Waste Management 55 (2016) 249–256

Contents lists available at ScienceDirect

Waste Management

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

Hollow fiber vs. flat sheet MBR for the treatment of high strength stabilized landfill leachate

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ARTICLE INFO

Article history:
Received 5 August 2015
Revised 27 November 2015
Accepted 20 December 2015
Available online 14 January 2016

Keywords:
Membrane bioreactor
Flat sheet
Hollow fiber
Stabilized leachate

ABSTRACT

The Membrane Bioreactor (MBR) technology is increasingly becoming a prominent process in the treatment of high-strength wastewater such as leachate resulting from the decomposition of waste in landfills. This study presents a performance comparative assessment of flat sheet and hollow fiber membranes in bioreactors for the treatment of relatively stable landfill leachate with the objective of defining guidelines for pilot/full scale plants. For this purpose, a laboratory scale MBR system was constructed and operated to treat a leachate with Chemical Oxygen Demand (COD) (3900–7800 mg/L), Biochemical Oxygen Demand (BOD_5) (~440–1537 mg/L), Total Phosphorus (TP) (~10–59 mg/L), Phosphate (PO_4^{3-}) (5–58 mg/L), Total Nitrogen (TN) (1500–5200 mg/L), and ammonium (NH_4^+) (1770–4410 mg/L). Both membranes achieved comparable BOD (92.2% vs. 93.2%) and TP (79.4% vs. 78.5%) removals. Higher PO_4^{3-} removal efficiency or percentage (87.3% vs. 81.3%) and slightly higher, but not statistically significant, COD removal efficiency were obtained with the hollow fiber membrane (71.4% vs. 68.5%). On the other hand, the flat sheet membrane achieved significantly higher TN and NH_4^+ removal efficiencies (61.2% vs. 49.4% and 63.4% vs. 47.8%, respectively), which may be attributed to the less frequent addition of NaOCl compared to the hollow fiber system.

1. Introduction

While at the bottom of the desirable hierarchy, landfilling, in many countries, continues to be a common element and often the only option adopted from the integrated system due to economic considerations. In landfills, leachate generation remains an inevitable consequence of the decomposition of the waste and the percolation of water through decomposing waste. Landfill leachate is invariably laden with various contaminants, whose characteristics are dependent on landfill age, precipitation, seasonal weather variation, and waste composition amongst other factors (Renou et al., 2008; Kulikowska and Klimiuk, 2008). Due to its complex and variable composition, leachate is difficult to treat (Tatsi and Zouboulis, 2002). Failing to properly treat the leachate is known to pollute the receiving environment (Kurniawan et al., 2006; El-Fadel et al., 1997). Thus, treating landfill leachate is increasingly subject to stringent environmental requirements to protect ground and surface water resources (Renou et al., 2008). Commonly adopted leachate management options include discharge into sewer systems for subsequent treatment with municipal wastewater (Çeçen and Çakiroğlu, 2001), recirculation (Rodríguez et al., 2004), evaporation followed by sludge disposal, and on-site treatment (Bodzek et al., 2006). In the latter context, various biological and physical/chemical technologies have been developed. Physical/chemical methods are usually adopted as pre/-post treatment or to remove specific pollutants (Renou et al., 2008). On the other hand, biological methods, which encompass several suspended and attached growth methods under either aerobic or anaerobic conditions, are often applied to treat the bulk of the biodegradable fraction in the leachate.

The Membrane Bioreactor (MBR) technology, a combination of membrane separation and biodegradation processes, is increasingly being recognized as the process treatment of choice for the treatment of high-strength wastewater, containing complex and recalcitrant compounds (Sutherland, 2010; Bilad et al., 2011). An MBR can be considered as a Conventional Activated Sludge (CAS) system with efficient membrane filtration that holds small particles (size <0.1 μm) (Santos et al., 2010). The main advantages of MBRs include the ability to replace the second stage of conventional wastewater treatment (i.e. gravity settling), produce a better quality effluent, and reduce reactor volume and footprint. Furthermore, an MBR is usually operated at a higher Mixed Liquor Volatile Suspended Solids (MLVSS), with values ranging between 8000 and 12,000 mg/L, as compared to the 2000–3000 mg/L range typically

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http://dx.doi.org/10.1016/j.wasman.2015.12.016
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reported in conventional activated sludge systems (Sutherland, 2010; Cornel and Krause, 2006; Alvarez-Vazquez et al., 2004).

