



AMERICAN UNIVERSITY OF BEIRUT

MBRS FOR THE TREATMENT OF HIGH STRENGTH OLD  
LANDFILL LEACHATE: HOLLOW FIBER VS. FLAT SHEET

by  
JIHAN MOHAMAD HASHISHO

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submitted in partial fulfillment of the requirements  
for the degree of Master of Science in Environmental Sciences  
to the Interfaculty Graduate Environmental Science Program  
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of the Faculty of Engineering and Architecture  
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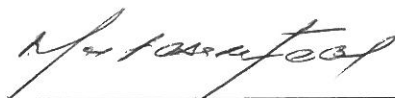
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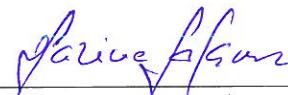
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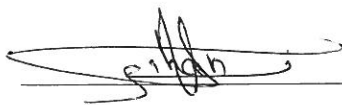
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## AN ABSTRACT OF THE THESIS OF

Jihan Mohamad Hashisho for Master of Science in Environmental Sciences  
Major: Environmental Technology

Title: MBRs for the treatment of high strength old landfill leachate: Hollow fiber vs. flat sheet

The generation of landfill leachate remains the main problem associated with municipal solid waste landfilling. In this study, membrane bioreactors (MBRs) were tested for leachate treatment through a combination of denitrification and aerobic processes to examine and compare the effectiveness of two membrane formats: hollow fiber and flat sheet.

For this purpose, a laboratory scale MBR was constructed and operated to treat a leachate with COD (3,900-7,800 mg/L), BOD<sub>5</sub> (439.74-1536.67 mg/L), TP (10.5-59 mg/L), PO<sub>4</sub><sup>3-</sup> (5-58), TN (1,500-5,200 mg/L), and NH<sub>3</sub> (1,770-4,410 mg/L). Both membranes, the flat sheet and hollow fiber, achieved comparable BOD (92.2 vs. 93.2%) and TP (79.4 vs. 78.5%) removals. However, while slightly higher COD and phosphate removal efficiencies were obtained with the Hollow Fiber membrane (71.4 vs. 68.5% and 87.3 vs. 81.3%, respectively), significantly higher TN and ammonia removal rates (61.2 vs. 49.4 and 63.4 vs. 47.8, respectively) were achieved by the Flat Sheet membrane. The experimental results contribute in filling a gap towards managing stabilized landfill leachate and providing guidelines for corresponding MBR applications

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## ABBREVIATIONS

CAS	:	Convetnional Activated Sludge
MBR	:	Membrane Bioreactor
MSW	:	Municipal Solid Waste
SBR	:	Sequencing Batch Reactor

# CHAPTER 1

## INTRODUCTION

### 1. Background

Municipal solid waste (MSW) generation has been continuously increasing as a result of population growth, development, and changes in lifestyles (Renou *et al.*, 2008). Landfilling remains the most common method for MSW disposal. In Lebanon, it is currently the prevailing alternative relied upon for the disposal of MSW. However, leachate formation and potential adverse impacts continue to be an inevitable consequence of landfilling due to inherent moisture content of the waste, biochemical, chemical and physical reactions and rain water percolation (Renou *et al.*, 2008). Thus, environmental monitoring is required during the design, operation, and post-closure period because of the produced leachate which presents serious threat to the nearby ground and surface water bodies, when left untreated (Wiszniowski *et al.*, 2006).

However, designing a common landfill leachate treatment system is difficult namely because of the versatility in landfill leachate quantity and quality. In this context, several conventional methods such as Conventional Activated sludge (CAS), sequencing batch reactors (SBR), aerated lagoons... have been commonly applied and investigated for the treatment of landfill leachate, namely young leachate with high BOD/COD (Renou *et al.*, 2008). Developments in membrane technology was also exploited to test the feasibility of treating leachate with various membranes: microfiltration (Piatkiewicz *et al.*, 2001), ultrafiltration (Tabet *et al.*, 2002), reverse osmosis (Peters, 1998), and nanofiltration (Trebouet *et al.*, 2001). More recently, combining membrane separation and biodegradation processes led to the development

of the membrane bioreactor (MBR) technology (Sutherland, 2010) which is increasingly recognized as the process of choice for the treatment of high-strength wastewater containing complex and recalcitrant compounds (Bilad *et al.*, 2011).

## **2. Objectives and Scope of Work**

The present study targets the treatability of the leachate generated from the landfill of Naameh. For this purpose, the leachate from the Landfill was first characterized for various indicators recognized as potential contaminants as well as a cause of eutrophication in receiving water bodies. A laboratory scale experimental program was developed and implemented to test a treatment system consisting of anoxic and aerobic tanks containing Flat Sheet and Hollow Fiber membrane modules.

## **3. Thesis structure**

Besides this introductory chapter, the thesis consists of two appendices which include the detailed results, discussions and conclusions.

- *Appendix A* is a review article. It is an elaborate critical review of the literature on the treatment of leachate using Membrane Bioreactor.
- *Appendix B* is a research article. It presents the analytical results and findings from including a comparison with the literature.
- *Appendix C* is a supplementary material section. It presents a summary of the data collected as well as pictures of the experimental setup adopted.

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# APPENDIX A MEMBRANE BIOREACTOR TECHNOLOGY FOR LANDFILL LEACHATE TREATMENT

## ABSTRACT

Controlled landfilling remains the most economic and commonly applied method for Municipal Solid Waste (MSW) disposal with leachate generation being an inevitable consequence of the decomposition of the waste and the percolation of water through decomposing waste. The Membrane Bioreactor (MBR) technology has been shown to be effective in treating such high strength wastewater streams because of its ability in retaining high biomass concentration while allowing for membrane separation. This paper presents a critical review of the application of the MBR technology for the treatment of leachate and evaluates reported performance while highlighting factors affecting MBR operation. The paper concludes with outlining existing gaps and future research needs to improve the understanding and performance of the MBR technology for leachate treatment.

## KEYWORDS

Municipal solid waste, Landfill leachate treatment, Membrane Bioreactor

## ABBREVIATIONS

*AC*= Activated Carbon; *Aer* = Aerobic; *An*= Anoxic; *Anammox* = ANaerobic AMMonium OXidation; *anMBR*= anaerobic MBR; *BNR*= Biological Nitrogen Removal; *BPA*= Bisphenol A; *BPAC*= Biological powdered activated carbon; *BOD*= Biochemical Oxygen Demand; *Cap*= *Capillary*; *CAS*= Conventional Activated Sludge; *COD*= Chemical Oxygen Demand; *DGGE*= Denaturing Gradient Gel Electrophoresis; *DO*= Dissolve Oxygen; *DOC*= Dissolved Organic Carbon; *EPS*= Extracellular Polymeric Substances; *Ext*= External; *FS*= Flat Sheet; *GAC*= Granular Activated Carbon; *HF*= Hollow Fiber; *HR*=

Hydrolytic Reactor; *HRT*= Hydraulic Retention Time; *LFL*= Landfill Leachate; *Inf*= Infinite; *M*= Medium; *MAP*= Magnesium Ammonium Phosphate; *MBR*= Membrane Bioreactor; *MF*= Microfiltration; *MLSS*= Mixed Liquor Suspended Solids; *MLVSS*= Mixed Liquor Volatile Suspended Solids; *MSBR*= Membrane coupled Sequencing Batch Reactor; *MSW*= Municipal Solid Waste; *MW*= Molecular Weight; *MW<sub>p</sub>*= Molecular Weight of the peak; *ND*= Not Detected; *NH<sub>3</sub>*= Ammonia; *NH<sup>4+</sup>*= ammonium; *NF*= Nanofiltration; *NLR*= Nitrogen Loading Rate; *NO<sub>3</sub><sup>-</sup>*= Nitrate; *NP*= Nonylphenol; *O*= Old; *OCP*= OrganoChlorine Pesticides; *PAC*= Powdered Activated Carbon; *PAH*= Polycyclic Aromatic Hydrocarbons; *RO*= Reverse Osmosis; *SAMBR*= Submerged Anaerobic Membrane Bioreactor; *SBR*= Sequencing Batch Reactor; *sCOD*= soluble COD; *SEM*= Scanning Electron Microscopy; *SHARON*= Single reactor system for High activity Ammonium Removal Over Nitrite; *SMBR*= Submerged Membrane Batch Reactor; *SMP*= Soluble Microbial Product; *SRT*= Solid Retention Time ; *SS*= Suspended Solids; *Sub*= Submerged; *SUVA*= Specific UV Absorbance; *TKN*= Total Kjeldahl Nitrogen; *TN*= Total Nitrogen; *TOC*= Total Organic Carbon; *TP*= Total Phosphorus; *TSS*= Total Suspended Solids; *Tub*= Tubular; *UASB*= Upflow Anaerobic Sludge Blanket; *UF*= Ultrafiltration; *VSS*= Volatile Suspended Solids; *WW*= WasteWater; *Y*= Young; *ZVI*= Zerovalent Iron

## **A.1. INTRODUCTION**

Solid waste management has evolved from open dumping to integrated systems involving various physical, biological, and/or thermal processes with landfills invariably constituting the last element in a system. Although at the bottom of the desirable hierarchy, landfilling remains an important element of most solid waste management schemes because all other elements result mostly in waste minimization with residuals inevitably disposed of in landfills, despite the emerging concept of “zero waste”. In fact, in many countries, landfills continue to be the most attractive element and often the only one adopted from the



integrated system due to economic considerations. In this context, leachate generation remains an inevitable consequence of the decomposition of the waste and the percolation of water through decomposing waste. Leachate is formed when the refuse moisture content exceeds its field capacity, which is defined as the maximum moisture that is retained in a porous medium without producing downward percolation. This process is influenced by many factors which can be divided into those that contribute directly to landfill moisture (rainfall, snowmelt, ground water intrusion, initial moisture content, irrigation, recirculation, liquid waste co-disposal, and refuse decomposition) and those that affect leachate or moisture distribution within the landfill (refuse age and pre-treatment, compaction, permeability, particle size, density, settlement, vegetation, cover, sidewall and liner material, gas and heat generation and transport) (El-Fadel *et al.* 1999; 2002; 2003). Once formed, leachate creates a non-uniform and intermittent percolation of moisture through the refuse mass, which results in the removal of soluble organic and inorganic compounds commonly encountered in the refuse at emplacement or are formed as a result of chemical and biological processes within the landfill. The by-products of these processes affect leachate quality particularly in the early stages of organic matter decomposition after refuse emplacement. In addition, leachate quality can exhibit considerable spatial and temporal variations depending upon site operations, refuse characteristics (composition and age), and internal landfill processes requiring varied management options.

Leachate percolation to underlying aquifers contaminates groundwater and threatens public health (Kurniawan *et al.* 2006; El-Fadel *et al.* 1997). Thus, the proper management and treatment of Landfill Leachate (LFL) is increasingly subject to stringent treatment standards to protect ground and surface water resources (Renou *et al.* 2008).

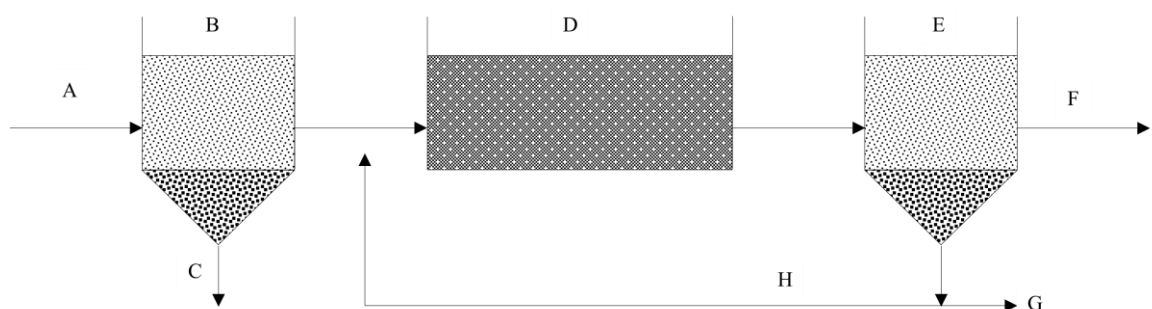
Historically various technologies encompassing a wide range of physical, chemical, and biological processes have been examined for the treatment of leachate (Renou *et al.* 2008; Wiszniowski *et al.* 2006). More recently, combining membrane separation and biodegradation processes led to the development of the Membrane Bioreactor (MBR) technology, which is increasingly recognized as the process of choice for the treatment of wastewater streams containing complex and recalcitrant compounds (Bilad *et al.* 2011).

