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Review

A review on the use of membrane technology and fouling control for olive mill wastewater treatment

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HIGHLIGHTS

- Olive mill wastewater can be effectively treated through of membrane technology.
- Different integrated membrane treatment processes are summarized and discussed.
- Appropriate fouling inhibition methods should be set upstream membrane operation.
- Feasible energetic and added-value compounds valorization of concentrate streams.
- Final treated effluent compatible for reuse in irrigation can be achieved.

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ABSTRACT

Olive mill effluents (OME) by-produced have significantly increased in the last decades as a result of the boost of the olive oil agro-industrial sector and due to the conversion into continuous operation centrifugation technologies. In these effluents, the presence of phytotoxic recalcitrant pollutants makes them resistant to biological degradation and thus inhibits the efficiency of biological and conventional processes. Many reclamation treatments as well as integrated processes for OME have already been proposed and developed but not led to completely satisfactory and cost-effective results. Olive oil industries in its current status, typically small mills dispersed, cannot afford such high treatment costs. Furthermore, conventional treatments are not able to abate the significant dissolved monovalent and divalent ions concentration present in OME. Within this framework, membrane technology offers high efficiency and moderate investment and maintenance expenses. Wastewater treatment by membrane technologies is growing in the recent years. This trend is owed to the fact of the availability of new membrane materials, membrane designs, membrane module concepts and general know-how, which have promoted credibility among investors. However, fouling reduces the membrane performances in time and leads to premature substitution of the membrane modules, and this is a problem of cost efficiency since wastewater treatment must imply low operating costs. Appropriate fouling inhibition methods should assure this result, thus making membrane processes for wastewater stream treatment both technically and economically feasible. In this paper, the treatment of the effluents by-produced in olive mills, generally called olive mill wastewaters, will be addressed. Within this context, the state of the art of the different pretreatments and integral membrane processes proposed up to today will be gathered and discussed, with an insight in the problem of fouling.

1. Introduction

In the last decades, the effluents generated by olive oil industries, commonly known as olive mills, have significantly increased as a result of the boost of the olive oil agro-industrial sector, also due to the technological conversion into continuous operation centrifugation processes. The olive oil sector has represented since several decades one of the most important industries in the Countries of the Mediterranean River Basin. In Spain, the main olive oil producer with more than 45% of the total olive oil production worldwide, there are more than 1700 olive mills currently authorized and operating. More than 1,400,000 tons of olive oil were produced in Spain during the 2013–2014 campaign, 70% of which were obtained in Andalucía where there are 850 olive mills, which yielded a production of 1,022,000 tons of olive oil as well as 4,778,451 tons of table olives, highlighting the provinces of Jaén, with more than 515,000 tons of olive oil and 2,229,000 tons of olives, Granada with 118,000 tons of olive oil and 509,383 tons of olives, and Córdoba with 219,000 tons of olive oil and 1,123,800 tons of olives.

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The production of olive oil employs a very significant number of people and is one of the main industrial activities, as in other countries of the Mediterranean Basin: Italy, Portugal, Greece and the Northern African countries — Syria, Algeria, Turkey, Morocco, Tunisia, Libya, Lebanon, Egypt (Fig. 1). Other countries such as France, Serbia and Montenegro, Macedonia, Cyprus, Turkey, Israel and Jordan also produce a considerable annual olive oil yield (International Olive Oil Council, IOOC, 2013–2014). Moreover, olive oil production is also rapidly becoming an emergent agro-food industry in China and other countries such as the USA, Australia and the Middle East. It is very worth highlighting the case of China, which exhibits favorable edaphoclimatic conditions for the growth of the olive trees, and is expected to develop a considerable olive oil production potential in the near future. Hence, the treatment of the olive mill effluents (OME) is already a task of global concern and is not anymore a problem constrained to a specific region.

The technological change of olive oil production, with the introduction of continuous centrifugation processes in replacement of the antique batch press methods, has ensured higher productivity, but have led to a significant increment of the effluents by-produced in these industries (OME) as well. Currently, an average-sized modern olive oil mill generates between 10 and 15 m^3 daily of wastewater derived from the centrifugation process, called olive mill wastewater (OMW), together with 1 m^3 of olive washing wastewater (OWW) per ton of processed olives. This raises several millions cubic meters of OME each year.

Whereas OWW is commonly composed of high concentration of suspended solids (mainly peel, pulp, ground, branches and leaves debris) derived from the washing procedure of the olive fruit, but low concentration of dissolved organic matter – depending on the water flow exchange rate in the washing machines during the fruit cleaning procedure – usually below the standard limits for discharge on superficial suitable terrains; on the other hand, discharge of untreated OMW to the ground fields and superficial waters bodies is currently prohibited in Spain, whereas in Italy and Portugal as well as in other European countries only partial discharge on suitable terrains is allowed. Straight discharge of OMW has been reported by several authors to cause strong odor nuisance, soil contamination, plants growth inhibition, underground leaks, water body pollution and hindrance of self-purification processes, as well as severe impacts to the aquatic fauna and to the ecological status, due to the presence of bio-refractory contaminants, including a wide variety of phenolic compounds, tannins, fatty acids and organohalogenated pollutants (Danellakis et al., 2011; Hodaifa et al., 2008, 2013a; Karaouzas et al., 2011; Martínez Nieto et al., 2011a; Ntougias et al., 2013). Due to the presence of high levels of refractory organic compounds, direct disposal of OMW to the municipal sewage treatment plants is also prohibited.