With increasingly stringent discharge standards, conventional treatment methods (biological or physico-chemical) are seldom adequate to meet the standards. In combining biological degradation and physical separation, the MBR technology has shown satisfactory results in treating old/stabilized landfill leachate (Alvarez-Vazquez et al., 2004). In the context of leachate treatment, the MBR has been shown to have high BOD removal rates (90–99%), irrespective of experimental conditions and leachate maturity. In contrast, the efficiency of MBR in removing Chemical Oxygen Demand (COD) is known to vary widely from as low as 25% (Jakopović et al., 2008) to as high as 90% (Chen and Liu, 2006; Puszczało et al., 2010; Aloui et al., 2009).

While the literature on the use of MBR in wastewater treatment is relatively rich, studies examining the impact of various membrane types in an MBR system on the treatment efficiency of high strength stabilized landfill leachate are limited. A large proportion of the literature studies on leachate treatment by MBRs have employed HF membrane modules with a fewer number adopting the FS membrane modules (Cui et al., 2003; Le-Clech et al., 2006). Furthermore, data on phosphorus removal achieved by MBR treating stabilized leachate is scarce, with no data on removal achieved by flat sheet MBRs. In this study, the two most common membrane types, hollow fibers and flat sheet, were compared by testing them in an MBR system to assess their effectiveness in treating stabilized high strength landfill leachate with the objective of defining guidelines for a pilot/full scale plant. During the process, several parameters, including phosphate and total phosphorus, were monitored at different locations of the experimental setup.

2. Materials and methods

2.1. Experimental setup

The experimental setup (Fig. 1) consisted of two Plexiglas denitrification tanks (D) with stirrer mixers (C) to prevent settlement of solids and two aerobic Plexiglas tanks (E, L), one of which was equipped with a Flat Sheet (FS) membrane (Kubota, 203), while the other was fitted with a Hollow Fiber (HF) membrane (ZW 10). A blower (M) with a rotameter (Omega-FL-3663C) to regulate the airflow from a central air compressor was attached to each membrane to provide aeration and help in scrubbing the membrane and eliminate-minimize potential fouling. In addition, two pressure sensors (F) (Omega DPG 1000ADA or DAR) connected to a digital display were used to trace variations in membrane pressure. Peristaltic pumps (Master Flex 07528-10 and 7550-22) (I, K), with variable speed and reverse operation modes, were used for the permeate suction and recirculation. Both systems were fed with landfill leachate from a common storage tank connected to the denitrification tanks by means of a multi-channel peristaltic pump (A, B). The systems were connected to a drain (H) to allow sludge wastage and hence control of the Solid Retention Time (SRT). FS and HF membrane modules were chosen in this study because they are the two most commonly used membrane types (Stephenson et al., 2000). Both membrane modules exhibit advantages and disadvantages (Cui et al., 2003; Le-Clech et al., 2006): FS modules are less prone to fouling and relatively easy to control but are more expensive than HF modules which are more prone to fouling but can withstand vigorous backwashing.

2.2. Operation and control

The influent leachate was collected weekly from an operational sanitary landfill in Naameh–Lebanon and transported to the laboratory for characterization and usage in the reactors. The landfill is part of an integrated regional solid waste management system and receives over 2000 tons per day of municipal waste composed of a large fraction of organic food waste with high moisture content.

The experiment was initiated by filling the reactors with leachate, opening the aeration valves in the aerobic tanks, and turning on the mixers in the anoxic tanks at low speeds (≈150 rpm). The flow rate was increased gradually until a Hydraulic Retention Time (HRT) of 100 h (4.2 days) was achieved. A sludge retention time of 30 days was selected based on previous data from full scale MBR plants treating old landfill leachate (Alvarez-Vazquez et al., 2004), where an SRT of 30 days achieved superior treatment performance (Hasar et al., 2009a,b).

Foaming in the membrane tanks was controlled using an antifoaming agent (Sigma A6426-from Sigma Aldrich) (few drops almost twice per week in the first month, every two weeks afterwards). The HF membrane was cleaned twice a week using sodium hypochlorite (NaOCl) solution, while the FS membrane was cleaned by gentle scraping of solids.

The manufacturer of the flat sheet membrane recommended chemical cleaning for the membrane modules once every six months only. In contrast, the hollow fiber membrane modules needed weekly cleaning through backwashing with NaOCl solution. The cleaning process is equivalent to a repeated backpulse field operating condition (aimed to dislodge fouling material) whereby the membrane is backwashed with NaOCl solution after reversing the direction of the flow through the pump. This Cleaning In Place (CIP) of the HF membrane is conducted to avoid removing the membrane module from the aerobic tank.

