Science of the Total Environment 612 (2018) 758–766

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Operation setup of a nanofiltration membrane unit for purification of two-phase olives and olive oil washing wastewaters

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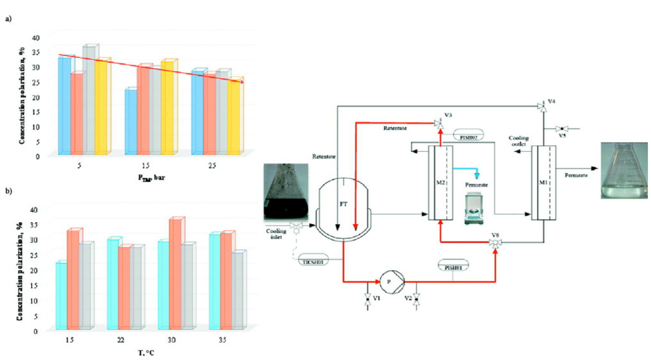
HIGHLIGHTS

- Nanofiltration (NF) process operating conditions insight and output was examined.
- EC was lowered to quality values acceptable for irrigation (1.9–2.0 mS cm^{-1}).
- The performance of the NF membrane ranged between 2.82 and 6.96 L h^{-1} m^{-2} bar^{-1}.
- The necessary overdesign of the membrane operation was estimated as 9.42–17.53%.
- Standards to reuse the purified effluent for irrigation or sewers discharge ensured.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 April 2017
Received in revised form 28 August 2017
Accepted 29 August 2017
Available online xxxx

Editor: D. Barcelo

Keywords:
Olive mill wastewater
Nanofiltration
Concentration polarization
Wastewater reclamation
Wastewater reuse

ABSTRACT

In this research work, the purification of olives and olive oil washing wastewaters from two-phase extraction mills by a novel polymeric NF membrane is addressed. The effluent was previously subjected to a physicochemical secondary-tertiary treatment previously optimized at pilot and industrial scales. Within the adequate operating conditions, suspended solids could be completely removed, and the EC was considerable lowered down to good quality values acceptable for irrigation purposes (1.9–2.0 mS cm^{-1}), whereas the chemical oxygen demand was reduced below 31.9 mg L^{-1}. The standards for discharging in public waterways or reusing the final treated effluent for irrigation with acceptable quality were therefore accomplished. Moreover, the performance of the NF membrane ranged between 2.82 and 6.96 L h^{-1} m^{-2} bar^{-1}, that is, a flux of up to 160 L h^{-1} m^{-2} at 25 bar. Furthermore, the 15-minute acid cleaning plus 15-minute alkaline/detergent cleaning could recover satisfactorily the permeability of the membrane. The necessary overdesign of the membrane operation was estimated as 9.42–17.53%, which meant a maximum required membrane area of 61.82 m^2. Hence, just 2 membrane modules should be implemented in a medium-sized mill to engineer the operation, boosting the economic feasibility of the proposed process both from operational and capital costs point of views.

1. Introduction

Olive mill wastewater (OMW) is a very heavily-polluted agro-industrial effluent (Niaounakis and Halvadakis, 2006; Paraskeva and Diamadopoulos, 2006). These wastewaters are generated during the production process of olive oil, mainly resulting from the washing of the fruits and from the centrifugation processes. Although historically based in the Mediterranean region - Spain, Italy, Portugal, Greece and Northern African countries - OMW it is now a task of global concern, as these industries are currently implemented or emergent in many other countries: France, Serbia and Montenegro, Macedonia, Cyprus,

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http://dx.doi.org/10.1016/j.scitotenv.2017.08.287

Turkey, Israel, Jordan, Iran, Iraq, and in the USA, Australia, and China (International Olive Oil Council, 2015).

These agro-industrial effluents present acid pH because of the content of organic acids in their composition, intense violet-dark color, considerable saline concentration, which confers them high electroconductivity (EC) and thus saline toxicity, as well as a strong odor (Hodaifa et al., 2013a; Niaounakis and Halvadakis, 2006; Stoller, 2011). The concentration in recalcitrant compounds, mainly phenolic compounds, tannins and organohalogenated pollutants, makes OMW utterly difficult to be degraded by conventional processes such as biological ones (Paraskeva and Diamadopoulos, 2006). As a result, the discharge of these effluents is controlled in most producing countries. The direct disposal of OMW to the ground fields has been reported to cause contamination of the soil and phytotoxic effects to the flora and fauna, and inhibition of auto-purification processes if they are directly discharged into water bodies. Also, if they leak to the underground they cause deleterious pollution of aquifers (Hodaifa et al., 2013a; Niaounakis and Halvadakis, 2006; Paraskeva and Diamadopoulos, 2006).

Legal actions have been undertaken in the last years in the producing countries in response to this. The straight disposal of untreated OMW to the ground fields and superficial waters bodies is nowadays prohibited in Spain, Italy, Portugal and other European countries, whereas in some others where policies are not so developed, just the partial and temporary-spatial discharge onto certain terrains is allowed. In addition to this, the direct discharge of OMWs to the municipal sewage collectors is prohibited too, given the high concentrations of refractory organic pollutants. Legal limits are established to prevent inhibition of the biological treatment processes that take place in wastewater treatment plants. In this direction, the European Union is currently committed through different financial programs with the regeneration of the used resources (https://ec.europa.eu/programmes/horizon2020), and this implies the reclamation and reuse of wastewaters from very diverse sources.

