Journal of Environmental Management 145 (2014) 341–348

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Olive mill wastewater treatment using a simple zeolite-based low-cost method

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A R T I C L E I N F O

Article history:
Received 11 April 2014
Received in revised form
8 July 2014
Accepted 9 July 2014
Available online

Keywords:
Olive mill wastewater
Treatment
Successive columns
Zeolite

A B S T R A C T

Olive mill wastewater (OMW), a liquid by-product of the olive oil industry, represents a severe environmental problem owing to its high pollution load. In this study, successive columns containing different types of natural materials were investigated for their OMW treatment efficiency. Passing OMW through three columns of gravel, fine sand, and a mixture of acidified cotton and zeolite (weight:weight ratio of cotton:clinoptilolite of 2:1), followed by treatment with activated charcoal (AC) and lime, was the best treatment in terms of the quality of water obtained. This treatment decreased concentrations of NO_3^-, B, K, P, and total fat in OMW by mean percentages of 78.0, 92.4, 66.6, 48.3, and 93.3%, respectively. Furthermore, it decreased OMW turbidity and electric conductivity (EC) by 96.8 and 48.4%, respectively. Most contaminants were removed from the OMW in the cotton/clinoptilolite column owing to the high sorption affinity of clinoptilolite on its active sites. The AC was efficient for organic particle removal; meanwhile, lime was used to raise the pH of the treated OMW (TOMW) from 2.9 to 5.1. This simple method enables us to obtain environmentally friendly TOMW that can be safely used for irrigation.

1. Introduction

Environmental pollution by toxins has accelerated dramatically in recent years due to increasing industrialization (Tiwari et al., 2008). Mankind's demand for resources and raw material treatments has intensified the ecological and economic contradictions in the industrial sector (Sen and Chakrabati, 2009). Widespread industrial development in urban areas has radically reduced land areas for waste disposal. Disposal of untreated industrial and domestic wastes into the environment affects the quality of the soil and groundwater and makes this soil and groundwater undesirable for use (Quazilbash et al., 2006). Water produced by industry generally contains potential hazardous wastes, such as heavy metals and toxic organic compounds, and carries these hazardous wastes into our environment (Azumi and Bichi, 2010). In this context, the waters produced by the olive oil industry usually contain hazardous materials that cause severe environmental problems (Mekki et al., 2007).

1.1. *Olive mill wastewater production*

Olive processing has been important in Mediterranean countries for centuries. Moreover, the growing interest in the consumption of olive oil as an essential of the Mediterranean diet has increased the importance of the olive oil sector in recent decades. Recently, the Kingdom of Saudi Arabia (KSA) started heavy olive oil production in the northern part of the Tabuk and Al-Jouf regions. According to the International Olive Oil Council (IOOC), worldwide production of olive oil was approximately 3,024,000 t during 2009/2010, and the European Union (EU) produced 74% of this total (Cara et al., 2012). Olive oil production involves producing considerable amounts of liquid effluent, which is referred to as olive mill wastewater (OMW). The OMW amounts to 0.5–3.25 m^3 per 1000 kg of olives, depending on the process used (Paraskeva and Diamadopoulos, 2006; Kapellakis et al., 2012). Furthermore, Mekki et al. (2013) recorded that the annual production of OMW in Mediterranean countries reached 30 million m^3 in the years 2005/2006. In the KSA alone, the annual olive oil production has been reported to be 80,000 t; consequently, the calculated OMW production could be around 160,000 m^3.

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http://dx.doi.org/10.1016/j.jenvman.2014.07.012
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1.2. Olive mill wastewater characteristics

Typically, OMW is composed of the water from the tissue of the fruit, the water used for the various stages of oil production, olive pulp, mucilage, pectin, and oil, among other things, suspended in a relatively stable emulsion. Moreover, it contains toxic organic compounds and inorganic compounds (Mekki et al., 2009, 2013). In general, OMW is characterized by an intensive violet–dark brown up to black color, strong specific olive oil smell, high degree of organic pollution (chemical oxygen demand (COD) of 40–220 g/L and biochemical oxygen demand (BOD) of 35–110 g/L), pH of 3–6, 25–45 g/L of organic compounds in total, high electrical conductivity (EC), high content of polyphenols (0.5–2 4 g/L), reduced sugars ($\leq$60% of the dry weight), and high solid matter content. Potassium is the predominant inorganic material (~4 g/L) in OMW (Niaounakis and Halvadakis, 2006). Typically, OMW is an acidic effluent with a high nutrient content that can be used to fertilize soil; however, it is very rich in toxic phenolic compounds (Aggelis et al., 2003). Additionally, the C/N ratio of OMW is unfavorable for biodegradation processes (Mekki et al., 2006a, 2013). Using OMW in irrigation without treatment increases soil salinity, which results from the presence of the main ionic species (K^+, Na^+, and HCO_3^-) of the OMW (Zenjari and Nejmeddine, 2001). In recent years, there has been increased attention directed toward finding the best methods to treat OMW and toward recycling both the organic matter (OM) and nutritive elements in the crop production system. Some OMW characteristics are favorable for agriculture, because this effluent is rich in water, OM, N, P, K, and Ca; however, other characteristics are unfavorable for agriculture, including the presence of phenolic compounds (Lesage-Meessen et al., 2001; Komilis et al., 2005; Mekki et al., 2006b).