This paper presents a critical review of the reported literature on the state and development trends in leachate treatment using the MBR technology. It defines the main determinants controlling the operation, performance, and limitations of the technology and concludes in outlining corresponding gaps and future research needs.

## A.2. MEMBRANE BIOREACTOR TECHNOLOGY

### A.1.1 MBR fundamentals

An MBR can be considered as a Conventional Activated Sludge (CAS) system with efficient membrane filtration that holds very small pore size of less than 0.1  $\mu\text{m}$  (Santos *et al.* 2010). It replaces the second stage of conventional wastewater treatment (i.e. gravity settling)



& A-2) MBRs have been used to treat domestic and industrial wastewater from various sources including food and meat, pharmaceutical, paper and pulp, textile, winery, and oil (Bolzonella *et al.* 2010; Abegglen & Siegrist 2006; Yang *et al.* 2006; Juang & Tsai 2006;

Badani *et al.* 2005).

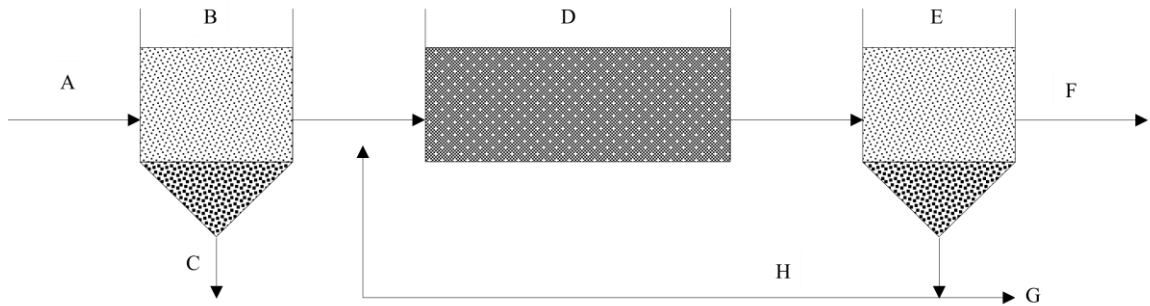


Figure A-1: Typical configuration of a Conventional Activated Sludge (CAS) system

A: Influent; B: Primary Clarifier; C: Return Sludge; D: Bioreactor; E: Secondary Clarifier; F: Treated Effluent; G: Waste Sludge; H: Return Sludge

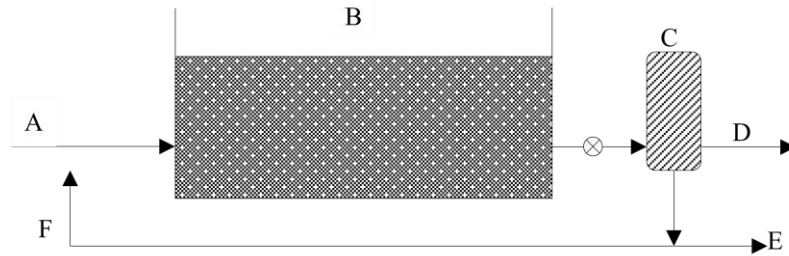


Figure A-2: Typical configuration of a Membrane Bioreactor (MBR) system

A: Influent; B: Bioreactor; C: Membrane; D: Treated Effluent; E: Waste Sludge; F: Return Sludge

In general, the submerged MBR can be found in the form of vertical flat sheets, horizontal or vertical hollow fiber and less frequently as tubes, although the latter format is preferred for side-stream operation. Whilst fluid dynamics and distribution might be easier to control for flat sheet and tubular membrane modules than for hollow fiber membranes, the latter are less expensive, can withstand vigorous backwashing and permit higher membrane density (Cui *et al.* 2003; Le Clech *et al.* 2006).

### A.2.2 MBR advantages and disadvantages

The MBR technology offers several advantages over the CAS system such as reduced foot print, high effluent quality, replacement of post-digestion settlement and clarification, less sludge production, ease of retrofitting to existing works and lower energy demand (although

the latter is inconclusive) (Sutherland 2010; Melin *et al.* 2006; Metcalf & Eddy 2003). When compared to CAS, MBRs produce a better quality effluent while offering a reduced reactor volume and footprint with the possibility of operating at a higher Mixed Liquor Volatile Suspended Solids (MLVSS) whereby the MLSS concentrations in MBR systems can reach 8,000-12,000 mg/L compared to 2,000-3,000 mg/L in a CAS system (Sutherland 2010). On the other hand, additional costs, related to the operation and maintenance of the MBR, are incurred with membrane systems particularly for membrane aeration, cleaning and replacement over the lifetime of the reactor (Cote *et al.* 2012).

### **A.2.3 MBR limitations and constraints**

Membrane fouling and foaming remain the main operational limitation against the widespread of the MBR technology (Le Clech *et al.* 2006). While there is no clear definition of the exact fouling phenomena occurring during filtration, Extracellular Polymeric Substances (EPS) have been widely reported to play a key role in this regard. More specifically, several studies have shown that the biomass supernatant “Soluble Microbial Product” (SMP) and its carbohydrate fraction are the main determinants affecting fouling (Rosenberger *et al.* 2005; Dvořák *et al.* 2011; Chang *et al.* 2002). The complex interaction of hydrodynamics, mass transfer, biological degradation, and existing compounds makes it difficult to isolate all the parameters that could help in predicting membrane fouling (Kraume & Drews 2010), although the most critical elements in this context include the mode of operation (constant flux vs. constant TMP) (Defrance & Jaffrin 1999; Vyas *et al.* 2002), the filtration time (short-term vs. long-term) (Yang *et al.* 2006; Zhang *et al.* 2006), the operating conditions and cleaning procedures (Trussell *et al.* 2006; Cui *et al.* 2003), and the initial stage of the membrane (new vs. cleaned) (Le Clech *et al.* 2003). Irrespective, MBR users have resorted to several methods to control fouling such as back-pulsing or bubbling (Prieske *et al.* 2010), the use of additives (Kraume & Drews 2010)

or sponge-like carriers (Ngo *et al.*, 2008), pre-settling of biomass (Ivanovic & Leiknes 2008), sludge granulation, and membrane surface change (Kraume & Drews 2010).

Foaming constitutes the other great challenge facing MBR application. It is a complex process that has not been extensively examined. Although there is no apparent difference between foaming in CAS and that in MBR systems, the latter can retain more EPS in the reactor thus contributing to more foaming even in the absence of foam-forming micro-organisms / filamentous bacteria (Cosenza *et al.* 2013; Di Bella & Torregrossa 2013). It is noteworthy that while fouling is generally known to increase with EPS levels, it has been reported that less fouling was observed with foaming events probably because bound EPS are trapped in the floating scum rendering the mixed liquor near the membrane less hydrophobic and resulting in a decrease in fouling rate (Cosenza *et al.* 2013).

#### A.2.4 MBR modeling

Given the limitations of experimental work, modeling studies become critical in complementing the design of MBR systems and corresponding performance prediction. Reported MBR modeling efforts have generally focused on biomass kinetics and membrane fouling (Table B-4).

Table A-1: A summary of modeling applications

Reference	Model	Application and main results
Wintgens <i>et al.</i> 2003 b	ASM	<ul style="list-style-type: none"> <li>– Full scale operational MBR plant</li> <li>– Good correspondence between the simulation results from ASM under steady state conditions and experimental values for ammonia, nitrate + nitrite and COD</li> </ul>
Urbain <i>et al.</i> 1998	SMP	<ul style="list-style-type: none"> <li>– MBR pilot plant under steady and transient state conditions</li> <li>– Good correspondence between SMP simulation results and measured values of VSS, effluent COD and nitrogen species under steady-state and transient-state conditions.</li> <li>– Inability of the SMP model to handle technical operational problems and sudden variation in wastewater quality.</li> </ul>
Silva <i>et al.</i>	SMP	<ul style="list-style-type: none"> <li>– MBR pilot plant under steady state conditions</li> </ul>

1998		<ul style="list-style-type: none"> <li>- Accurate predictions of the concentrations of nitrogen species and sludge as well as of the soluble COD trend in the effluent</li> </ul>
Lu <i>et al.</i> 2001	ASM1-SMP	<ul style="list-style-type: none"> <li>- Completely mixed bioreactor treating synthetic wastewater</li> <li>- Good correspondence for soluble nitrogen and soluble COD concentrations under steady state conditions</li> <li>- Underestimation of MLSS levels</li> </ul>
Liu <i>et al.</i> 2003	Empirical hydrodynamic	<ul style="list-style-type: none"> <li>- Internal loop air-lift reactor</li> <li>- Ability of the model to illustrate hydrodynamic effects on membrane fouling, despite its unsuitability for operational and design purposes.</li> </ul>
Meng <i>et al.</i> 2005	Fractal permeation	<ul style="list-style-type: none"> <li>- Evaluation of permeability of cake formed during the microfiltration of activated sludge</li> <li>- Inability of the model to show how operational conditions and parameters influence cake resistance</li> </ul>
Li & Wang 2008	Sectional resistance	<ul style="list-style-type: none"> <li>- Characterization of membrane fouling in submerged MBR</li> <li>- Ability of the model to account for cleaning cycles and characterization of fouling development over time</li> <li>- Ability of the model to capture general trends only and unsuitability to accurately model membrane fouling phenomena</li> </ul>

In the context of biomass kinetics models, the Activated Sludge Model (ASM) family was first developed to simulate the activated sludge process but their ability to simulate the MBR process has not been adequately validated (Ng & Kim 2007). The ASM family presents several advantages such as the simple presentation in matrix format which permits better understanding of the biological treatment and hence developing efficient experimental design, as well as the incorporation of biological phosphorus removal which is a key parameter in biological treatment (in ASM2 and ASM2d) (Ng & Kim 2007). Further, the Soluble Microbial Product (SMP) model has exhibited satisfactory results in characterizing the biomass in MBR systems with acceptable to high accuracy mainly because of the ability of MBRs to operate at high SRT (Furumai & Rittmann 1992; Urbain *et al.* 1998). The model reportedly exhibited good performance in simulating measured VSS, effluent COD and nitrogen species and addresses the interaction between nitrifying and heterotrophic bacteria. It presents two major advantages over the ASM family through its ability to simulate the biomass in MBRs without calibration using experimental data and its formulation-governing equations require less input parameters. And finally, the ASM1-SMP hybrid system was reportedly able to quantify

COD and soluble nitrogen concentrations with acceptable accuracy; although it has underestimated MLSS levels (Lu *et al.* 2001). In the context of membrane fouling, the fractal permeation and the resistance-in-series models have shown good results although further experimental verification is reportedly still needed (Lee *et al.* 2002). In addition, the resistance-in-series model in particular appears to be most promising because it takes into consideration cleaning cycles and was reported to predict well temporal changes in permeability (Winitgens *et al.* 2003). Other reported efforts include the empirical hydrodynamic model which has been reported to explore the impact of hydrodynamics conditions on the membrane fouling rate and the mixed liquor cross-flow velocity, however it is reportedly too simple to describe and predict adequately the complex phenomenon of fouling in MBRs and the sectional resistance model that was equally reported to lack accuracy (Ng & Kim 2007).