2.3. Analytical methods

Throughout the experimental program (127 days), samples were collected twice a week from the feed tank and permeate and once per week from all tanks. Samples were analyzed for several indicators including pH, Biochemical Oxygen Demand (BOD_5), Total Nitrogen (TN), ammonium (NH_4^+), COD, Total Phosphorus (TP), and phosphate (PO_4^{3-}) according to Standard Methods of the American Public Heath Association (APHA) (APHA, 2005). The pH was measured using a Thermo Scientific Orion 3 STAR pH Benchtop meter, the DO for BOD determination was measured using a WTW Oxi 538 Oximeter, while other parameters were quantified using spectro-photometric analysis that was performed using a HACH DR/2010 Spectrophotometer. Collected data were then analyzed using R 3.03 statistical software (R Core Team, 2014). Differences between the two MBR systems with respect to the achieved removal efficiencies of various parameters were quantified. Statistical comparison was conducted by running paired *t*-tests, when the data were normally distributed, and the paired Wilcoxn Signed Rank test, when the normality assumption was violated. The confidence level was set to 95% (a significance level of 0.05).

3. Results and discussion

Leachate characterization (Table 1) showed high levels of TN (1500–5200 mg N/L), pH values ranging between 8.08 and 8.87, and a low BOD_5/COD ratio (0.07–0.22), all reflecting a stabilized leachate (Aloui et al., 2009; Jakopović et al., 2008; Trebouet et al., 1999). However, ammonium and TP levels (1770–4410 mg/L and 10.5–59 mg/L, respectively) were high and more typical of a young leachate (1400–10,250 mg/L for ammonium and 1.6–655 mg/L for TP).

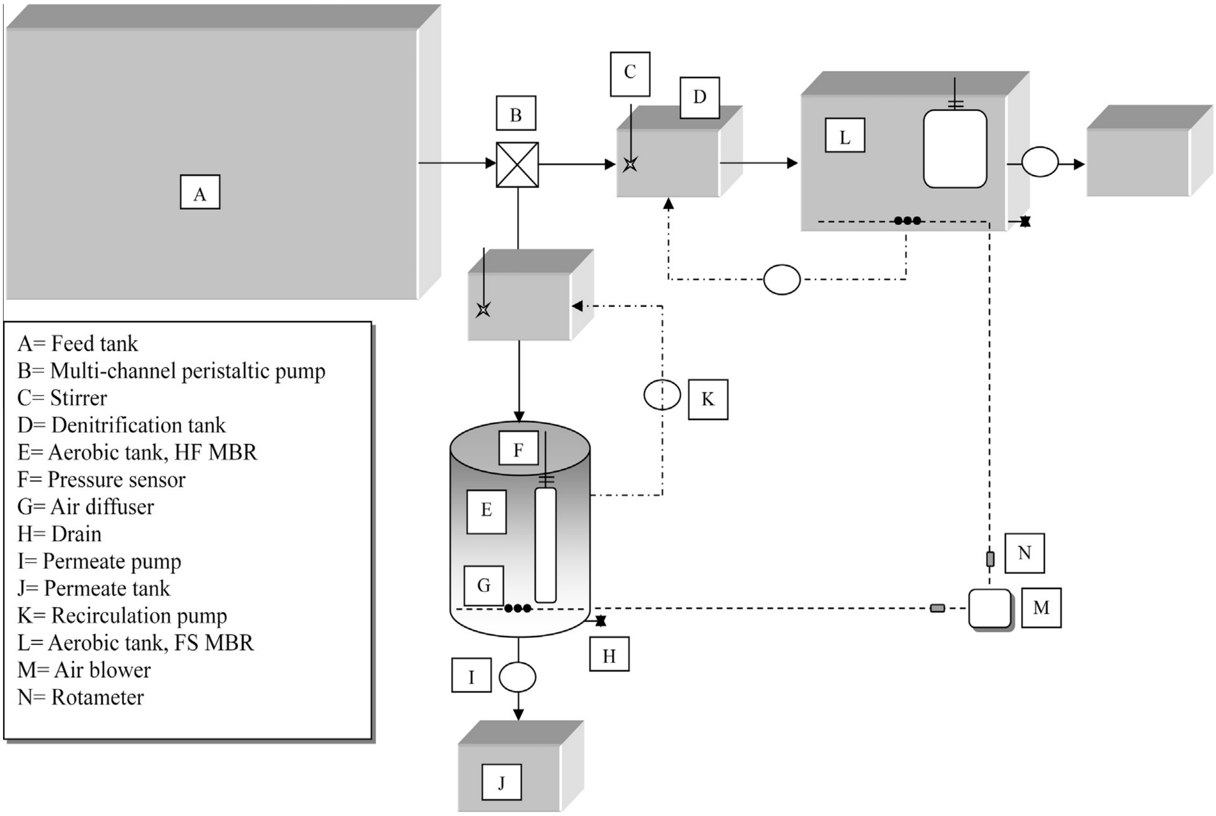

Fig. 1. Experimental setup.

