Design of Experiments; EE – Encapsulation Efficiency; FTIR – Fourier Transform Infrared Spectroscopy; L-MPs – Loaded-Microparticles; MC – Moisture Content; MPs – Microparticles; NPs – Nanoparticles; OOWW – Olive oil wastewater; OLE – Olive Leaf Extract; PO-MPs – Polymer-only Microparticles; PY – Product Yield; Ref – References; RSM – Response Surface Methodology; SC – Swelling Capacity; Tg – Glass Transition Temperature; TPC – Total Phenolic Content; WAI – Water Absorption Index; WSI – Water Solubility Index;

lipophilic extracts are easily emulsified in aqueous phases containing water-soluble polymers.

As regards, the majority of phenolic compounds present in the olive drupes are retained in aqueous phases as the octanol/water partition coefficients of these compounds are, generally, favorable to the aqueous phase. Therefore, a considerable amount of phenolic compounds (around 98%) are retained in OOBP, such as in OOWW (around 53%) and OOP (approximately 45%) (El-Abbassi et al. 2014).

Dissimilarly, the incorporation of hydrophilic extracts is not favored by the spray-drying technique. Therefore, alternative approaches as emulsification techniques should be applied to obtain extracts-loaded microparticles from OOWW and OOP. Contrary to extracts obtained through the spray-drying technique, these extracts should be incorporated in polymers soluble in organic solvents.

Concerning the encapsulation of olive leaf extracts, in the eight studies found, three of them presented results regarding the encapsulation of extracts through the spray-drying process. Out of three studies found had employed emulsification strategies to encapsulate olive leaf extracts. The application of nanoprecipitation was proposed by Kesente et al. (2017) as an alternative encapsulation approach for the incorporation of extracts in a polymeric shell intended for cosmetic applications; therefore, this study was not included in the revision presented in Table 3. Additionally, two studies regarding the encapsulation of olive leaf extracts by freeze-drying were also found in the literature review.

A considerable variety of polymers from natural to synthetic origin were used as encapsulating materials of olive leaf extracts as insulin, soybean oil, chitosan, maltodextrin, sodium caseinate, and Arabic gum. The authors Taghvaei et al. (2014) used a blend of soybean oil with maltodextrin to study the storage stability of the encapsulated extract. On their turn, Mohammadi et al. (2016) used a complex of whey protein concentrate and pectin to study the release behavior during mid-term storage conditions.

Regarding the encapsulation of OOP extracts, all of the three studies found in the literature used maltodextrin as the encapsulating agent, being the spray-drying technique the most applied one. Supercritical assisted atomization was proposed as an alternative encapsulation technique for the inclusion of OOP extracts in maltodextrin polymeric matrix by Aliakbarian et al. (2017).

About the encapsulation of OOWW extracts, only two studies were found. They evaluated the possibility of incorporation of OOWW extracts in polymeric matrices by spray-drying and freeze-drying encapsulation processes.

The inclusion of olive leaf extract in a β-cyclodextrin matrix was studied by Ourtzinos et al. (2007). Results demonstrate some of the main advantages of using inclusion/encapsulation techniques: the extract was efficiently protected against oxidation. Complexes were stable at high temperatures, unlike the pure extract. Additionally, the olive leaf extract in β-cyclodextrin increased the aqueous solubility of phenolic compounds residues.

The encapsulation of olive leaf extracts in insulin was performed by Urzúa et al. (2017). Results pointed out that the total phenolic content of particles was considerably lower than the extract (around 21% of the initial phenolic content). The authors tested the incorporation of loaded microparticles in starch–gluten matrices. Like previously, essential features of encapsulation approaches were verified: the microencapsulation of the extract had a protective role in the degradation of phenolic compounds when atmospheric frying was applied. In the case of using the vacuum frying approach, the protective effect was not observed due to lower frying temperature.

In two complementary studies, Mohammadi et al. (2016), evaluated the particle size distribution alongside time when particles were produced by emulsification processes. In both studies, the authors concluded that the encapsulation of olive leaf extract in whey protein concentrate–pectin complex improved antioxidant capacity and allowed a sustained release of bioactive compounds.