### **A.3. MBR FOR LANDFILL LEACHATE TREATMENT**

#### **A.1.1 Performance evaluation**

Much of the reported literature on the application of MBR technology for landfill leachate treatment focus on the performance of the MBR in removing Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD) and ammonia under various operating conditions. In general, the MBR technology offers the flexibility of a stable performance despite the wide variation in leachate quality and quantity (Chen & Liu 2006; (Laitinen *et al.* 2006; Sadri *et al.* 2008; Hua *et al.* 2009; Akkaya *et al.* 2010). While many studies adopted pretreatment and/or post treatment, table A-2 presents removal efficiencies pertaining to the MBR alone. In general, high BOD removals (90-99%) from the leachate were attained with MBR treatment irrespective of experimental conditions and leachate maturity, with the exception of one recent study that reported 75-99% (Akgul *et al.* 2013),

although the efficiency was mostly greater than 90%. In contrast, the efficiency in removing COD varied significantly from as low as 23% (Jakopović *et al.* 2008) to as high as 98% (Hua *et al.* 2009). On the other hand, the MBR technology showed more promise and effectiveness in removing nitrogenous compounds than conventional biological systems even under variable influent concentration and operating conditions (Sadri *et al.* 2008; Visvanathan *et al.* 2007). Note that high removal efficiencies (up to 99%) were obtained mostly when the influent ammonia concentrations were low because high levels of nitrogen (> 1,000 mg/L as ammonium or organic nitrogen) result in the inhibition of nitrobacter and nitrosomonas species (Ince *et al.* 2013; Ahn *et al.* 2002), thus necessitating pre-treatment such as ammonia-stripping (Hasar *et al.* 2009 b; An *et al.* 2006; Wichitsathian *et al.* 2004)



Table A-2: Performance of MBR in treating landfill leachate

Reference	Location	Scale	Process	Membrane configuration	Influent characteristics			Operational conditions		Removal efficiency		
					COD, mg/L	BOD/COD (age)	NH <sub>3</sub> , mg/L	HRT days	SRT days	COD %	BOD %	NH <sub>3</sub> %
Akgul <i>et al.</i> 2013 <sup>c</sup>	Turkey	Lab	UASB+MBR+SHARON+Anammox	(Tub)	28,000–37,000 1500–2000 <sup>a</sup>	0.7–0.37 Y, Y+O	250–2500		5 d	30–85	75–99	
Brown <i>et al.</i> 2013	Canada	Lab	MBR (compost leachate)	Sub (HF)	116,000		2720	95	-	99.7		≈100
Campagna <i>et al.</i> 2013	Turkey	Full	MBR+NF	Ext (UF)	16360		>400	-	-	84.4		88 <sup>d</sup>
Ince <i>et al.</i> 2013	Turkey		Jet loop MBR	Ext (Tub)	13225	0.44 (Y)	2200–2500	1.35 and 2.93	2.16 and 5.33	80–85		3.2–21.3
Insel <i>et al.</i> 2013 <sup>c</sup>	Turkey	Lab	MBR+NF+RO	Sub (UF)	18,685		1245		30	89		83.5
Sanguanpak <i>et al.</i> 2013	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9240	0.629 (Y)	--	1	Inf.	87	99	
Thanh <i>et al.</i> 2013	Vietnam		MBR	Sub	1200–1400 <sup>a</sup>		68 ± 26	3.5–14.6 h	30	Up to 97.5		≤92.0 ± 1.5
Zhang <i>et al.</i> 2013 a <sup>c</sup>	China	Lab	Fenton oxidation+MBR+RO	Sub(HF)	1200–1600 <sup>a</sup>	0.09–0.12	550–725	4	45	83–87.5		72–95
Boonyaraj <i>et al.</i> 2012 a	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9,389	0.746	105–174	1	Inf.	87	97	83–91
Boonyaraj <i>et al.</i> 2012 b	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9306	0.72 (Y+O)	138	1	Inf.	87	97	90
Coban <i>et al.</i> 2012	Turkey	Full	Ammonia stripping+MBR+NF	Ext (UF)	24,000	0.33	2313 (24000 cod)	-	-	93.75		98
Hua & Zhang 2012 <sup>c</sup>	China	Full	MBR+NF+RO and MBR+NF+NF	Ext (UF)	30,000	0.5 (Y)	2200 (30000)	-	-	≈97	≈99	98.9–99.6
Litas <i>et al.</i> 2012	Greece	Pilot	SMBR (SBR) Mixture of LFL+Synthetic WW 1:1	Sub (FS)	1772	(O)	269	9	-	95		98.2–99.2
Mahmoudkhani <i>et al.</i> 2012 <sup>c</sup>	Iran	Lab	MBR+RO	Sub (HF)	68250±8000	0.65	1470	15	55	97	99	99.45
Lv <i>et al.</i> 2012	China	Pilot	MBR	Sub (HF)	3600–9700	0.31–0.65	200–620 <sup>a</sup>	4		95	100	61.7
Santinelli <i>et al.</i> 2012	Italy	Pilot	MBR	Sub (HF)	802		--	-	-			
Bai <i>et al.</i> 2011	China	Lab	Anoxic-oxic hybrid MBR Diluted leachate	Ext	500–4500	-	150–1400	-	-	Up to 90		≤60
Chiemchaisri <i>et al.</i> 2011	Thailand	Pilot	2-stage MBR (anoxic tank+ aerobic MBR)	Sub(HF)	2605–7318	(O+Y) mixed feed	218–1750	0.5 (MBR tank)	-	60–78	99	80–97
Akkaya <i>et al.</i> 2010 <sup>c</sup>	Turkey	Lab	UASB+MBR+MAP	Sub	4250 <sup>a</sup>	(M)	2315.4 <sup>a</sup>	-	-	10–70		35
Trzcinski <i>et al.</i> 2010	UK	Lab	3-stage (HR-SAMBR-MBR)	Sub (FS)	150–1300		--	Variabl e	300	30–90		
Li <i>et al.</i> 2010 <sup>c</sup>	China	Pilot	Anaerobic pretreatment/air-lift bioreactor	Ext(UF/Tub)	4670–6700	(Y)	820–960	-	-	87		100
Puszczalo <i>et al.</i> 2010 <sup>c</sup>	Poland	Lab	Mixture of 10% LFL+ synthetic WW/SBR	Sub (MF/Cap)	3000–3500	0.06(O)	950–1550	2–3	15	89	>98	>95
Aloui <i>et al.</i> 2009 <sup>c</sup>	Tunisia	Lab	Stirred tank reactor	Ext (MF/Tub)	7100–8000	0.18(O)	1000–2800	2–3	-	70–77	>90	≈90
Feki <i>et al.</i> 2009 <sup>c</sup>	Tunisia	Lab	MBR/electrochemical oxidation	Ext(Tub)	6500–8000	0.09 (O)	1500	-	-	61	100	72.8
Hasar <i>et al.</i> 2009 a	Turkey	Bench	Mixture of LFL+ domestic WW	Sub (HF)	8500–14200 +750–2400 <sup>a</sup>	0.4–0.67 (Y)	1100–2150	3.6–6.0 h	5–30	72–99	-	
Hasar <i>et al.</i> 2009 b <sup>c</sup>	Turkey	Lab	Ammonia stripping+ coagulation/ flocculation pretreatment + aer/an-MBR+RO	Sub (HF)	8500–19200 ~7300 <sup>a</sup>	0.4–0.7 (Y)	200–1000 <sup>a</sup>	3.6–16.4 h	10–50	60–90		87–98
Hua <i>et al.</i> 2009	China	Pilot	UASB/MBR	Sub(HF)	40,000–75,000 1440–25600 <sup>a</sup>	0.42–0.52 (Y)	380–1800	0.5	14	98	99	≈100
Ratanatamskul, & Nilthong, 2009	Thailand	Lab	BPAC-MBR	Sub (HF)	5000–6000 1000 <sup>a</sup>	~0.1 (O)	--	1	Inf.	83		
Svojitzka <i>et al.</i> 2009	Germany	Bench	Compartmentalized activated sludge tank	Ext (UF/Tub)	2200	<0.05	1200	70–170 h	100	≈30	91	90–99
Jakopović <i>et al.</i> 2008 <sup>c</sup>	Croatia	Pilot	Stirred tank reactor	Sub (HF)	1400–2800	0.46	--	8 h	-	23		
Sadri <i>et al.</i>	Canada	Lab	Stirred tank reactor	Sub (HF)	2737–4079	0.11–0.18	662±176	1–3.5	30, 60	54–	>97	>99

Reference	Location	Scale	Process	Membrane configuration	Influent characteristics			Operational conditions		Removal efficiency		
					COD, mg/L	BOD/COD (age)	NH <sub>3</sub> , mg/L	HRT days	SRT days	COD %	BOD %	NH <sub>3</sub> %
2008						(O)				78		
Tsilogeorgis <i>et al.</i> 2008	Greece	Bench	MSBR	Sub (UF/HF)	1391–3977	(O)	200–279	10	infinite	40–60		≈100
Xu <i>et al.</i> 2008	China	Pilot	Combined anaerobic pre-treatment and MBR	Air-lift Ext (UF/HF)	10,084 9357 <sup>a-b</sup>	0.71 (Y)	--	9.5	-	89	>99	
Judd <i>et al.</i> 2006	UK	Lab	MBR	Ext (Tub)	2701	(O)	21.77–588	5	30			73–99
Robinson <i>et al.</i> 2007 <sup>c</sup>	UK	Full	3 aerobic biological tanks in series	Ext (UF/Tub)	5000	0.05	2000	-	-	76	>96	≈100
Sang <i>et al.</i> 2007	Vietnam	Lab	Stirred tank reactor	Sub (MF)	4000–39,600	>0.68 (Y)	--	50 – infinite	-	84–97		
Visvanathan <i>et al.</i> 2007	Thailand	Lab	Thermophilic MBR	Sub	12000±1000	0.39–0.65 (M)	1000–1700	1	-	62–79	>97	60–75
An <i>et al.</i> 2006	China	Pilot	Anoxic +aerobic zone in tank	Sub (MF)	1500	-	500	8.5	-	75	-	80–99
Bodzek <i>et al.</i> 2006	Poland	Lab	Mixture of 10% LFL+synthetic WW	Ext(UF/Tub)	442 <sup>a</sup>	-	390 <sup>a</sup>	-	-	82.4	98.3	62.8
Canziani <i>et al.</i> 2006	Italy	Pilot	MBR+MBBR	Sub (Tub)	6,316	0.3 (O)	1000–1500	-	>45	Up to 75		≥90
Chen & Liu 2006	China	Pilot	Air-lift bioreactor	Air-lift Ext (UF/HF)	4200–15900 <sup>b</sup>	-	--	1.8–12.9	-	70–96	>99	
Laitinen <i>et al.</i> 2006	finnish	Pilot	Dual-tank MBR (SBR +MBR)	Sub (HF)	2200±230	(Y)	210±90	2–5	35–60	>80	>97	>97
Chaturapruek <i>et al.</i> 2005 <sup>c</sup>	Thailand	Lab	Ammonia stripping pretreatment/stirred tank reactor	Sub (HF)	8000–9000	0.40–0.45 (M)	1700–1800	1	-	~70	~>95	
Schwarzenbeck <i>et al.</i> 2004 <sup>d</sup>	Germany	Full	2 reactors in series (denitrification+ nitrification)+AC filter	MF	136–1980	~0.2	120	-	-	65	95	97
Wichitsathian <i>et al.</i> 2004	Thailand	Lab	Ammonia stripping /stirred tank reactor	Sub (MF/HF)	8000±1000	0.4±0.05 (M)	1700±100	0.66–1	-	60–66 72–76 b	94–98 b	
Setiadi & Fairus 2003	Indonesia	Lab	Stirred tank reactor	Ext(MF/HF)	1800	0.15–0.17	114.8	1	32	31.3	98	66
Ahn <i>et al.</i> 2002 <sup>c</sup>	South korea	Full	Aeration basin with anoxic +aerobic parts	Sub(MF/HF)	400–1500	(O)	200–1400	-	-	~38	97	

Cap: Capillary; Ext: External; FS: Flat Sheet; HF: Hollow Fiber; M: Medium; MF: Microfiltration; O: Old; Sub: Submerged; Tub: Tubular; UF: Ultrafiltration; Y: Young

<sup>a</sup> Concentrations after pretreatment or dilution.

<sup>b</sup> COD values in terms of the soluble COD.

<sup>c</sup> Applied post-treatment to MBR (efficiencies are for MBR only)

<sup>d</sup> Combined efficiency for primary clarifier + MBR

### A.3.2 Removal of micropollutants and metals

Municipal Solid Waste (MSW) invariably contains hazardous substances resulting from biochemical reactions within the landfill (Banar *et al.* 2006) and the uncontrolled discharge of household and industrial chemicals that contributes to leachate toxicity (Slack *et al.* 2005; Kjeldsen *et al.* 2002) and represents a serious threat to receiving water bodies (Baun *et al.* 2004; Alkalay *et al.* 1998). However, since micropollutants contribute little to the levels of Total Organic Carbon (TOC) and COD in leachate, the removal of TOC and COD may not necessarily imply a significant decrease in the levels of trace contaminants (Xu *et al.* 2008). In this context, MBR systems present the advantage of improving the decomposition of these micro-pollutants due to their ability of maintaining high sludge concentrations resulting in high removal efficiencies (Boonyaroj *et al.* 20012b) (Table A-3).