Table 1
Comparison of leachate quality with literature reported ranges.

Parameter	Units	Measurement methods	Literature		Experimental	
			Old leachate[b]	Fresh leachate[b]	Range	Average[c]
pH		SM[a] 4500-H+ B	7.3–8.8	4.9–6.7	8.1–8.9	8.43
BOD_5	mg/l	SM 5210 B	50–4,200	9,500–80,795	440–1,537	695
COD	mg/l	SM 5220 D	685–15,000	44,000–115,000	3,900–7,800	5,978
NH_4^+	mg-N/l	Hach 8155	39–1750	1,400–10,250	1,770–4,410	2,464
TN	mg-N/L	Hach 10072			1,500–5,200	2,543
TP	mg/l	SM 4500 P B (5), E	1.27–19.9	1.6–655	10.5–59	31
PO_4^{3-}	mg/l	SM 4500-P-E			5–58	30

[a] Standard methods.
[b] Reported in Tatsi and Zouboulis, 2002.
[c] Average of all encountered values measured throughout the experiment.

3.1. Performance assessment

Samples from the influent, denitrification, aerobic, and effluent tanks for both MBRs were collected and analyzed on a weekly basis. The temporal variations of the tested indicators (Fig. 2 for the flat sheet membrane system and Fig. 3 for the hollow fiber membrane system) elucidate the contribution of denitrification and the aerobic tanks towards the treatment process. Table 2 compares the influent to the effluent concentrations as well as the removal efficiencies of pollutants for the two systems.

The results show that BOD_5 removal rates across the two MBRs were comparable (Table 2). The effluent BOD_5 remained invariably lower than 62 and 74 mg/L for the FS and HF membrane systems, respectively, although the Organic Loading Rate (OLR) fluctuated between 0.94 and 1.87 g COD/L.d. While the leachate strength in this study (COD: 3,900–7,800 mg/L) was on the higher end of those reported in the literature for old stable leachate (136–8,000 mg/L), similar, and sometimes better, COD attenuation rates (68–71%) were achieved albeit at relatively lower HRT (4.2 days) and SRT (30 days) (vs. HRT of 1–10 days and SRT of 15-infinite days) (Table 3). However, the effluent characteristics exceeded the local standards for the discharge in water bodies, for BOD (50 for FS and 55 for HF vs. 25 mg/L), COD (1868 for FS and 1689 for HF vs. 125 mg/L, TN (968 for FS and 1265 for HF vs. 30 mg/L) ammonium (922 for FS and 1283 for HF vs. 10 mg/L) but not for TP (6.3 for FS and 6 for HF vs. 10 mg/L). Note that the aerobic tanks experienced higher values of COD than the influent tank, which can be attributed to further hydrolysis of organics in the leachate (Nikolaou et al., 2010) and the disintegration of dead cells.

The high COD removal efficiency (83–87%) reported by Zhang et al. (2013) pertains to weaker pre-treated leachate undergoing Fenton oxidation prior to being fed to the MBR system. Ratanatamskul and Nilthong (2009) also reported 83% COD removal, when treating a weaker leachate with an influent COD of 1000 mg/L. Their results were based on running a combined Biological Powdered Activated Carbon (BPAC)-MBR system, whereby