In the early times of encapsulation progress, when the research of encapsulation and characterization techniques had started, Kosaraju, D'ath, and Lawrence (2006) efficiently

incorporated olive leaf extracts in chitosan microparticles. Even though the results were quite far from expected (encapsulation efficiency around 27%), in fact, these authors made use of some relevant characterization techniques in encapsulation processes as the Fourier Transform Infrared Spectroscopy (FT-IR) and the dynamic scanning calorimetry (DSC). Later, these authors encapsulated olive leaf extracts using sodium caseinate as the encapsulating material. They evaluated the retention of antioxidant activity of phenolic compounds after the encapsulation process, and they found the maximum radical scavenging activity around 60%. Therefore, they concluded that, is possible significant retention of antioxidant activity of the initial extract, after encapsulation procedures.

The antioxidant activity of olive leaf extract encapsulated was evaluated by Taghvaei et al. (2014), separately, in Arabic gum and maltodextrin. Moreover, particles storage stability was evaluated for 20 days. They verified that only a small amount of phenolic compounds were released during the 20 days at 55 °C. The encapsulation approach employed was able to protect the extract from decomposition during storage at 55 °C.

The comparison of the encapsulation of two different OOBP extracts (olive leaf extract and olive kernel extract) was performed by Chanioti, Siamandoura, and Constantina (2016). After the optimization of extraction conditions, they evaluated a vast number of distinguishing features of the obtained powder through microparticles characterization techniques. It can be concluded that even though the encapsulation efficiency of olive leaf extracts was slightly higher (around 1%), the antioxidant capacity of encapsulated olive kernel extract was surprisingly high.

The studies found regarding the encapsulation of OOP extracts are based on the optimization of operational encapsulation conditions. However, the results presented by Aliakbarian et al. (2017), revealed that the total phenolic content of OOP extract-loaded particles was higher using the supercritical assisted atomization in optimized conditions when compared to the results obtained by Paini et al. (2015) and Aliakbarian et al. (2018) when they encapsulated OOP extracts by spray-drying using maltodextrin in optimized conditions. In the study conducted by Paini et al. (2015), microcapsules were stable under storage conditions for 70 days at 5 °C in darkness. It was only verified a degradation of 21% after increasing the storage temperature up to 25 °C. The results from light exposure of microparticles revealed a 66% loss in phenolic compounds after 48 h.

Regarding the studies of the encapsulation of OOWW extracts, the results obtained by Petrotos et al. (2012) demonstrated once again the protective effect of encapsulated phenolic compounds: when they incorporated loaded particles in yogurt, they verified a protective effect against undesirable pH drop during yogurt storage.

The results presented in this literature review are outstanding significant as in all the studies, the authors revealed the adequate protection of phenolic compounds present in different OOBP as olive leaves, kernel, olive oil wastewater, and olive oil pomace. Encapsulation approaches allowed the adequate protection of extracts from oxidation, high temperatures, also demonstrating the protective effect of the phenolic compounds at atmospheric frying when leaf-extract loaded particles were incorporated in a starch matrix for frying applications.

Accordingly, new insights in the research areas of (i) OOBP management and disposal, in (ii) microencapsulation techniques and (iii) fortification of foods with phenolic-rich extract loaded microparticles may unlock innovative applications of OOBP in the future. Alternative scientific areas will benefit from the microencapsulation of phenolic compounds rich-extracts obtained from olive oil by-products. Firstly, it will aid the OOBP management and disposal. Afterward, the food industry may beneficiate from the extraction and incorporation of OOBP extracts in particles for the obtainment of added-value products, resistant of a wide range of environmental conditions and simultaneously allowing a delayed or sustained release of phenolic compounds that have been demonstrated positive health benefits, increasing food products shelf-life.

Identification and quantification of phenolic compounds

For the valorization from olive oil by-products is crucial the quantification of the total phenolic content of the obtained extracts from OOBP, the quantification of the amount of phenolic-rich extracts encapsulated and the amount of phenolic compounds released over time. Moreover, the quantification of the total amount of phenolic compounds released over time is paramount for the evaluation of phenolic compounds release pattern both in the fortified foods as well as during the gastrointestinal release. Simultaneously, it could be intended for the evaluation of the radical scavenging potential of extracts, loaded microparticles, and the phenolic compounds released.

Additionally, to the quantification of the total phenolic content and the total antioxidant capacity, it may be required the identification of specific phenolic compounds during the different instars of the valorization of phenolic compounds present in OOBP.