Table A-3: Performance of MBRs in removing micropollutants and metals

Reference	Micropollutants /Metals	Influent concentration, µg/L	% Removal
Wintgens <i>et al.</i> 2003 a	Bisphenol A (BPA)	≈600-7000	95
	Nonylphenol (NP)	≈9-300	85
Xu <i>et al.</i> 2008	Σ OrganoChlorine Pesticides	203.5 µg/L	6
	Σ Polycyclic Aromatic Hydrocarbons	485.2-1,188.2	1
	Σ Technical 4-NP	92-482	98
Svojitka <i>et al.</i> 2009	BPA	2100 µg/L	99.99
Matošić <i>et al.</i> 2008	Diacetone sorbose	1,420-2,570	30
	Diacetone alpha-ketogulonic acid	80-430	69
	Propyphenazone	85-130	16
Boonyaroj <i>et al.</i> 2012 a	PAHs, phenolic compounds and phtalic acid esters	16.1-1,1412	50-76
Santinelli <i>et al.</i> 2012	Chromium	136	93
	Nickel	48.6	58
Fatone <i>et al.</i> 2008	Chromium	12-41	Up to 50
	Copper	8-44	Up to 61
	Nickel	21-65	Up to 22
Brown <i>et al.</i> 2013	Aluminum	39,800	99.93
	Arsenic	634	97
	Barium	1190	98.99
	Boron	4890	82.74
	Chromium	401	98.75
	Cobalt	220	97.27
	Iron	50	99.87
	Lead	811	99.9

Manganese	51,000	99.95
Molybdenum	147	93.2
Nickel	589	88.79
Strontium	27,100	89.19
Titanium	1370	99.49
Vanadium	136	98.53
Zinc	13,500	99.5

### A.3.3 Effect of HRT and SRT

The HRT is a key parameter affecting the size and performance of the MBR (Ren *et al.* 2005; He *et al.* 2005). While the COD removal was reported to increase with increasing BOD/COD ratio in conventional systems, the effect of HRT was less evident. For younger leachate with BOD/COD ratios ranging between 0.4 and 0.8, the COD removal increased with increasing HRT. For older leachate, HRT had no considerable impact on COD removal, even at high HRTs (24-45 days). Sadri *et al.* (2008) tested the aerobic MBR under various organic loading rates and at different HRT and SRT combinations. They reported that although the membrane required more frequent cleaning when decreasing the HRT, the effectiveness in Total Suspended Solids (TSS) and VSS removal was not affected. Further, they observed no considerable changes in the removal of metals, BOD and NH<sub>3</sub> at different SRT-HRT combinations. However, the toxicity removal decreased from 100% to 75% when the HRT decreased from 3.5 into 2 days. It is worth noting that reported HRT in MBR treating landfill leachate ranged from as low as 3.5 h to as high as 95 days; however most studies adopted an HRT between 12h and 3 days which is lower than HRTs adopted in conventional aerobic biological systems also treating landfill leachate (1-20 days) (Kurniawan *et al.* 2010).

Similarly the SRT is another key parameter that affects the performance of MBRs (Massé *et al.* 2006; Han *et al.* 2005). In fact, one of the main advantages of MBRs over CAS is the decoupling of the HRT and SRT, which permits operations in small volume reactors under

shorter HRT (Ahmed & Lan 2012). A longer SRT offers sufficient time for the development of slow-growing bacteria and permits the growth of specialized microbial species necessary for the decomposition of slowly-biodegradable compounds (Matošić *et al.* 2008; Setiadi & Fairus 2003; Hasar *et al.* 2009 b). Note however that excessively high SRTs might negatively affect the performance of MBR systems whereby COD removal reportedly decreased when increasing the SRT to 50 days (Hasar *et al.* 2009 b) while low COD and phosphate removals were reported when operating at infinite SRT (Tsilogeorgis *et al.* 2008) which might be due to the accumulation of inert matter in the MBR resulting in lower specific biomass activity (Pollice *et al.* 2008 a, b; Li *et al.* 2006).

#### **A.3.4 Mixing domestic wastewater and leachate**

Much work examined the suitability and viability of the treatment of leachate mixed with municipal WW using MBR. While concerns have been reported about potential biological inhibition (i.e. heavy metals) and overloading (i.e. high COD, TN and BOD) (Çeçen & Aktaş 2004), several studies demonstrated that MBR systems were attractive options in areas that required the co-treatment of leachate and sewage (Bodzek *et al.* 2006; Bohdziewicz *et al.* 2008; Hasar *et al.* 2009 a; Puszczalo *et al.* 2010; Litas *et al.* 2012).

#### **A.3.5 Pre treatment and Post-treatment**

Gel permeation chromatography has shown that the organic matter in leachate is distributed in different fractions of high and low molecular weight depending on the age of the leachate. The high MW fraction (MW of the peak (MWp) = 11480-13182 Da) has a low biodegradability but can be converted into low MW compounds with high biodegradability using ozonation (Chaturapruek *et al.* 2005; Jakopović *et al.* 2008). On the other hand, while the low MW fraction (MWp = 158-275 Da) is mostly biodegradable, yet its residue could

permeate through the membrane resulting in high soluble COD (sCOD) in the MBR effluent (Chen & Liu 2006). This necessitates the adoption of a pre-treatment and/or post-treatment in MBR systems to ensure good effluent quality, especially with increasingly stringent discharge standards. In this context, various technologies have been explored with different levels of success as a polishing treatment for leachate effluent from MBR systems such as *Nanofiltration* (Insel *et al.* 2013, Li *et al.* 2010, Ince *et al.* 2010, Jakopović *et al.* 2008; Robinson 2007; Hua & Zhang 2012), *Microfiltration* (Ince *et al.* 2010), *Electrochemical Oxidation* (Feki *et al.* 2009; Aloui *et al.* 2009), *Reverse Osmosis* (Ahn *et al.* 2002; Bohdziewicz *et al.* 2008; Hasar *et al.* 2009 b; Mahmoudkhani *et al.* 2012; Zhang *et al.* 2013 a, Insel *et al.* 2013; Hua & Zhang 2012; Puszczalo *et al.* 2010). *PAC*, *GAC*, *FeCL3* with and without *polyelectrolyte*, and *polymeric adsorbents* (e.g. XAD7HP and XAD4) (Trzcinski *et al.* 2011); *Zeravalent Iron (ZVI)* (Zhang *et al.* 2013 b), *Coagulation-Flocculation* (Hasar *et al.* 2009 b), *Ammonia Stripping* (Bohdziewicz *et al.* 2008; Hasar *et al.* 2009 b), *Fenton Oxidation* (Zhang *et al.* 2013 a); *precipitation with Magnesium Ammonium Phosphate (MAP)* (Akkaya *et al.* 2010), *SHARON and Anammox* (Akgul *et al.* 2013). Note that the MBR itself was examined as a post-treatment for the effluent of UASB treating medium aged leachate which reduced the variation in UASB effluent quality (Akkaya *et al.* 2010).

#### **A.4. VARIATIONS IN MBR TECHNOLOGY**

Variations in the MBR system have been explored including Anaerobic MBR (anMBR), Jet-loop MBRs, and biological powdered activated carbon (BPAC)-MBR. Although anaerobic digestion systems present some advantages in terms of low sludge yield and production of methane (Farhadian *et al.* 2007), their utilization has been limited by the slow-growing methanogenic bacteria whose doubling time might take from 12 hours to as high as 1 week. However, since the MBR allows for better retention of biomass, it results in

greater COD removal efficiency for less biodegradable influent (Zhang *et al.* 2007). Hence several studies examined the anMBR performance for the treatment of leachate or leachate-WW mixture under various operating conditions (i.e. SRT, HRT, temperature, OLR, flux...) (Taskan & Hasar 2012; Trzcinski *et al.* 2011; Zayen *et al.* 2010; Trzcinski & Stuckey 2009 a, b; Bohdziewicz *et al.* 2008; Trzcinski & Stuckey 2010) while others investigated the accompanying microbial populations (Trzcinski *et al.* 2010; Trzcinski & Stuckey 2012). Since anMBR are more prone to fouling than aerobic membranes (Saddoud & Sayadi 2007), the flux decline during operation (Taskan & Hasar 2012) in addition to the need for frequent cleaning were reportedly the most limiting factors in the application of anMBRs. However, anaerobic dynamic MBR which relies on the formation of a cake layer on the mesh or fabric supporting material for the separation of solids from the liquid (Ersahin *et al.* 2012), offers the advantage of lower cost and high membrane flux but its application for the treatment of leachate remains very limited (Xie *et al.* 2014). Under thermophilic conditions, the use of membranes to separate microorganisms from the treated effluent permitted the preservation of the advantages of thermophilic treatment (low sludge yield, rapid biodegradation, rapid inactivation of pathogenic microorganisms, high loading rates, low retention time) without compromising the effluent quality due to poor settling properties resulting from high temperatures (Krishna & Van Loosdrecht 1999). However, thermophilic MBRs have shown a higher level of fouling due to elevated concentration of EPS when compared to mesophilic MBRs probably because thermophilic sludge contains higher levels of small particles than mesophilic sludge (Visvanathan *et al.* 2007).

In a jet loop reactor, dispersion is performed by a liquid jet drive (Farizoglu & Keskinler 2006; Dirix & Wiele 1990; Dutta & Raghavan 1987) whereby liquid is injected at high velocity to cause a fine dispersion of liquid and gaseous phases. The formation of bubbles permits better oxygen transfer to the microorganisms and grants homogeneous biomass

dispersion (Ince *et al.* 2013). As such up to 85% COD removal efficiency was achieved using a jet-loop MBRs for the treatment of LFL (Ince *et al.* 2013).

Finally, the BPAC-MBR was explored for the treatment of leachate and achieved 83, 85, 97 and 68% removal efficiency of COD, color, Total Kjeldahl Nitrogen (TKN) and Total Phosphorus (TP) respectively (Ratanatamskul & Nilthong 2009).

These new aspects of MBR application for leachate treatment need to be further investigated because of their advantages over their conventional MBR counterparts. For example the anaerobic treatment requires no aeration energy and produces biogas (methane) that permits the conversion of bio-energy in leachate into valuable renewable energy. Further, the aerobic thermophilic treatment offers several advantages such as: low sludge yield, rapid biodegradation, rapid inactivation of pathogenic microorganisms and high loading rates, thus decreasing the retention time and the associated capital cost. The jet-loop systems require less space and offer the flexibility in treating high organic load wastewaters. Finally, the PAC addition has been shown to alleviate membrane fouling and lead to improved COD removal due to coincident biodegradation and adsorption, namely of biologically recalcitrant compounds.

#### **A.5. EXISTING GAPS AND FUTURE NEEDS**

While the MBR technology has shown promising results in treating landfill leachate, much remains to be desired in terms of better understanding its performance towards achieving high removal efficiency at the lowest possible cost which requires careful selection of parameters that ensure optimum performance (Figure A-3).



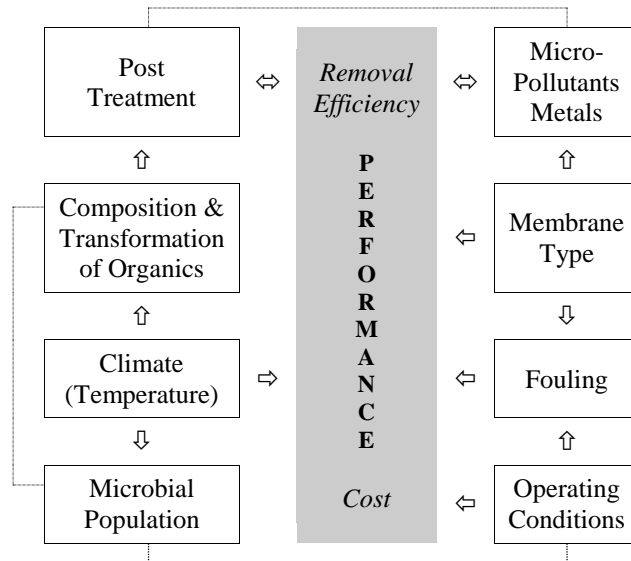


Figure A-3: Factors affecting the performance in MBR systems

In this context, *membrane fouling* remains the most challenging factor that might impede the development of the MBR technology, especially in treating high strength leachate. While appreciable efforts have been dedicated to fouling in municipal wastewater treatment systems, limited work targeted landfill leachate. It is also critical to define optimum *operating conditions* (HRT, SRT, aeration pattern and frequency, leachate circulation pattern, filtration modes...) as well as *membrane type and material* that result in minimum fouling and hence reduce associated maintenance and operating costs. Such knowledge will enhance the optimization of the design and operation of MBR systems.

Equally important is a need to examine leachate at a micro-prescriptive rather than only from a macro-perspective as generally expressed in terms of COD, BOD, NH<sub>3</sub> and TOC. Characterization of the *composition and fate of organic matters* in leachate during the treatment process especially when *post-treatment*, such as NF and RO, is required, because the occurrence of certain polymeric substances, organics and trace materials might lead to biofouling of NF/RO membranes and hence adversely affects the removal efficiency (Insel *et al.* 2013). Similarly, *micropollutants* resulting from biochemical reactions (Banar *et al.*

2006), represent a serious threat to receiving water bodies (Baun *et al.* 2004). However, since these micropollutants contribute little to TOC and COD in LFL, the removal of the latter may not imply a significant decrease in trace contaminants and hence the need to better characterize their occurrence and removal efficiencies.

