### **ARTICLE IN PRESS**

#### Waste Management xxx (2016) xxx-xxx

Contents lists available at ScienceDirect



Waste Management



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

## Incorporation of an anaerobic digestion step in a multistage treatment system for sanitary landfill leachate

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#### ARTICLE INFO

Article history: Received 16 December 2015 Revised 27 April 2016 Accepted 27 April 2016 Available online xxxx

Keywords: Anaerobic digestion Landfill leachate Chemical precipitation Microfiltration Reverse osmosis GC-MS

#### ABSTRACT

A combined process of anaerobic digestion (AD), lime precipitation (P), microfiltration (MF) and reverse osmosis (RO) was developed for the treatment of landfill leachate (LFL). The raw LFL contained high amount of organic matter with an elevated humic acids concentration. During the anaerobic digestion step, the organic loading rate was increased progressively up to 3.3 g COD  $L^{-1} d^{-1}$ . The upflow anaerobic fixed bed reactor showed a great performance in terms of COD removal efficiency and biogas production. During precipitation experiments, lime dose was optimized to obtain the maximum reduction of conductivity to prevent the fouling of RO membranes. This process was compared to a second one in which the AD step was eliminated. Both treatment plans achieved similar removal efficiencies. However, AD step significantly improved the process by reducing the needed lime dose by 50%. It has also increased MF and RO fluxes by 35% and 40% at a steady state, respectively. The dominant fouling mechanism was cake layer formation during both MF tests. This process seems to be a promising approach for the treatment of LFL and its industrial application should be further investigated.

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#### 1. Introduction

Landfilling is one of the most used methods for municipal solid waste disposal around the world (Dong et al., 2014). However, the generation of the landfill leachate (LFL), a dark effluent of remarkably variable composition with recalcitrant compounds (Müller et al., 2015), is a major pressing issue surrounding the operation of sanitary landfills (Dong et al., 2014; Amor et al., 2015). It may cause severe pollution to the groundwater aquifers as well as to the adjacent surface waters (Huang et al., 2014). The development of an efficient LFL treatment system plays an important role in preventing such potential threats. Many treatment technologies such as conventional chemical, physical and biological processes are in practice for the treatment of leachate (Insel et al., 2013; Fernandes et al., 2015; Hassan et al., 2016).

Although the discharge standards for rejection of the treated leachate regarding a wide range of inorganic and organic trace elements are not clearly defined in Tunisia, these substances have a negative effect on the environment and may lead to bioaccumulation in the ecological system. Reverse osmosis (RO) has been one of the most widely used methods for leachate treatment due to its

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http://dx.doi.org/10.1016/j.wasman.2016.04.030 0956-053X/© 2016 Elsevier Ltd. All rights reserved. ability to retain both organic and inorganic contaminants (Bohdziewicz et al., 2008). Due to the low filterability of leachate compared to other wastewaters (Insel et al., 2013), the performance of such treatment process would be mainly limited by the membrane fouling and the high osmotic pressures involved (Renou et al., 2008). Membrane fouling is an important issue in the treatment process as it causes a major reduction in flux and shortens the membrane life and then increases the capital and operational costs of the treatment plant (Hasar et al., 2009). To reduce membrane fouling, Renou et al. (2008, 2009) have employed lime precipitation subsequent to RO process. The optimum lime dose was adjusted to obtain the maximum reduction of conductivity. Such pretreatment has led to the preferential removal of calcium, carbonate ions, organic compounds including humic acids, and thus reduced the risk of irreversible membrane fouling. Still the combination of physico-chemical and biological methods is required for the efficient treatment of leachate. Furthermore, many researchers have stated that biological technology was one of the most cost-effective methods to treat landfill leachate, especially for young leachate with a high biodegradability (Canziani et al., 2006; Castillo et al., 2007; Yang and Zhou, 2008).

With the above aforementioned, this study investigated the incorporation of an anaerobic digestion process as a first step of a multistage strategy based on the aforementioned patented technology. The system combines: (1) an anaerobic digestion step

Please cite this article in press as: Zayen, A., et al. Incorporation of an anaerobic digestion step in a multistage treatment system for sanitary landfill leachate. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.04.030 2

(removal of biodegradable organic fraction in leachate) followed by (2) lime precipitation (reduction of conductivity and humic substances), (3) microfiltration (removal of suspended matter generated during the precipitation step) and (4) a final reverse osmosis step (complete removal of the remaining organic and inorganic matter). The objectives of this study are to evaluate the performance of this integrated process and in particular, to assess the impact of the anaerobic digestion step on the whole process. Thus, this first process was compared to a second treatment plan during which the anaerobic treatment is eliminated. Organic compounds in the raw leachate and after the reverse osmosis step were also investigated by gas chromatography coupled to mass spectrometry (GC–MS).

#### 2. Materials and methods

#### 2.1. Landfill leachate sampling

Landfill leachate was sampled from the controlled discharge of Sousse (Tunisia). This discharge has come into operation since July 2008 and receives 365 tons/day collected at three transfer centers. If not immediately analyzed, LFL was stored at 4 °C until use. Table 1 shows the physicochemical characteristics of the analyzed LFL.