The evaluation of the total phenolic content is generally performed employing the Folin-Ciocalteu method. This spectrophotometer method is reported to be widely used for the estimation of the total amount of phenolic compounds present in extracts obtained from olive oil by-products (Cioffi et al. 2010). This colorimetric method is based on the electron transference between the Folin-Ciocalteu reagent and phenolic compounds. Alternatively, it can be defined as a method that detects calorimetrically a product obtained from the reduction of phosphotungstic-phosphomolibdic complex. Even though is easy to implement, this method presents several drawbacks as the possibility of the interference of other reducing agents as ascorbic acid, vitamin E, vitamin C, carotenoids, reducing sugars, and even certain aminoacids. Moreover, an additional limitation is the impossibility of the detection and quantification of individual compounds (Singleton and Lamuela-Raventós 1999).

For the quantification of radical scavenging activity/capacity of extracts obtained from OOBP, the encapsulated amount of antioxidants in polymeric shells and the amount of antioxidants released from microparticles over time can be accomplished determining the presence of alternative radicals as (i) the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical and (ii) the 2,2′-azinobis(3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS) radical. Standardized assays can also be used for the quantification of the radical scavenging activity as (i) the oxygen radical absorbance capacity (ORAC) assay, (ii) the superoxide dismutase (SOD) assay, (iii) the ferric reducing antioxidant potential (FRAP) assay, among others (Dudonné et al. 2009). The radical scavenging activity can be alternatively evaluated through the formation of Cu^{2+}-antioxidants complexes, as described by Jabeen et al. (2017).

For the identification and quantification of specific phenolic compounds present in OOBP, it is commonly applied a chromatographic technique as the reverse-phase (RP) high-performance liquid chromatography (HPLC). The detection of phenolic compounds using a RP-HPLC can be performed using two detectors–the diode array detector (DAD) and the fluorescence (FL) detector–that can be used individually or simultaneously. The detection and quantification of phenolic compounds extracted from OOBP are based on the ultraviolet absorption spectra in the case of using the DAD and on the maximum fluorescence excitation and emission wavelengths in the case of using the FL detector. Different compounds present alternative maximum absorption (in the case of the DAD) wavelength (λ_{max}) and differing maximum excitation and emission wavelengths in the case of using the FL detector (Alu'datt et al. 2010; El-Abbassi, Kiai, and Hafidi 2012). The compounds found in high amounts in olive oil by-products present alternative maximum absorption wavelengths (e.g. λ_{max} of hydroxytyrosol: 236 nm and 280 nm; λ_{max} of tyrosol: 234 nm and 275 nm; λ_{max} of oleuropein: 244 nm and 275 nm) (Klen 2014). Some compounds found on OOBP (e.g., tyrosol, hydroxytyrosol, vanillic acid, apigenin-7-O-glucoside) present a maximum fluorescence wavelength of excitation and emission at 280 nm and 330 nm, respectively (Obied et al., 2007). However, even though when both detectors are combined, the identification of complex or unknown phenolic compounds can be a challenging task.

Additionally, when occurs, the co-elution of compounds that present similar spectra and/or are verified low limits of detection, and quantification of phenolic compounds is compromised (Suárez et al. 2009). In these cases, is widely recommended to use a mass spectrometer (MS), which allows a specific analysis based on the molecular ion and fragmentation patterns for the identification of dubious compounds. The confirmation of the presence of specific phenolic compounds in OOBP can be performed associating an electrospray ionization (ESI) interface with liquid chromatography (LC)-MS through the fragmentation of the molecular ion and the identification of the respective molecular mass (De Marco et al. 2007). The authors Cardoso et al. (2011) identified seven new isomers of oleuropein and ligstroside derivates using ESI-MS and ESI-MS^n.