While most studies were reportedly conducted at moderate temperatures, a gap exists in exploring the suitability of the MBR technology to treat leachate under wider *climatic conditions* that may affect *microbial populations*, biochemical reactions and *transformation of organics*, as well as *membrane fouling* often necessitating the adoption of cooling/heating elements thus adding to the overall *energy costs* of the system.

The understanding of microbial dynamics within MBRs remains in its early development with more efforts needed to delineate the effect of SRT, temperature, leachate recirculation (ratio and configuration), aeration pattern and frequency (continuous vs. intermittent) on microbial populations including optimum conditions for the enrichment of selected microorganisms targeting the biodegradation of specific micropollutants and xenobiotics. This latter aspect is imperative when handling high strength wastewater such as leachate, which might contain toxicants that require enrichment of certain types of bacteria targeting those toxicants.

On the other hand, although it is commonly perceived as an expensive technology, the overall *economics* of MBRs may indeed be more attractive because pre- and post-treatment requirements are reduced particularly in the context of stabilized LFL. Limited to no work has been reported in this context and hence the overall cost of MBR systems including polishing steps and methods to reduce associated energy costs, needs further examination.

Last but not least, scaling up from laboratory studies to field scale requires particular attention and testing to ensure long-term operation under variable loading conditions. While promising results have been obtained in this regard from limited previous studies, it remains risky to extrapolate results particularly because of the fluctuating properties of leachate in quantity and quality.

Systematic studies targeting the above constraints are prerequisites for enhancing the development and widespread of the MBR technology. In this context, a framework is depicted in Figure A-4 consisting of an integrated experimental and modeling approach with the objectives and methods outlined in Table A-4

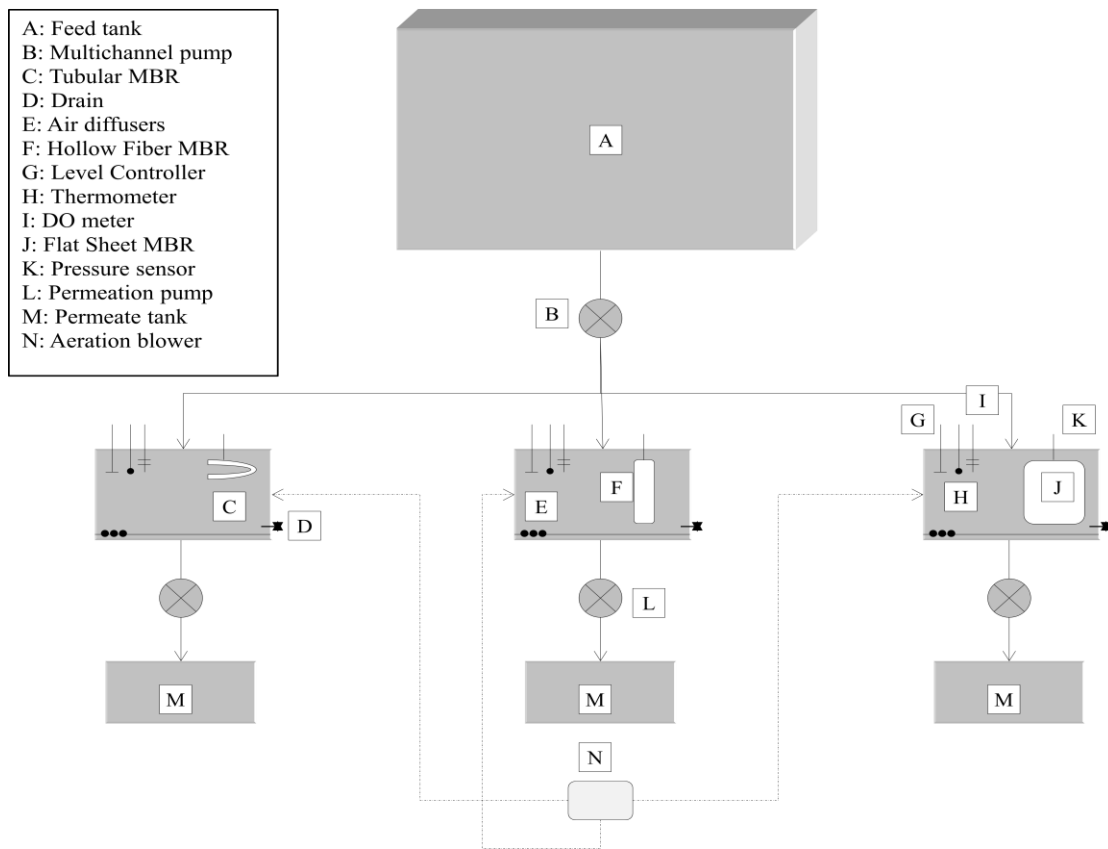


Figure A-4: Proposed experimental setup

Table A-4: Suggested experimental program

Objective / Outcome	Methods	Justification	Constraints/Challenges
Start-up and steady state operation of multiple MBR systems with different membrane types	<ul style="list-style-type: none"> <li>Collect leachate from a landfill and store at 4°C for future use to reduce fluctuations in the influent</li> <li>Equip reactors with different membrane types (i.e. tubular, hollow fiber, and flat sheet) and proper instrumentation (pressure sensors, thermostats, influent flow meters, aeration rotameters, level control, and a data acquisition system)</li> <li>Assure hydraulic performance. initiate leachate feeding, and inoculate reactors with activated sludge from a wastewater or leachate treatment plant.</li> <li>Test under various operating conditions: SRT (5 to 60 days), aeration pattern (continuous vs. intermittent, and temperature (10 to 45°C).</li> <li>Replace membrane modules prior to the initiation of each SRT if deemed necessary.</li> </ul>	Limited comparative assessments on the impact of membrane type and operating conditions on the performance of MBRs in treating landfill leachate	<ul style="list-style-type: none"> <li>Resources (budget and time)</li> <li>Foaming especially during the start up phase: consider anti-foaming agent such as Sigma A6426</li> <li>Membrane fouling: consider air backpulsing</li> </ul>
Treatment performance as a function of membrane types and varied operating conditions	Monitor periodically (three times per week) indicators of interest Dissolve Oxygen (DO), pH, COD, BOD, alkalinity, phosphorus and nitrogen compounds, Dissolved Organic Carbon (DOC) in addition to heavy metals and micropollutants (such as BPA, NP, OCP, PAHs, phenolic compounds, phthalates...) at several locations in the system.	Limited studies targeted the removal of metals and micropollutants under variable operating conditions with different membrane types	Advanced and relatively expensive analytical equipment for the analysis of metals and micro-pollutants
Correlation between membrane type, operating conditions and fouling propensity	<ul style="list-style-type: none"> <li>Monitor carbohydrate and protein fractions of bound and soluble EPS (i.e. SMP) in aerobic tanks</li> <li>Evaluate the resistance across membranes according the procedure presented by Meng <i>et al.</i> 2008 as a function of organic Loading rate (Trussell <i>et al.</i> 2006)</li> </ul>	While much work have been reported on membrane fouling, most efforts targeted municipal wastewater and not leachate. Irrespective fouling processes are still not adequately understood and remain the main impediment for MBR systems.	Resources (budget and time)
Definition of Molecular weight (MW) distribution	Carry MW distribution of influent and effluent leachate with sequential filtration using UF membranes with variable diameters (1 to 500 um). Particle size and MW distribution of organic matter are related to their biodegradability and helps in evaluating the appropriate treatment technology.	Provides micro-perspective insight on leachate and contributes to scarce knowledge on the composition and fate of organic matters during treatment.	Resources (budget and time)
Correlation between biological components/processes and system performance under various operating conditions to shed light on observed behavior and validate developed relationships	Collect mixed liquor samples (weekly) from the aerobic tanks and biofilm samples from the membrane surfaces (bi-weekly) for microbial analysis (16S rRNA gene pyrosequencing; quantitative polymerase chain reaction) and Scanning Electron Microscopy (SEM) to define the need for bioaugmentation of certain bacteria and monitor its effect on the removal efficiency of micropollutants and the overall system performance	There is a lack of fundamental information on microbial communities in MBR systems treating leachate as well as the effect of operating conditions and membrane type on the dynamics of these communities.	Besides resources (budget and time), the selection of candidate bacteria in a bioaugmentation process is complex and may affect to original microbial communities

### **A.1.1 MBR modeling**

The modeling of MBR systems is increasingly becoming critical for its performance prediction and system design analysis for the development of the technology and its widespread use. In this context, while the ASM family has reportedly been appropriate for the characterization of biomass kinetics in MBR systems, much remains to be desired to examine the suitability of ASMs for modeling MBR systems particularly in the context of 1) assessing the effect of higher MLSS levels and SRTs on biomass; and 2) understanding the difference between the CAS and MBR process. Although the SMP model (Urbain *et al.* 1998) is equally promising to shed more light on the effects of filtration when compared to clarification under both steady and transient conditions, it lacks a user-friendly interface and does not simulate biological phosphorus removal. While an attempt at integrating SMP within the ASM1 to simulate MBRs (Lu *et al.* 2001), more efforts in this hybrid direction can prove useful in understanding the MBR technology.

In the context of fouling, the empirical model reported by Liu *et al.* (2003), although relatively simple to apply, could not simulate the complex membrane surface processes under varied operating conditions and failed to reproduce experimental data during the calibration of the membrane fouling rate. On the other hand, the application of the Fractal permeation model (based on Darcy's law and fractal theory) to determine the permeability of the cake on membrane surfaces failed to simulate the impact of varied operating conditions on the cake resistance (Meng *et al.* 2005). Similarly, the sectional resistance

model that considers the contribution of uneven cake formation<sup>1</sup> was equally non-suitable to simulate membrane fouling although it was reportedly able to delineate general trends (Li & Wang 2006). Finally, the integration of ASM1-SMP hybrid and resistance-in-series (Lee *et al.* 2002), could not be validated and its applicability remains unknown, while the integration of ASM3 and resistance-in-series (Wintgen *et al.* 2003 b) was unable to simulate temporal permeability consistently (Geissler *et al.* 2005).

An integrated framework combining the experimental program outlined above with a modeling program (Figure A-5) is needed to enhance the prediction of the MBR performance and system design analysis for the development of the technology and its widespread use. Such a framework should encompass key processes of the ASM family (such as the growth and decay of heterotrophs and autotrophs) which could be derived from the *Composition & Transformation of Organics* as well as the analysis of *Microbial Population* characterization. The inclusion of a biological algorithm targeting the transformation and fate of SMP and EPS is vital because of their contribution to membrane fouling and potential role in the formation of trihalomethanes in subsequent treatment steps. Equally important is the algorithm that simulates the effect of the membrane type on organics removal and the formation of the cake layer and its role in enhancing the treatment performance through biological filtration in addition to its contribution to membrane resistance. The latter could be obtained based on the *correlation between membrane type and fouling* propensity under different *operating conditions* (i.e. T, HRT, SRT, aeration,

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<sup>1</sup> due to coarse bubbles occurrence during the aeration of submerged MBRs contributing to membrane module cleaning by scouring its surface, thus resulting in non-uniform fouling due to the irregular distribution of shear force from aeration.

and loading rate).

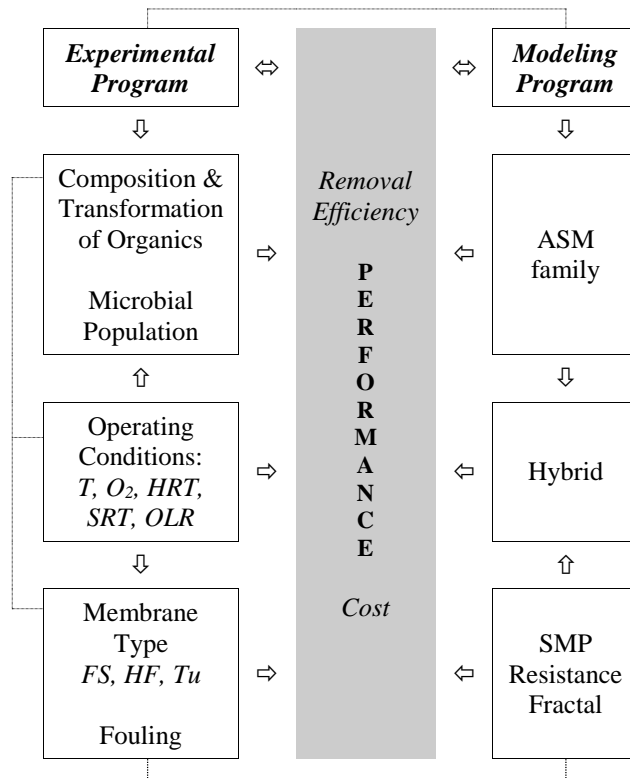


Figure A-5: Framework towards improving the performance and understanding of MBR systems

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# APPENDIX B HOLLOW FIBER VS FLAT SHEET MBR FOR THE TREATMENT OF HIGH STRENGTH STABILIZED LANDFILL LEACHATE

## ABSTRACT

This study presents a performance comparative assessment of flat sheet and hollow fiber membranes in bioreactors for the treatment of relatively stable landfill leachate. For this purpose, a laboratory scale MBR system was constructed and operated to treat a leachate with COD (3,900-7,800 mg/L), BOD<sub>5</sub> (~440-1,537 mg/L), TP (~10-59 mg/L), PO<sub>4</sub><sup>3-</sup> (5-58), TN (1,500-5,200 mg/L), and NH<sub>3</sub> (1,770-4,410 mg/L). Both membranes achieved comparable BOD (92.2 vs. 93.2%) and TP (79.4 vs. 78.5%) removals. However, while slightly higher COD and phosphate removal efficiencies were obtained with the Hollow Fiber membrane (71.4 vs. 68.5% and 87.3 vs. 81.3%, respectively), significantly higher TN and ammonia removal rates (61.2 vs. 49.4 and 63.4 vs. 47.8, respectively) were achieved by the Flat Sheet membrane. The experimental results contribute in filling a gap towards managing stabilized landfill leachate and providing guidelines for corresponding MBR applications.