## 2.2. Experimental set-up and reactor operation during anaerobic digestion of LFL

A pilot scale upflow anaerobic fixed bed bioreactor (UAFB) was previously adapted to treat LFL. The reactor, made of plexiglass, consists of a circular column with 100 cm in height and 25 cm in diameter and offers an effective volume of 20 L (Fig. 1). The fixed bed was packed with plastic carriers (Type HIFLOWE with a specific area of 200 m<sup>2</sup> m<sup>-3</sup>). The temperature was maintained constant at 37 °C by circulating water through the water jacket of the reactor. Daily produced methane was measured directly by using an alkaline solution, prior to the gas meter, which effectively absorbs all the carbon dioxide in the off gas (Guwy, 2004).

The hydraulic residence time (HRT) was maintained constant during all the treatment process (HRT = 4.5 d) and the organic loading rate (OLR) was increased from  $1 \text{ g COD } \text{L}^{-1} \text{ d}^{-1}$  to 3.3 g COD L<sup>-1</sup> d<sup>-1</sup> by decreasing the dilution of the raw LFL.

#### 2.3. Lime precipitation

Different lime concentrations were tested in order to determine the optimum dosage allowing a maximal reduction of electrical conductivity. The precipitation was performed following stages:

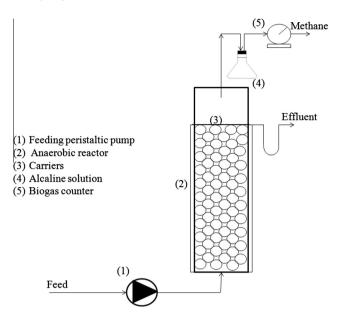


Fig. 1. Schematic diagram of the upflow anaerobic fixed bed reactor.

(1) 500 ml including leachate and lime milk ( $100 \text{ g L}^{-1}$ ) were prepared to have the desired concentration; (2) the stirrer was turned on for a rapid mixing stage of 5 min at 300 rpm; (3) the stirrer speed was decreased to allow a slow mixing for 30 min at 50 rpm; (4) the resulting suspension was allowed to naturally settle for 30 min; (5) the supernatant was withdrawn and pH as well as EC were measured in order to determine the optimum lime dosage.

#### 2.4. Microfiltration/reverse osmosis setup and operation

A schematic of the experimental filtration unit is shown in Fig. 2. During this test, only the dead end filtration cell system was used. Pressure was generated by a gear pump (Gather Industrie). Additional pressure was provided by a nitrogen gas cylinder. The filtration pressure was set to a fixed value through setting pump frequency and gas pressure. The fluid velocity was set in order to get a turbulent flow. LFL was held in a stainless steel feedwater reservoir with a total capacity of 500 ml. The concentrate stream was collected separately. All experiments were conducted at room temperature (15 °C ± 2 °C).

Microfiltration was conducted using PVDF membranes (Nadir PM MV020).

#### Table 1

Physico-chemical characteristics of the raw landfill leachate and the landfill leachate at different treatment stages for both treatment plans.

	RLFL	AD		Lime precipitation				Microfiltration				Reverse osmosis			
		Value	(%)	P1	(%)	P2	(%)	MF1	(%)	MF2	(%)	RO1	(%)	RO2	(%)
рН	8.107	8.181		10.902		12.631		_a		_a		_a		_a	
$EC (mS cm^{-1})$	59.4	43	27.61	18.3	57.44	42.8	27.95	17.5	4.37	40.3	5.84	0.33	98.11	3.98	90.12
$COD (mg O_2 L^{-1})$	15,200	3647	76.01	2007	44.97	7700	49.34	1808	9.91	5599	27.29	48.5	97.32	66.6	98.81
$BOD_5 (mg O_2 L^{-1})$	4599.2	908.58	80.24	645.24	28.98	2251.5	51.05	550	14.76	960.6	57.33	13.68	97.51	27.36	97.15
TOC (mg $L^{-1}$ )	5480	1487	72.86	1014	31.81	2960	45.98	948	6.51	2396	19.05	24.3	97.44	30.27	98.74
TSS (mg $L^{-1}$ )	1.28	0.44	65.62	0.06	86.36	0.82	35.94	ND	100	0.51	37.80	ND	100	ND	100
Alkalinity (mg $CO_2 L^{-1}$ )	3255	3175	2.46	620	80.47	2870	11.83	134	78.39	657	77.11	70.1	47.69	127	80.67
HS (mg $C_{HS} L^{-1}$ )	1112	487	56.20	127	73.92	451	59.44	ND	100	373	17.29	ND	-	ND	100
TKN (mg $L^{-1}$ )	2300.9	1980.6	13.92	545	72.48	1270	44.80	437.5	19.72	1060	16.67	33.2	92.41	171.5	83.82
$[NH_4^+]$ (mg L <sup>-1</sup> )	1400.3	1360.3	2.86	369	72.89	970	30.73	272,5	26.15	745	23.19	24.7	90.93	73	90.2
Abs 254 nm (diluted 1:25)	2.704	0.8823	67.37	0.4669	47.08	1.5429	42.94	0.3346	28.33	1.0488	32.02	0.0303	90.94	0.0429	95.9

Removal percentage in each treatment stage was calculated relatively to the previous step.

<sup>a</sup> pH was set to 10 during the microfiltration and reverse osmosis steps (in accordance with the pH range of MF and RO membranes).

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