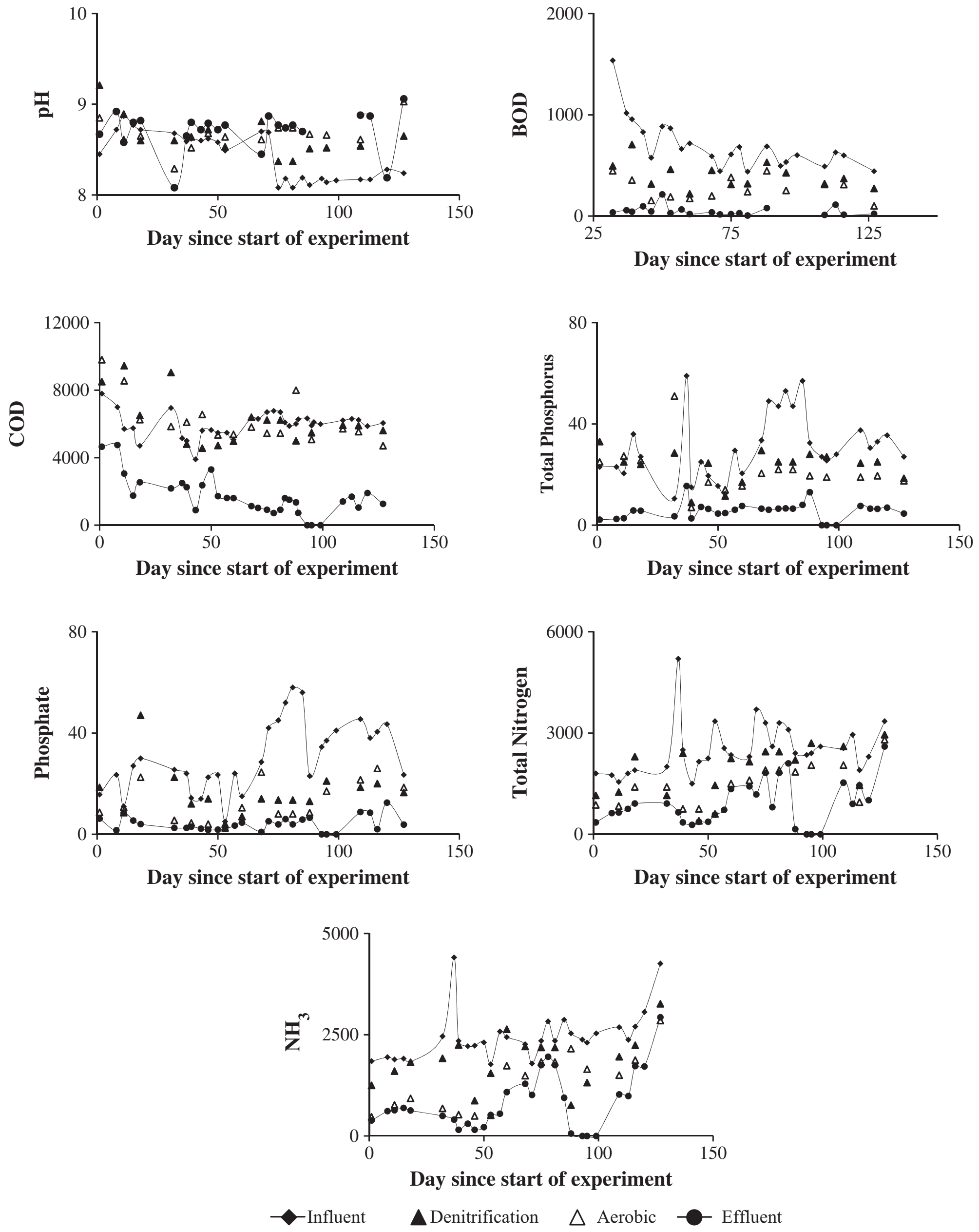

Fig. 2. Temporal variation of main performance indicators: flat sheet membrane.

the Powdered Activated Carbon (PAC) contributed to the removal of biologically recalcitrant-resistant compounds. Other superior COD removal rates were reported by Litas et al. (2012) and Puszczało et al. (2010) (95% and 89%, respectively), who resorted to mixing the leachate with synthetic wastewater that helped preventing the toxic effect of the high ammonium concentrations, typically encountered in leachate, and providing an extra phosphorus source (Hasar et al., 2009a).

The average COD removal efficiency achieved with the HF membrane was slightly higher as compared to the FS system. Yet, these differences are not statistically significant (paired Wilcoxon sign rank test, *p*-value = 0.918), mostly due to the high variability in the removal rates in both systems. Overall, the COD concentrations in the effluent of both systems were relatively high due to the presence of refractory compounds, typically associated with leachate exhibiting a low BOD_5/COD ratio (0.07–0.22). Better COD removal efficiencies could be obtained when applying the aerobic MBR technology to the treatment of young leachate with a higher biodegradability (i.e. higher BOD/COD ratio).