## KEYWORDS

Membrane Bioreactor, Flat Sheet, Hollow Fiber, stabilized leachate

### A.1. INTRODUCTION

While at the bottom of the desirable hierarchy, landfilling remains an important element of integrated solid waste management because other elements result mostly in waste minimization with residuals inevitably disposed of in landfills, despite the emerging concept of “zero waste”. In many countries, landfills continue to be the most attractive element and often the only one

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. On the other hand, landfill leachate is invariably laden with various contaminants with characteristics dependent on landfill age, precipitation, seasonal weather variation, and waste composition amongst other factors (Renou *et al.* 2008; Kulikowska & Klimiuk 2008). Due to its complex and variable composition, leachate is a difficult wastewater to treat (Tatsi & Zouboulis 2002). Its biodegradability is reduced when the landfill becomes older due to the formation of high molecular weight species (Kang *et al.* 2002; Lopez *et al.* 2004) and the removal of easily biodegraded carbonaceous compounds.

Leachate percolation to underlying aquifers contaminates groundwater and threatens public health (Kurniawan *et al.* 2006; El-Fadel *et al.* 1997). Thus, the proper management and treatment of landfill leachate is increasingly subject to stringent environmental requirements to protect ground and surface water resources (Renou *et al.* 2008). Alternatives for leachate management include discharge into sewer systems for subsequent treatment with municipal wastewater (Çeçen & 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 technologies have been examined for the treatment of leachate encompassing a wide range of biological and physical/chemical processes. While physical/chemical methods are usually adopted as pre/post treatment or to remove a specific pollutant (Renou *et al.* 2008), biological methods which encompass several suspended and attached growth methods under either aerobic or anaerobic conditions, are usually applied to remove the bulk of leachate with high BOD..



More recently, combining membrane separation and biodegradation processes led to the development of the Membrane Bioreactor (MBR) technology (Sutherland 2010) which is increasingly recognized as the process of choice for the treatment of high-strength wastewater containing complex and recalcitrant compounds (Bilad *et al.* 2011). An MBR can be considered as a Conventional Activated Sludge (CAS) system with efficient membrane filtration that holds small pore size of less than 0.1  $\mu\text{m}$  (Santos *et al.* 2010). It replaces the second stage of conventional wastewater treatment (i.e. gravity settling) but produces a better quality effluent while offering a reduced reactor volume and footprint with the possibility of operating at a higher Mixed Liquor Volatile Suspended Solids (MLVSS) ranging between 8,000 and 12,000 mg/L compared to 2,000-3,000 mg/L in a conventional activated sludge system (Sutherland 2010; Cornel & Krause 2006; Alvarez-Vazquez *et al.* 2004).

In the context of leachate treatment, considerably higher BOD removals (90-99%) were attained with MBRs irrespective of experimental conditions and leachate maturity. In contrast, the efficiency of MBR in removing COD varied more widely from as low as 25% (Jakopović *et al.* 2008) to as high as 90% (Chen & Liu 2006; Puszczalo *et al.* 2010; Aloui *et al.* 2009). Some efforts were dedicated to examine the viability of treating leachate with municipal wastewater using MBRs while recognizing potential biological inhibition and plant overloading associated with leachate quality (Çeçen & Aktaş 2004; Bodzek *et al.* 2006; Hasar *et al.* 2009 a, Puszczalo *et al.* 2010; Ahmed & Lan 2012). 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

more satisfactory results in treating old / stabilized landfill leachate (Alvarez-Vazquez et al. 2004).

While the literature on the use of MBR in wastewater treatment is relatively rich, studies examining the impact of various membranes in an MBR system on the treatment efficiency of high strength stabilized landfill leachate are limited. Further, although nutrients are important to control prior to the release of treated wastewater, due to possible eutrophication, the reviewed literature on the treatment of stabilized leachate using MBR has shown scarce data on phosphorus compounds removal using MBR and no data using the Flat sheet MBR. 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 such a leachate with the objective of defining guidelines for a pilot/full scale plant.

## **B.2. MATERIALS AND METHODS**

The experimental setup (Figure B-1) consisted of two Plexiglas denitrification tanks (D) with stirrer mixers (C) to prevent settlement of solids and two aerobic tanks (E, L) also made of Plexiglas with one equipped with a flat sheet membrane (Table B-1) and the other with a hollow fiber membrane (Table B-1). 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) and were connected to a drain (H) to allow sludge wastage and hence control the Solid Retention Time (SRT).

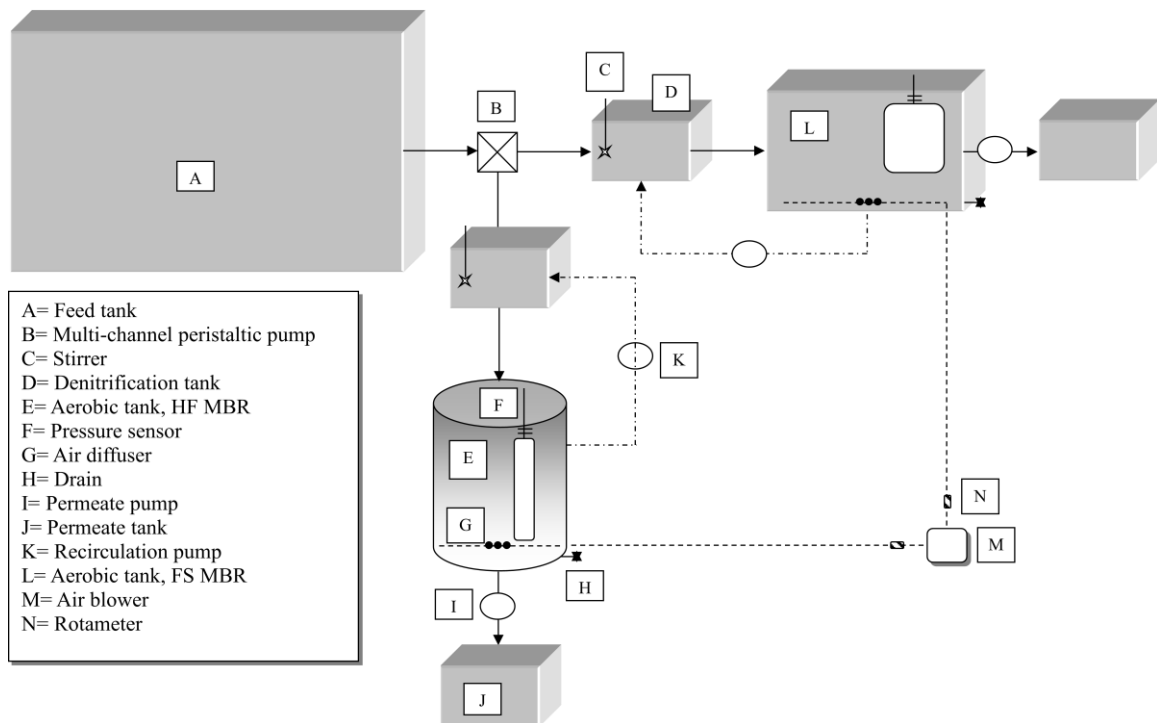


Figure B-1: Experimental setup

Table B-1: Properties of membrane modules used

	<b>Flat Sheet (ACWA)</b>	<b>Hollow Fiber (Zenon)</b>
Model	Kubota 203	ZW 10
Membrane type	Microfiltration	Ultrafiltration
Materials	Chlorinated Polyethylene	Neutral hydrophilic
Manufacturer	Kubota	GE
Nominal Pore size, $\mu\text{m}$	0.2	0.03
Surface area, $\text{m}^2$	0.1	0.93

Leachate was collected weekly from a nearby operating sanitary landfill and transported to the Environmental Engineering Research Center at the American University of Beirut to feed the system. 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 speed ( $\approx 150$  rpm). The flow rate was increased gradually until an HRT of 100 hours (the value was chosen based on previous data from full scale MBR plants treating old landfill leachate (Alvarez-Vazquez *et al.* 2004) and an SRT of 30 days were reached (whereby superior treatment performance has been reported in the literature for 30 day SRTs (Hasar *et al.* 2009 a and b)). The membrane tanks exhibited foaming which was controlled using anti-foaming agent (from Sigma Aldrich) that was added in small quantities as needed (few drops, almost twice per week in the first month then around every two weeks). In addition, the Hollow Fiber membrane was cleaned twice a week using Sodium Hypochlorite (NaOCl) solution while the FS membrane was cleaned by gentle scraping of solids. 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 and analyzed for several indicators including pH, BOD<sub>5</sub> (five day- Biochemical Oxygen Demand), Total Nitrogen (TN), ammonia (NH<sub>3</sub>), COD, TP and phosphate (PO<sub>4</sub><sup>3-</sup>). These indicators were selected following an initial screening of leachate quality that showed them to be most significant and hence they were used as indicators to evaluate the performance of the experimental system in subsequent tests.

### **B.3. RESULTS AND DISCUSSION**

Leachate characterization (Table B-2 B-2) showed high levels of TN (1,500-5,200 mg N/L) and NH<sub>3</sub> (1,770-4,410 mg N/L) together with pH values ranging between 8.08 and 8.87 and a low BOD<sub>5</sub>/COD ratio (0.07-0.22) reflecting a stabilized leachate (Aloui *et al.* 2009; Jakopović *et al.* 2008; Trebouet *et al.* 1999). However, when putting the obtained values in the context of literature, the BOD<sub>5</sub> and COD levels (439.7-1,536.7 mg/L and 3,900-7,800 mg/L) fall within the range reported for stabilized leachate (50 – 4,200 mg/L and 685 – 15,000 mg/L) while the

ammonia and TP levels (1,770-4,410 mg/L and 10.5-59 mg/L) fall within the literature reported range for young leachate (1,400 – 10,250 mg/L and 1.6 – 655 mg/L). This might be due to the fact that the leachate was derived from a landfill in a developing country, Lebanon, where the discharge of high nutrient waste, such as detergents and agricultural wastes, is not controlled while the other studies reported in literature pertain to developed countries such as Greece (Tatsi & Zouboulis 2002; Tsilogeorgis *et al.* 2008) Poland (Puszczalo *et al.* 2010) and Germany (Svojitka *et al.* 2009).

Table B-2: Comparison of leachate quality with literature reported ranges

Paramter	Units	Old leachate*	Fresh leachate*	Range	Average
pH		7.3 – 8.8	4.9 – 6.7	8.08-8.87	8.43
BOD <sub>5</sub>	mg/l	50 – 4200	9500 – 80795	439.7-1,536.7	695
COD	mg/l	685 – 15000	44000 – 115000	3,900-7,800	5978
Ammonia	mg-N/l	39 – 1750	1400 – 10250	1,770-4,410	2,464
TN	mg-N/L			1,500-5,200	254
TP	mg/l	1.27 – 19.9	1.6 – 655	10.5-59	31
PO <sub>4</sub> <sup>3-</sup>	mg/l			5-58	30

\* (Tatsi & Zouboulis 2002)

### B.3.2 Performance Assessment

Temporal variations of tested indicators are presented in **Error! Reference source not found.** and B-3 for the Flat Sheet Hollow Fiber systems, respectively. Table B-3 summarizes the ranges of the chemical analysis results of the influent and effluent of both systems with corresponding removal efficiencies throughout the experimental program.