In this scenario, the European Directive 2000/60/CE took the lead in establishing the legal framework to confer utmost protection to water, highlighting the use of regenerated wastewater. Moreover, European Environmental Regulations will become more stringent in virtue of the '*H2020 Horizon*' (http://ec.europa.eu/programmes/horizon2020/). Water scarcity specially concerns agricultural irrigation, which demands more than 70% of the total water consumption worldwide (Food and Agricultural Organization (FAO), 2003). However, there is a big potential to use regenerated wastewater for irrigation purposes, which would result in a solution with very positive environmental and economic impacts.

Geographical dispersion of olive oil mills, as well as their small size and the seasonality of olive oil production, are handicaps that have typically made the management of OME quite cost-ineffective. Furthermore, the physico-chemical composition of OME is extremely variable as it depends on several factors such as the type of olive oil extraction process, the edaphoclimatic conditions of the region and cultivation parameters, as well as the type, quality and maturity of the kind of processed olives. OME typically exhibit intense violet-dark color, acid pH, strong odor, considerable saline toxicity reflected by high electroconductivity values, and very heavy organic pollutants load (Table 1).

In the continuous two-phase extraction process water injection is only performed in the final vertical centrifugation step, therefore the volume of liquid effluent derived from the production process (OMW-2) is reduced by one fifth on average if compared to the amount required for the three-phase system (Table 2). On the other hand, much of the organic matter remains in the solid waste, which contains more moisture than the pomace from the three-phase system (60_70% in two-phase systems vs. 30_45% in three-phase ones, OMW-3) and hence OMW-2 exhibits lower pollutants degree, too: the measured chemical oxygen demand (COD) in OMW-2 is commonly in the range 4–16 g/L in contrast with up to 30–200 g/L for OMW-3. Inorganic compounds including chloride, sulfate and phosphoric salts of potassium, calcium, iron, magnesium, sodium, copper and traces of other elements are also common traits of OMW (Niaounakis and Halvadakis, 2006; Paraskeva and Diamadopoulos, 2006).

A wide variety of stand-alone and integrated processes for the treatment of OMW have already been proposed and developed but have not yet led to completely satisfactory results, such as lagooning or natural evaporation and thermal concentration (Annesini and Gironi, 1991; Paraskeva and Diamadopoulos, 2006), composting (Bouranis et al., 1995; Cegarra et al., 1996; Papadimitriou et al., 1997), treatments with clay (Al-Malah et al., 2000) or with lime (Aktas et al., 2001), physico-chemical procedures including coagulation–flocculation (Martínez Nieto et al., 2011b; Sarika et al., 2005), electrocoagulation (Inan et al., 2004; Tezcan Ün et al., 2006) and biosorption (Hodaifa et al., 2013a; Martínez Nieto et al., 2010; Stasinakis et al., 2008), advanced oxidation processes comprising ozonation (Cañizares et al., 2006, 2007 and Cañizares et al., 2009), Fenton's reaction (Hodaifa et al., 2013b) and photocatalysis (Chatzisymeon et al., 2009; Sacco et al., 2012), electrochemical treatments (Inan et al., 2004; Papastefanakis et al., 2010, Tezcan Ün et al., 2008) and hybrid processes (Ammary, 2005; Beltran-Heredia et al., 2001; Grafias et al., 2010; Khoufi et al., 2006; Lafi et al., 2009; Rizzo et al., 2008).

Various authors have noticed that common biological treatment processes – active sludge – are not effective for the treatment of OMW

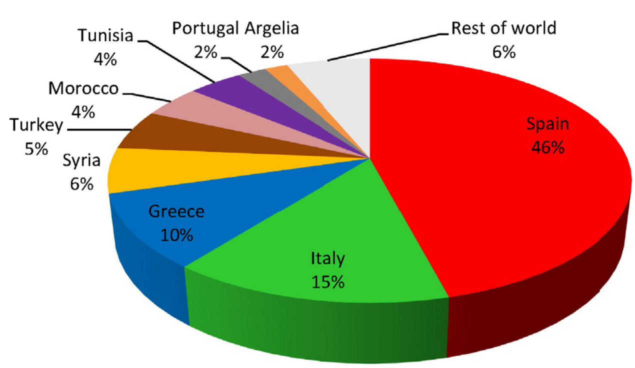

Fig. 1. Olive oil production worldwide (International Olive Oil Council, IOOC 2014).

Table 1
Characteristics of the effluents of batch and continuous (three-phase and two-phase) olive oil extraction processes.

Process	Effluent	COD, g/L	BOD_5, g/L	TSSs, g/L	pH	EC, mS/cm	TPh, g/L
Olives cleaning	OWW	0.8–2.2	0.3–1.5	8–18	5.5–6.6	2.5–3.0	0–0.1
Batch press	OMW-P	130–130	90–100	10–12	4.5–5.0	2.0–5.0	1.0–2.4
Three phase	OMW-3	30–200	5–45	5–35	3.5–5.5	2.0–7.9	0.3–7.5
Two phase	OMW-2	4–16	0.8–6.0	2–7	3.5–6.0	1.5–2.5	0.1–1.0

COD: chemical oxygen demand; BOD_5: biological oxygen demand; TSS: total suspended solids; EC: electric conductivity; TPh: total phenols; OWW: olive washing wastewater; OMW-P: press OMW; OMW-3: three-phase OMW; OMW-2: two-phase OMW.

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