While the leachate utilized in this study exhibited higher levels of TP (31 ± 13 mg/L) as compared to other mature leachate (1.27–24 mg/L) (Tatsi and Zouboulis, 2002; Xie et al., 2010), both membranes achieved high and comparable TP removal rates (78.5 ± 9.0% and 79.4 ± 7.8% for the FS and HF, respectively). These rates are higher than those previously reported in the literature (60.5–74.3%) (Gürel and Büyükgüngör, 2011; Liu et al., 2012; Liu and Lv, 2012). As for phosphate, influent concentrations were high. Phosphate removals for the HF were consistently greater than 83%, except for two instances when the rate dropped to lower than 35%. FS removal efficiencies showed large variability with removal efficiencies ranging between 58% and 96.8% (expect for one instance where the rate dropped to 24%). The removal efficiencies between the two MBR systems were found to be statistically different (paired Wilcoxon Signed Rank test; *p*-value = 0.0236), with better

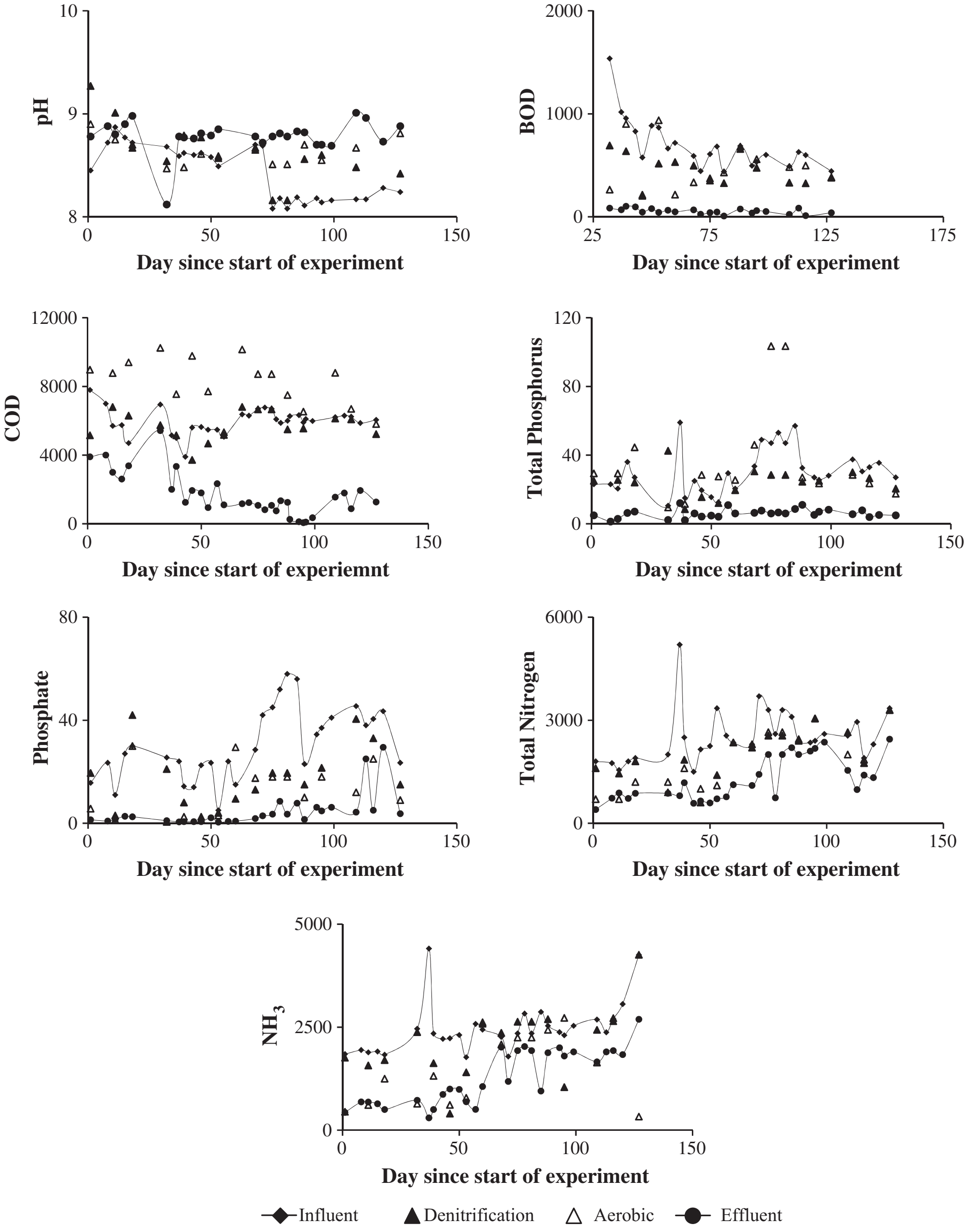

Fig. 3. Temporal variation of main performance indicators: hollow fiber membrane.

Table 2
MBR performance assessment.