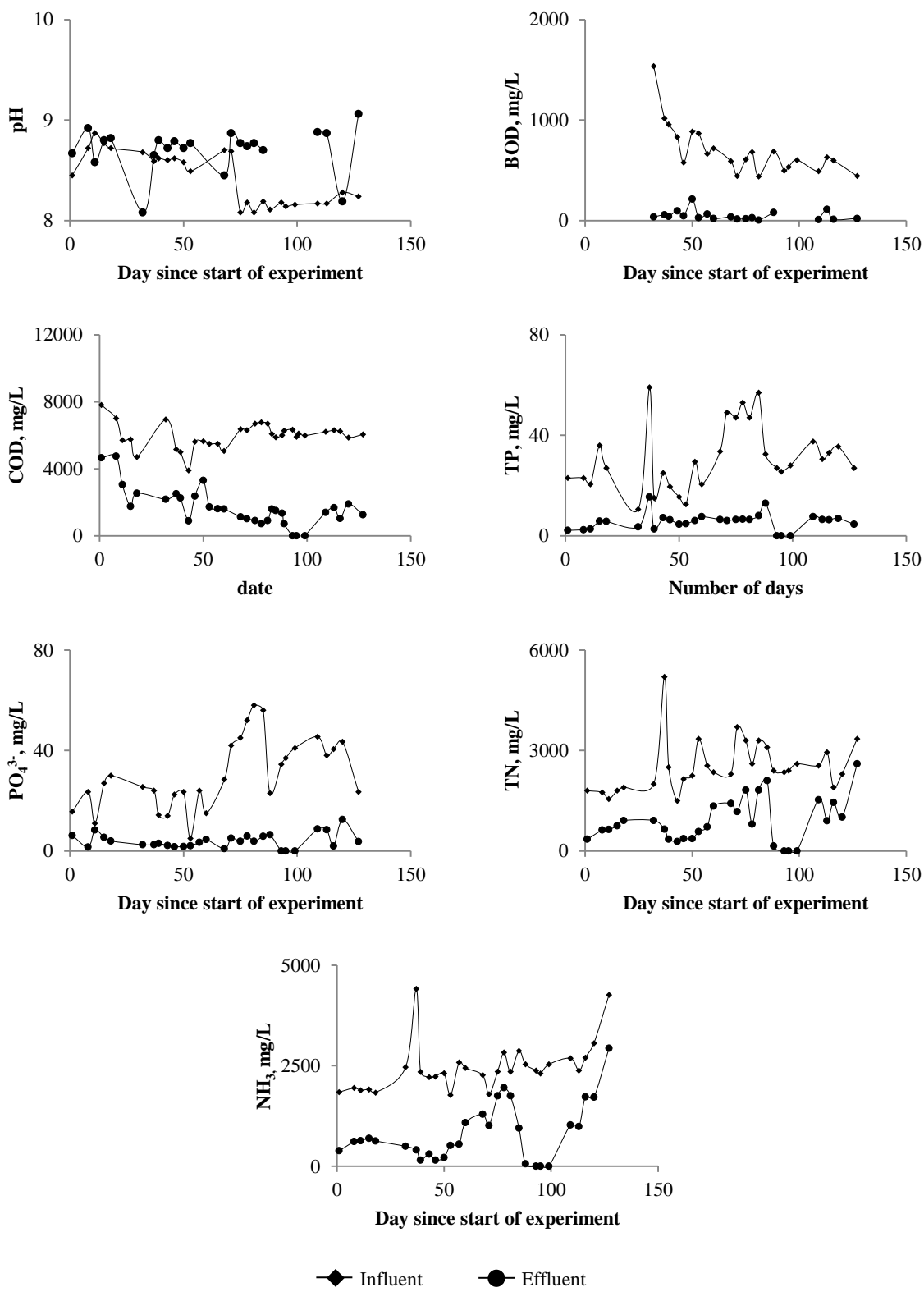


Figure B-2: Temporal variation of main performance indicators: Flat Sheet membrane

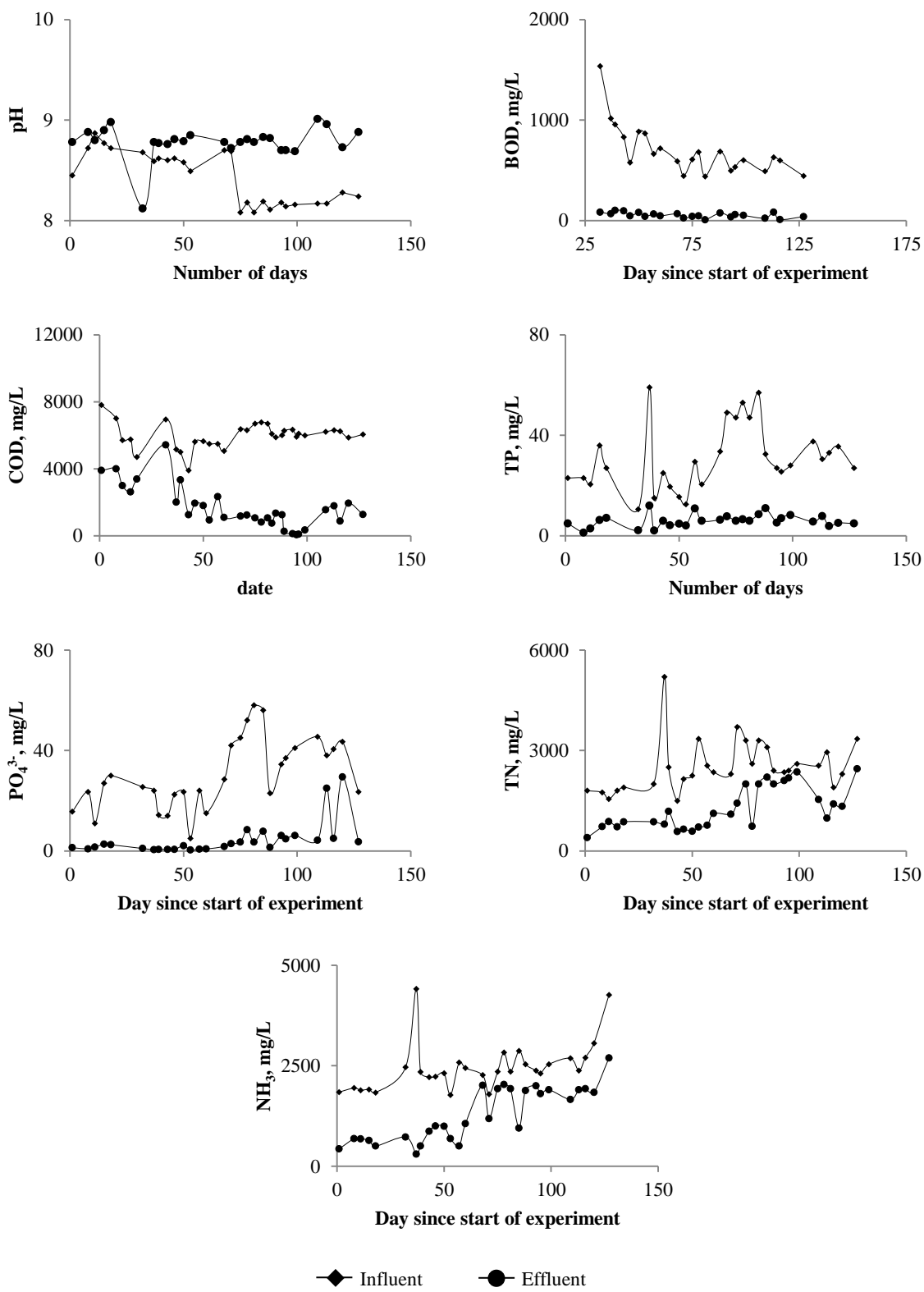


Figure B-3: Temporal variation of main performance indicators: Hollow Fiber membrane

Table B-3: MBR performance assessment

Parameter	Average Influent, mg/L	Average FS_Effluent, mg/L	Average HF_Effluent, mg/L	Average FS Removal Efficiency, %	Average HF Removal Efficiency, %
BOD	695±250.6	50±49.4	55±26.1	93.2±5.9	92.2±3.1
COD	5,978±664.3	1,868±650.7	1,689±1142.5	68.5±13.1	71.4±19.9
TP	31±13	6.3±2.9	6±2.6	78.5±9	79.4±7.8
PO <sub>4</sub> <sup>3-</sup>	30.5±13.8	4.5±2.8	4.5±6.7	81.3±15.6	87.3±15.6
TN	2,543±773	986±618.7	1,265±627.7	61.2±20.6	49.4±21.8
NH <sub>3</sub>	2,464±617.4	922±695.7	1,283±668.2	63.4±22.7	47.8±23.7

± refers to Standard Deviation values

Comparable BOD<sub>5</sub> removal rates were obtained with the FS and HF membranes (93.2±5.9 vs. 92.2±3.1 %, respectively). Despite the fluctuation in the OLR (0.94 and 1.87 g COD/L.d), the effluent BOD<sub>5</sub> was invariably lower than 62 mg/L and 74 mg/L for the FS and HF membrane systems, respectively. A slightly higher average COD removal efficiency was obtained with the HF membrane (71.4±19.9 vs. 68.5±13.1 %) with a maximum removal efficiency of 98% compared to 89.3% for the FS membrane. The effluent COD of both systems was relatively high due to the presence of refractory compounds typically associated with leachate exhibiting a low BOD<sub>5</sub>/COD ratio (0.07-0.22). Young leachate contains high levels of free volatile acids, BOD<sub>5</sub>, COD, NH<sub>3</sub> and alkalinity (Alvarez-Vazquez *et al.* 2004). However, with the ageing of the landfill, large refractory organic molecules are released from the solid waste and dissolved into the leachate. Hence, better COD removal efficiencies could be obtained when applying the aerobic MBR technology for the treatment of young leachate with high biodegradability (i.e. high BOD/COD).

The discharge of wastewater with high nutrient content (nitrogen and phosphorus) into water systems can stimulate excessive algae growth (eutrophication), deplete dissolved oxygen and hence cause aquatic toxicity. Hence, prior to its release into surface or ground water bodies, leachate treatment is necessary to lower its nutrient content and preclude eutrophication potential (Philips *et al.* 2002; Asano *et al.* 2007; Zhang *et al.* 2013 b). The leachate utilized in this study has shown higher levels of phosphorus (31±13 mg/L) than



previously reported values for similar mature landfill leachate by Tatsi & Zouboulis (2002) (1.27-19.9 mg/L) and Xie *et al.* (2010) (10-24 mg/L). Nevertheless, both membranes achieved high and comparable TP removal rates (78.5±9 and 79.4±7.8 % for the FS and HF, respectively). These removal rates are higher than the value of 65% reported for the treatment of slaughterhouse wastewater using MBR (with influent TP of 16.25 mg/L) (Gürel & Büyükgüngör 2011), 60.51% reported for the treatment of municipal wastewater using combined MBR (consisting of 3 zones anoxic, anaerobic, aerobic) (Liu *et al.* (2012), and 74.3% reported for the treatment of municipal wastewater using sequencing batch MBR (with influent TP of 4.2-5.8 mg /L) (Liu & Lv 2012). As for phosphate, it was also found at high levels, and higher phosphate removals were obtained with the HF membrane (87.3±15.6 %) when compared to the FS membrane (81.27±15.6%).

Unfortunately, there exists no mechanism for ammonia removal under landfills methanogenic conditions leading to its accumulation (Kurniawan *et al.* 2010; Kaczorek & Ledakowicz 2006) representing long-term concerns that extend beyond the landfill lifetime (Berge *et al.* 2006) and necessitates nitrogen removal before the discharge of leachate into surface water since nitrogenous LFL causes eutrophication, aquatic toxicity as well as the release of nitrous oxide into the atmosphere (Philips *et al.* 2002). In this study, leachate characterization showed that NH<sub>3</sub> was the main fraction of TN (around 96.8%). The concentrations of NH<sub>3</sub> (1,770-4,410 mg/L) was higher than the range of 39-1,750 mg/L reported for old leachate by Tatsi & Zouboulis (2002) and 1,700-2,000 mg/L reported by Xie *et al.* (2010). The FS membrane achieved significantly higher TN and NH<sub>3</sub> removal rates than 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<sub>3</sub>). In general, moderate NH<sub>3</sub> removal rates were achieved by both systems, probably due to the inhibition effect of high ammonia concentration (higher than 1,000 mg/L) exerted on nitrobacteria and nitrosomonas species (Ince *et al.* 2013; Ahn *et al.*

2002) necessary for nitrification. In fact, high ammonia levels (more than 1,000 mg/L as ammonium or as organic nitrogen), encountered in old LFL result in inhibition of nitrobacter and nitrosomonas species (Ince *et al.* 2013; Ahn *et al.* 2002), an increase in biomass washout, as well as a reduction in microbial activity and hence decrease the rate of nitrification in biological systems (Ahmed & Lan 2012) thus necessitating special pre-treatment for NH<sub>3</sub> removal, such as ammonia stripping. Indeed, the inhibiting effect of high NH<sub>3</sub> concentrations on ammonia and nitrite oxidation has been widely reported (An *et al.* 2006; Wichitsathian *et al.* 2004). When applying ammonia stripping prior to the MBR, the NH<sub>3</sub> removal efficiency of the MBR increased from 25% to 35-42% at an HRT of 16-24 hours (Wichitsathian *et al.* 2004). The low ammonia removal efficiency can be attributed to ammonia toxicity, which was validated when the COD removal efficiency improved from 63% to 74% after the stripping process.