One important breakthrough in olive oil production was the technological change of the first continuous three-phase centrifuges for more modern two-phase ones, although the three-phase centrifugation system is still surviving in many countries due to the scarcity of financial support needed for the technological upgrade (Mendoza et al., 1996). These new centrifuges lead to the by-production of lower volumes of liquid effluents, since water injection is not needed or at least minimized (Mendoza et al., 1996). However, very high volumes of OMW are still produced, presenting very high organic matter concentration, hence the problem is not solved yet.

A wide variety of treatment processes have been studied by researchers all over the world for the reclamation of these effluents (Ioannou-Ttofa et al., 2017), highlighting lagooning or natural evaporation and thermal concentration (Annesini and Gironi, 1991; Paraskeva and Diamadopoulos, 2006), composting (Cegarra et al., 1996; Papadimitriou et al., 1997), treatments with clay (Al-Malah et al., 2000) and with lime (Aktas et al., 2001), physico-chemical processes including coagulation-flocculation (Martínez Nieto et al., 2011; Sarika et al., 2005), electrocoagulation (Inan et al., 2004; Tezcan Ün et al., 2008) and biosorption (Hodaifa et al., 2013b; Martínez Nieto et al., 2010), advanced oxidation processes comprising ozonation (Cañizares et al., 2007, 2009), Fenton's reaction (Hodaifa et al., 2013a; Ochando-Pulido et al., 2012), photocatalysis (Chatzisymeon et al., 2009; Ochando-Pulido et al., 2015a), electrochemical treatments (Papastefanakis et al., 2010; Tezcan Ün et al., 2008) and combined processes (Ammary, 2005; Khoufi et al., 2006; Rizzo et al., 2008).

Other major handicaps faced in relation to the management of these effluents rely on the small size of these factories, which are also dispersed geographically. These two aspects certainly limit the possibility of success of the proposals. Most processes presented up to the date for the treatment of OMW are rather cost-ineffective, and the olive oil industry in its current status cannot assume such high costs. Among the proposed treatment processes, chemical remediation strategies based on advanced oxidation processes (AOPs) seem a feasible solution. Particularly, Fenton is the most cost-effective, on the premises that it does not require nor high energetic costs (it can be operated efficiently at ambient temperature and pressure) nor high capital investment (Cañizares et al., 2009; Hodaifa et al., 2013b).

On another hand, membrane processes have also been examined for the purification of these effluents. Membrane technologies imply low to moderate costs in contrast with energy-intensive conventional separation processes, and are easily scalable too (Field and Pearce, 2011). Moreover, depending on the membrane type and material, high rejection and selectivity - necessary for the purification of target species in wastewater treatments - can be achieved (Stoller and Ochando Pulido, 2015). Owed to these key features, and because of the improvement in terms of membrane know-how, membranes have been gradually implemented in water and wastewater treatment lines (Field and Pearce, 2011; Le-Clech et al., 2006; Ochando-Pulido et al., 2014). Concretely, the specific selectivity towards small pollutants and the lower energy consumption of nanofiltration (NF) membranes have triggered their use as tertiary treatment in integrated wastewater treatment processes (Beril Gönder et al., 2011; Boussu et al., 2007; Ellouze et al., 2012; Iaquinta and Stoller, 2009; Korzenowski et al., 2011; Luo et al., 2012; Ochando-Pulido, 2016; Paraskeva et al., 2007; Pizzichini et al., 2005; Stoller et al., 2013; Van der Bruggen et al., 2001; Wei et al., 2010).

Up to the moment, few works on OMW treatment with membranes have been published (Conidi et al., 2014; Coskun et al., 2010; Di Lecce et al., 2014; Paraskeva et al., 2007; Stoller, 2011; Zagklis et al., 2015; Zirehpour et al., 2012). What is more, the authors of the present paper have detected one common lack in most of the published literature on the topic, regarding the insight of the operating conditions of the performance and output of the membranes. The adequate set-up of the operating framework of a membrane is of paramount importance in terms of productivity and rejection efficiencies towards the different target species. Moreover, there is a knowledge gap in the available scientific literature which reports mainly membrane treatment processes for OMW of olive mills operating with the three-phase system, but very scarce is published on the modern two-phase technology that is prevalent in Spain and is now being implemented in Portugal, Greece and Italy, the main producers worldwide (Ochando-Pulido et al., 2012; Ochando-Pulido and Stoller, 2015).

In former work by the authors (Hodaifa et al., 2013a), OMW from two-phase mills was conducted to a treatment consisting in a core Fenton process for the abatement of the high organic matter load. In the present research study, the performance of a novel thin-film-composite (TFC) polymeric NF membrane for the ulterior purification of the effluent is fully examined in terms of productivity (permeate flux), rejection towards target species, membrane cleaning for the effective recovery of its permeability, and finally the required membrane area of the plant. The set-up of the adequate operating framework of a specific feedstream-membrane system determines in fact the feasibility and cost-effectiveness of the process at industrial scale. Engineers design membrane processes with an excessive oversized capacity, up to >300%, triggering both the investment and operating costs unnecessarily (Stoller and Ochando Pulido, 2015). In other words, the lack of knowledge of the process represents an additional cost for this industry that must be minimized to permit its successful competitiveness.

Finally, the standards to reuse the purified effluent for irrigation or recycling as irrigation water were checked, with the aim of closing the loop to reduce the environmental impact of the olive oil production process.

2. Experimental

2.1. Olive mill wastewater (OMW) samples

The samples of OMW were taken from various olive mills located in the region of Andalucía (Spain), specifically in the provinces of Granada