1.3. Olive mill wastewater treatment

Subsequently, Kapellakis et al. (2012) reported the widespread use of evaporation ponds to manage OMW, which can be attributed to the low cost of this method. However, evaporation ponds are associated with odor development, leaking of OMW to surface waterways or groundwater, and relatively high area requirements in regions with low evaporation rates. Many different treatment methods of OMW have been proposed previously, including aerobic treatment, anaerobic digestion, and composting (Sayadi and Ellouz, 1995; Ehaliotis et al., 1999; Kissi et al., 2001; Marques, 2001). However, an environmentally safe and cost-effective solution to OMW treatment has not yet been found (Zagklis et al., 2013). Most studies devoted to building efficient treatment technologies for OMW are not economically feasible owing to the short olive oil season, typically biennial olive harvest cycle, and the small isolated area of olive mills (Zenjari and Nejmeddine, 2001).

The main objectives of this study were as follows: to evaluate the physiochemical characteristics of the KSA's olive mill wastewater; to investigate a suitable and feasible method for olive wastewater treatment that will decrease toxic compound concentrations; and to assess the suitability of treated water for irrigation purposes.

2. Materials and methods

2.1. Collection and preservation of olive mill wastewater

Five hundred liters of fresh OMW was collected during the 4th quarter of 2013 from the National Agricultural Development Company (NADEC) Al-Jouf Project, which is located in the Al-Jouf region about 1250 km north east of Riyadh City, KSA. At this site, olive oil is produced by the centrifugal method. The OMW samples were collected in plastic containers (20 L) and transported immediately to a laboratory at King Saud University and stored at approximately 8 °C.

2.2. Analytical methods

OMW chemical analyses were carried out to assess the quality of water according to procedures described by Matiti (2004), and then different types of water treatments were examined. Water reaction (pH) was determined using a pH meter (pH meter, CG 817). Total soluble salts were measured by using an EC meter in units of dS m^{-1} at 25 °C (Test kit Model 1500_20 Cole and Parmer). Bicarbonate (HCO_3^-) concentration was determined by titration with sulfuric acid (H_2SO_4), while Cl concentration was determined by titration with silver nitrate ($AgNO_3$) (Matiti, 2004). Sulfate (SO_4^{-2}) concentration was determined by the turbidity method (Tabatabai, 1996), and the nitrate (NO_3^-) concentration was determined by the phenoldisulfonic acid method (APHA, 1998). Soluble Na and K concentrations were determined using a flame photometer (Corning 400). Phosphate concentration was determined using the method described by Matiti (2004). Boron was determined using the azomethine-H method (Keren, 1996). Concentrations of Ca, Mg, Cd, Co, Cu, Fe, Mn, Ni, Pb, and Zn were determined using inductively coupled plasma (ICP) spectrometry (Perkin Elmer Model 4300DV). UOMW was digested by nitric acid (HNO_3) (Matiti, 2004) and then injected into the ICP. TOMW was injected directly into the ICP after acidification by nitric acid and without digestion.

2.3. Treatments of olive mill wastewater

2.3.1. Treatment of olive mill wastewater by the sedimentation and flotation method (S and F)

Sedimentation and flotation (S and F) is a solid–liquid separation technique using gravity settling to remove suspended solids having densities higher or lower, respectively, than water (Kapellakis et al., 2012). The removal of suspended solids from wastewater by gravity separation is one of the most widely used unit operations in wastewater treatment (Al-Farraj et al., 2013). In this study, physical separation was carried out using glass cylinders with diameter of 20 cm and length of 24 cm. After 6 h of gravity separation, two kinds of suspended solid were observed: a floating part (containing total fat content of 9.39%) and a precipitated part (containing total fat content of 1.64%).

2.3.2. Treatment of olive mill wastewater using successive steps

After OMW physical separation, the water was extracted using a siphon method and then passed through six different designed treatment methods, with each treatment consisting of 3–4 successive columns containing different natural materials (Table 1).

The columns used in this experiment were made of transparent polyvinyl chloride (PVC) and had an internal diameter of 10 cm and a length of 40 cm. The columns were sealed from the bottom using two pieces of gauze fabric firmly held by strings and tape. The columns were packed with the material to 25 cm height. The bulk densities of the sand columns were 1500 kg m^{-3}. The packing process was conducted in 10 cm increments to avoid segregation of particles. The columns were mounted vertically on a wooden holder inside the laboratory and kept at a controlled temperature of 22 ± 2 °C. Each treatment system was replicated three times. Cumulative TOMW versus time were collected for analysis. The contact times between the OMW and substrates were 1 min for gravel, 10 min for fine sand, and 0.5 min for the cotton:clinoptilolite column. The grass and unactivated charcoal columns recorded contact times of just a few seconds. Five physiochemical parameters were