Parameter	Mean influent mg/L (±1 sd)	Mean FS_Effluent mg/L (±1 sd)	Mean HF_Effluent mg/L (±1 sd)	Mean FS_Removal efficiency % (±1 sd)	Mean HF_Removal efficiency % (±1 sd)	Local discharge standards[a] mg/L
BOD_5	695 (250.6)	50 (49.4)	55 (26.1)	93.2 (5.9)	92.2 (3.1)	25
COD	5,978 (664.3)	1868 (650.7)	1,689 (1142.5)	68.5 (13.1)	71.4 (19.9)	125
TP	31 (13)	6.3 (2.9)	6 (2.6)	78.5 (9)	79.4 (7.8)	10
PO_4^{3-}	30.5 (13.8)	4.5 (2.8)	4.5 (6.7)	81.3 (15.6)	87.3 (15.6)	–
TN	2543 (773)	986 (618.7)	1265 (627.7)	61.2 (20.6)	49.4 (21.8)	30
NH_4^+	2464 (617.4)	922 (695.7)	1283 (668.2)	63.4 (22.7)	47.8 (23.7)	10

± Refers to standard deviation values.
[a] MoE (2001).

removal efficiency achieved with the HF system. The lower phosphate removal in the FS tank may be attributed to the higher nitrate concentration in the anoxic tank since high levels of nitrate in anoxic reactors have been reported to negatively affect the uptake and release of phosphate (Monclús et al., 2010; Puszczało et al., 2010).

Nitrogen in the leachate was primarily in the form of NH_4^+ (around 96.8% of the TN). The influent NH_4^+ concentrations

Table 3

Literature reported values of old leachate COD removal efficiency using MBR technology.

Reference	Location	Scale	Process	Membrane configuration	Influent characteristics		Operational conditions		Removal efficiency COD, %
					COD, mg/L	BOD/COD (age)	HRT, days	SRT, days	
This study	Lebanon	Lab	Anoxic-aerobic	Sub (FS)	3900–7800	0.12 (O)	4.16	30	68.5
This study	Lebanon	Lab	Anoxic-aerobic	Sub (HF)	3900–7800	0.12 (O)	4.16	30	71.4
Zhang et al. (2013)[b]	China	Lab	Fenton oxidation + MBR + RO	Sub (HF)	1200–1600[a]	0.09–0.12	4	45	83–87.5
Litas et al. (2012)	Greece	Pilot	SMBR (SBR) mixture of LFL + Synthetic WW (1:1)	Sub (FS)	1772	(O)	9	–	95
Chiemchaisri et al. (2011)	Thailand	Pilot	2-stage MBR (anoxic tank + aerobic MBR)	Sub (HF)	2605–7318	(O + Y) mixed feed	0.5 (MBR tank)	–	60–78
Akkaya et al. (2010)[b]	Turkey	Lab	UASB + MBR + MAP	Sub	4250[a]	(M)	–	–	10–70
Puszczało et al. (2010)[b]	Poland	Lab	Mixture of 10% LFL + s synthetic WW/SBR	Sub (MF/Cap)	3000–3500	0.06(O)	2–3	15	89
Aloui et al. (2009)[b]	Tunisia	Lab	Stirred tank reactor	Ext (MF/Tub)	7100–8000	0.18(O)	2–3	–	70–77
Feki et al. (2009)[b]	Tunisia	Lab	MBR/electrochemical oxidation	Ext (Tub)	6500–8000	0.09 (O)	–	–	61
Ratanatamskul and Nilthong (2009)	Thailand	Lab	BPAC-MBR	Sub (HF)	5000–6000 1000[a]	~0.1 (O)	1	Inf.	83
Svojitka et al. (2009)	Germany	Bench	Compartmentalized activated sludge tank	Ext (UF/Tub)	2200	<0.05	2.92–7.08	100	<30
Sadri et al. (2008)	Canada	Lab	Stirred tank reactor	Sub (HF)	2737–4079	0.11–0.18 (O)	1–3.5	30, 60	54–78
Tsilogeorgis et al. (2008)	Greece	Bench	MSBR	Sub (UF/HF)	1391–3977	(O)	10	Infinite	40–60
Robinson (2007)[b]	UK	Full	3 aerobic biological tanks in series	Ext (UF/Tub)	5000	0.05	–	–	76
Canziani et al. (2006)	Italy	Pilot	MBR + MBBR	Sub (Tub)	6316	0.3 (O)	–	>45	Up to 75
Schwarzenbeck et al. (2004)[b]	Germany	Full	2 reactors in series (denitrification + nitrification)+AC filter	MF	136–1980	~0.2	–	–	65
Setiadi and Fairus (2003)	Indonesia	Lab	Stirred tank reactor (hazardous waste)	Ext (MF/HF)	1800	0.15–0.17	1	32	31.3
Ahn et al. (2002)[b]	South Korea	Full	Aeration basin with anoxic + aerobic parts	Sub (MF/HF)	400–1500	(O)	–	–	~38