Other alternatives have been exploited to confirm the presence of specific phenolic compounds or identify them as the time-of-flight mass spectrometry (TOF-MS), which allows the determination of the m/z through time measurement. The authors Lozano-Sánchez et al. (2014) identified several phenolic compounds and their related derivates present in OOP as hydroxytyrosol, oxidized hydroxytyrosol, decarboxymethyl-oleuropein aglycone, decarboxymethyl-ligstroside aglycone, luteolin, and 10-hydroxy-oleuropein aglycone. It is also described the use of rapid resolution liquid chromatography (RRLC) and ultra-performance liquid chromatography (UPLC) associated with MS (Herrero et al. 2011; Lozano-Sánchez et al. 2011). The reduction of the particle size in the column results in the increase of the linear velocity and the peak capacity–the number of separated peaks per unit of time–also increases. The RRLC and the UPLC systems use smaller particles ($<2\,\mu$m) than the typical HPLC system (particle size $\geq 3\,\mu$m), which contributes to an increase of sensibility and the speed of samples running. The authors Suárez et al. (2009) compared the chromatographic separation of phenolic compounds extracted from olive cake by HPLC and UPLC. They successfully separated phenolic compounds in 24 minutes using the UPLC and in 60 minutes, using the HPLC apparatus.

From this perspective, during the choice of the analytical method to employ should be considered which are the main purposes of the study (if it is to identify specific compounds increasing the complexity of the process; if it is only necessary to quantify the total amount of phenolic compounds), the availability of analytical apparatus, the feasibility of the study, the time of analysis, the consumption of solvents among others.

A combination of chromatographic and spectrophotometric quantification methods should be carefully considered as both methods' present limitations.

Perspectives and future challenges

Olive oil has been produced since ancient times; however, the awareness for the problematic of olive oil by-products management and disposal is considerably recent.

In the regard of the conscious awareness of the negative environmental impact of phenolic compounds present in olive oil by-products (olive oil pomace, olive leaves, olive oil wastewater and olive kernel) combined with the finding of innovating alternative valorization strategies for extraction and encapsulation approaches, arise a potential pathway for the obtainment of added-value food products fortified with encapsulated phenolic-rich extracts.

It is expected that encapsulation approaches associated with extraction techniques of phenolic compounds with solvent generally recognized as safe will contribute positively to the environmental problem as well as improve or redesign added value products for the food industry.

In the best expectations, research will be focused on the combination of extraction and encapsulation techniques for

the recovery of phenolic compounds from olive oil by-products, addressing the environmental problem associated to the presence of phenolic compounds in olive oil by-products, answering the market demand of innovative products. Moreover, it is expected that comprehensive studies should be conducted in the scope of the fortification of foods with phenolic-rich extracts loaded microparticles. An integrated approach regarding the fortification of foods with loaded microparticles and in vivo absorption, transportation, and release of phenolic compounds enclosed in emulsions should be considered and evaluated.

Conclusions

The olive oil is one of the most consumed edible oils around the world, mostly due to the success of the Mediterranean diet in which olive oil intake is associated to many health benefits as some compounds present in olive oil may prevent cardiovascular diseases, may lower the blood pressure, may have a positive effect on inflammation processes and in endothelial dysfunction.

The worldwide recognition of the importance of the consumption of olive oil fostered its production intensively. Olive oil can be extracted from olive drupes through continuous and discontinuous processes. During olive oil production, different olive oil by-products are generated as olive leaves, olive oil wastewaters, olive oil pomace, and olive stones/kernel.

These olive oil by-products constitute, nowadays, an environmental problem regarding its disposal and management. Moreover, the majority of phenolic compounds remain in the olive oil by-products (around 98%) due to their higher affinity for aqueous systems rather than oily phases. Therefore, another problem arises: the high content of phenolic compounds in olive oil by-products is responsible for the phytotoxic and biotoxic of these by-products.

One of a potentially innovative strategy to valorize olive oil by-products is to recovery phenolic compounds which have many health benefits from these by-products through the obtainment of extracts that can be encapsulated. Encapsulation strategies may protect the extracted phenolic compounds from olive oil by-products for the external environment, improve extracts antioxidant capacity and stability to mid to long term storage, to high temperatures, oxidation conditions, and undesired pH drops, allowing a delayed or sustained release of these valuable compounds.

Olive oil by-products extracts encapsulated in polymeric matrices for further incorporation in complex matrices as foods, is expected to aid the olive oil sector regarding the problematic of the disposal and management of olive oil by-products and also aid food industry to reinvent through the obtainment of added-value products as fortified foods with phenolic-rich extract loaded microparticles that may exhibit outstanding radical scavenging activities.

Disclosure statement

Authors declare no conflict of interest or any financial benefit from direct or indirect applications of the research.

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