AC: Activated Carbon; BPAC: Biological Powdered Activated Carbon; Cap: Capillary; Ext: External; FS: Flat Sheet; HF: Hollow Fiber; LFL: Landfill Leachate; M: Medium; MAP: Magnesium Ammonium Phosphate; MF: Microfiltration; MBBR: Moving-Bed Biofilm Reactor; MSBR: Membrane Sequencing Batch Reactor; O: Old; RO: Reverse Osmosis; SBR: Sequencing Batch Reactor Sub: Submerged; Tub: Tubular; UASB: Up-flow Anaerobic Sludge Blanket; UF: Ultrafiltration; WW: Waste Water; Y: Young.

[a] Concentrations after pretreatment or dilution.

[b] Applied post-treatment to MBR (efficiencies are for MBR only).

(1770–4410 mg/L) were found to be higher than the ranges reported for old leachate by both Tatsi and Zouboulis (2002) (39–1750 mg/L) and by Xie et al. (2010) (1700–2000 mg/L). The achieved NH_4^+ removal rates differed across the two MBR systems. The FS membrane achieved significantly higher TN and NH_4^+ removal rates as compared to the HF membrane (61.2 ± 20.6% vs. 49.4 ± 21.8% for TN and 63.4 ± 22.6% vs. 47.8 ± 23.7% for NH_4^+; *t*-test *p*-value <0.05). These differences may be attributed to the more frequent addition of NaOCl required for the HF system cleaning. The compound may inhibit the growth of nitrobacter or nitrosomonas that are reportedly necessary for nitrogen removal (AWWA, 2006). Similarly, the moderate NH_4^+ removal rates achieved by both systems may be attributed to the potential inhibiting effect of high ammonia concentrations (>1000 mg/L) on nitrobacter and nitrosomonas species (Ince et al., 2013; Ahn et al., 2002; An et al., 2006; Ahmed and Lan, 2012; Wichitsathian et al., 2004). High levels of NH_4^+ in leachate are a result of the lack of a mechanism capable of removing ammonia under the landfills' methanogenic conditions, leading to its accumulation (Kurniawan et al., 2010; Kaczorek and Ledakowicz, 2006). As such, establishing a pre-treatment for NH_4^+ removal prior to MBR treatment could be vital; ammonia stripping could be a suitable option. Failing to do so could have long-term concerns that extend beyond the landfill lifetime (Berge et al., 2006), whereby the discharge of high ammonium leachate into surface water may cause the release of nitrous oxide into the atmosphere and promote eutrophication and aquatic toxicity (Philips et al., 2002).

4. Conclusion

The performance of the flat sheet MBR and hollow fiber MBR for the treatment of high strength stabilized landfill leachate was demonstrated at a laboratory scale using leachate collected from an active landfill. The leachate showed characteristics typical of both fresh and old leachate. Comparable BOD and TP removal rates were obtained with the FS and HF membranes (93.2% vs. 92.2% for BOD, 78.5% and 79.4% for TP). Yet, slightly higher but not statistically significant COD removal efficiencies were obtained with the HF membrane (71.4% vs. 68.5%). While phosphate removal rates were higher for HF membrane (87.3%) when compared to the FS membrane (81.3%), the FS membrane achieved significantly higher TN and ammonium removal rates when compared to the rates for the HF membrane (61.2% vs. 49.4% for TN and 63.4% vs. 47.8% for ammonium). Nevertheless, additional polishing treatment will be required for compliance with local standards for discharge into water bodies. The effluent characteristics exceeded local standards for BOD, COD, TN ammonium but not for TP. The results help in anticipating potential constraints that may be faced at the full scale leachate treatment plant, whereby a successful system should account for the non-biodegradable COD fraction using a pre/post physical/chemical process (such as coagulation/flocculation) and the high NH_3 concentration using ammonia stripping for instance, to reduce the influent NH_3 concentration.

Acknowledgements

The authors are indebted to ACWA and GE Power & Water for providing the membranes and for their guidance during the installation of the experimental setup. Special thanks are extended to the Council for Development & Reconstruction for providing access to the Naameh landfill as well as to Laceco and the landfill management for facilitating and assisting during leachate samples collection.

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