The percent removal of pollutants from the FS and HF membranes were calculated and compared in an effort to assess the impact of the membrane type on the removal efficiency. Paired t-tests were conducted, when the pollutant concentrations did not show significant deviations from normality based on the Shapiro test. When the data violated the normality assumption, the non-parametric paired Wilcoxon Signed-Rank test was used instead. The data indicates that at the 95% confidence interval ( $\alpha = 0.05$ ), the removal efficiency of the two membranes is not statistically different for TP, BOD<sub>5</sub>, and COD. However, the HF membrane appears to be significantly better than FS in reducing the concentration of PO<sub>4</sub><sup>3-</sup> in the reactor. For TN and NH<sub>3</sub>, the FS membrane was found to be significantly better than the HF in reducing the influent concentration. A summary of the results is shown in Table B-4.

Table B-4: A summary of statistical analysis results

Parameter	Mean Removal Efficiency (%)		Test	P-value
	FS	HF		
TP	78.5	79.4	Wilcoxon Signed-Rank	0.1538
PO <sub>4</sub> <sup>3-</sup>	81.3	87.3	Wilcoxon Signed-Rank	0.02361 *
TN	61.2	49.4	t-test	0.04419 *
NH <sub>3</sub>	63.4	47.8	t-test	0.001723 *
BOD	93.2	92.2	Wilcoxon Signed-Rank	0.06629
COD	68.5	71.4	Wilcoxon Signed-Rank	0.9184

\* significant at the 95% confidence level

### B.3.3 Comparative Assessment

Table B-5 compares average removal efficiencies of various indicators with those reported in the literature using the MBR technology or hybrid systems with MBRs treating old leachate.

Table B-5: Comparison with removal efficiencies reported in the literature

Reference	Location	Scale	Process	Membrane configuration	Influent characteristics		Operational conditions		Removal efficiency
					COD, mg/L	BOD/COD (age)	HRT, days	SRT, days	COD, %
Zhang <i>et al.</i> 2013 a <sup>c</sup>	China	Lab	Fenton oxidation+MBR+RO	Sub(HF)	1200-1600 <sup>a</sup>	0.09-0.12	4	45	83-87.5
Litas <i>et al.</i> 2012	Greece	Pilot	SMBR (SBR) Mixture of LFL+Synthetic WW (1:1)	Sub (FS)	1772	(O)	9	-	95
Chiemchaisri <i>et al.</i> 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 <i>et al.</i> 2010 <sup>c</sup>	Turkey	Lab	UASB+MBR+MAP	Sub	4250 <sup>a</sup>	(M)	-	-	10-70
Puszczalo <i>et al.</i> 2010 <sup>c</sup>	Poland	Lab	Mixture of 10% LFL+synthetic WW/SBR	Sub (MF/Cap)	3000–3500	0.06(O)	2-3	15	89
Aloui <i>et al.</i> 2009 <sup>c</sup>	Tunisia	Lab	Stirred tank reactor	Ext (MF/Tub)	7100–8000	0.18(O)	2–3	-	70–77
Feki <i>et al.</i> 2009 <sup>c</sup>	Tunisia	Lab	MBR/electrochemical oxidation	Ext(Tub)	6500-8000	0.09 (O)	-	-	61
Ratanatamskul, & Nilthong 2009	Thailand	Lab	BPAC-MBR	Sub (HF)	5000–6000 1000 <sup>a</sup>	~0.1 (O)	1	Inf.	83
Svojitka <i>et al.</i> 2009	Germany	Bench	Compartmentalized activated sludge tank	Ext (UF/Tub)	2200	<0.05	2.92-7.08	100	<30
Sadri <i>et al.</i> 2008	Canada	Lab	Stirred tank reactor	Sub (HF)	2737–4079	0.11–0.18 (O)	1–3.5	30, 60	54–78
Tsilogeorgis <i>et al.</i> 2008	Greece	Bench	Membrane sequencing batch reactor (MSBR)	Sub (UF/HF)	1391–3977	(O)	10	infinite	40–60
Robinson <i>et al.</i> 2007 <sup>c</sup>	UK	Full	3 aerobic biological tanks in series	Ext (UF/Tub)	5000	0.05	-	-	76
Canziani <i>et al.</i> 2006	Italy	Pilot	MBR+MBBR	Sub (Tub)	6,316	0.3 (O)	-	>45	Up to 75
Schwarzenbeck <i>et al.</i> 2004 <sup>c</sup>	Germany	Full	2 reactors in series (denitrification+nitrification)+AC filter	MF	136–1980	~0.2	-	-	65
Setiadi & Fairus 2003	Indonesia	Lab	Stirred tank reactor (hazardous waste)	Ext(MF/HF)	1800	0.15–0.17	1	32	31.3
Ahn <i>et al.</i> 2002 <sup>c</sup>	South korea	Full	Aeration basin with anoxic +aerobic parts	Sub(MF/HF)	400–1500	(O)	-	-	~38
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-	0.12 (O)	4.16	30	71.4

					7800				
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Cap: Capillary; Ext: External; FS: Flat Sheet; HF: Hollow Fiber; M: Medium; MF: Microfiltration; O: Old; Sub: Submerged; Tub: Tubular; UF: Ultrafiltration; Y: Young

<sup>a</sup> Concentrations after pretreatment or dilution.

<sup>b</sup> COD values in terms of the soluble COD.

<sup>c</sup> Applied post-treatment to MBR (efficiencies are for MBR only)

While the leachate strength in this study (COD: 3,900-7,800 mg/L) was on the higher range of those reported in the literature (136-8,000 g/L), similar, and sometimes better, attenuation was achieved albeit the relatively lower HRT (4.16 days) and SRT (30 days) (vs. HRT of 1-10 days and SRT of 15-inf. days). Note that Zhang et al. (2013 a) achieved 83-87.5% COD removal efficiency after the treatment with Fenton oxidation that resulted in a weaker influent leachate for the MBR system. Litas et al. (2012) and Puszczalo et al. (2010) reported higher removal efficiencies of 95 and 89% after mixing the leachate with synthetic wastewater. The blending scenario can prevent the toxic effect of relatively high ammonium concentrations normally encountered in leachate. The addition of wastewater might also provide extra phosphorus source that can improve the Carbon: Nitrogen: Phosphorus (C:N:P) ratio resulting in a better performance (Hasar *et al.* 2009 a). In addition, Ratanatamskul & Nilthong (2009) reported 83% COD removal when treating weaker leachate with influent COD of 1,000 mg/L and by adopting the Biological Powdered Activated Carbon (BPAC)-MBR system. The enhanced performance could be due to the simultaneous adsorption and biodegradation whereby Powdered Activated Carbon (PAC) contributed to the removal of biologically recalcitrant-resistant compounds.

#### **B.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 a leachate with COD (3,900-7,800 mg/L), BOD<sub>5</sub> (439.7-1,536.7 mg/L), TP (10.5-59 mg/L), PO<sub>4</sub><sup>3-</sup> (5-58), TN (1,500-5,200 mg/L), and NH<sub>3</sub>-N (1,770-4,410 mg/L) as indicators. Almost 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 COD removal efficiency was obtained with the HF membrane (71.4 vs. 68.5%) with a maximum removal efficiency of 98% compared to 89.3% for the FS membrane, similarly higher phosphate removals were

obtained with the HF membrane (87.3 %) when compared to the FS membrane (81.3%). The FS membrane achieved significantly higher TN and ammonia removal rates than the HF membrane (61.2 vs. 49.4% for TN and 63.4 vs. 47.8% for ammonia). The results help in anticipating potential constraints that might be faced at the full scale leachate treatment plant whereby a successful system should target the non-biodegradable COD fraction using a pre/ post physical/chemical process as well as the high NH<sub>3</sub> concentration using ammonia stripping to reduce the influent NH<sub>3</sub> concentration and prevent its effect on nitrification.

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## APPENDIX C Supplementary material

### C.1. SUMMARY OF DATA COLLECTED

#### C.1.2 pH variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
1	8.45	9.21	9.27	8.85	8.9	8.67	8.78
8	8.72					8.92	8.88
11	8.87	8.89	9.01	8.61	8.75	8.58	8.8
15	8.77					8.8	8.9
18	8.72	8.6	8.67	8.65	8.7	8.82	8.98
32	8.68	8.6	8.54	8.29	8.47	8.08	8.12
37	8.59					8.65	8.78
39	8.62	8.64	8.79	8.52	8.48	8.8	8.77
43	8.6					8.72	8.76
46	8.62	8.72	8.77	8.68	8.61	8.79	8.81
50	8.58					8.72	8.79
53	8.49	8.53	8.57	8.64	8.59	8.77	8.85
68	8.7	8.81	8.65	8.61	8.67	8.45	8.78
71	8.69					8.87	8.72
75	8.08	8.37	8.16	8.74	8.51	8.77	8.78
78	8.18					8.74	8.81
81	8.08	8.37	8.16	8.74	8.51	8.77	8.78
85	8.19					8.7	8.83
88	8.11	8.51	8.56	8.67	8.7		8.82
93	8.18						8.7
95	8.14	8.52	8.6	8.66	8.55		8.7
99	8.16						8.69
109	8.17	8.54	8.48	8.61	8.67	8.88	9.01
113	8.17					8.87	8.96
120	8.28					8.19	8.73
127	8.24	8.65	8.42	9.03	8.81	9.06	8.88

### C.1.3 Total Phosphorus variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
1	23	33.00	25.00	25.00	29.33	2.17	4.90
8	23					2.43	1.29
11	20.5	25.00	25.50	27.33	29.33	2.76	2.88
15	36					5.80	6.30
18	27	24.00	24.00	25.50	44.50	5.70	7.10
32	10.5	28.50	42.50	51.00	9.50	3.50	2.20
37	59					15.50	12.00
39	15	9	8.5	7	11.5	2.7	2.1
43	25					7.2	6
46	19.5	24.5	15.5	17	28.5	6.4	4.2
50	15.5					4.6	4.8
53	12.5	11.5	12	14	27.5	4.8	4.1
57	29.5					6.1	10.9
60	20.5	17	19.5	15.5	25.5	7.6	6
68	33.5	29.5	30.5	20.5	46	6.5	6.4
71	49					6.1	7.7
75	47	25	28.5	22	103.5	6.5	6
78	53					6.6	6.6
81	47	25	28.5	22	103.5	6.5	6
85	57					8	8.6
88	32.5	28	24.5	19.5	27.00	13	11
93	27						5.2
95	25.5	27	25	19	23.5		7
99	28						8.2
109	37.5	24.5	30	19	28.5	7.6	5.6
113	30.5					6.5	7.8
116	33	25	26.5	19.5	23.5	6.4	3.9
120	35.5					6.9	5.1
127	27	18.5	20.5	17.5	17.5	4.6	4.9

#### C.1.4 Phosphate variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
1	15.6667	18.5	19.5	8.67	5.67	6.17	1.33
8	23.5					1.57	0.86
11	11	8.5	3	10.67	1.33	8.40	1.60
15	27					5.40	2.70
18	30	47	42	22.50	30.00	4.00	2.50
32	25.5	22.5	21	5.50	0.50	2.50	1.00
37	24					2.5	0.5
39	14.3333	12	8	4.5	2.5	3	0.6
43	14					2.2	0.6
46	22.5	14	2.5	4	1.5	1.7	0.6
50	23.5					1.8	2.1
53	5	4	4	2.5	3	2.1	0.4
57	24					3.4	0.7
60	15	7	9.5	10.5	29.5	4.6	0.8
68	28.5	14	13	24.5	17.5	0.9	1.8
71	42					5.1	2.9
75	45	13.5	19.5	8	18	3.9	3.5
78	52					6	8.5
81	58	13.5	19.5	8	18	3.9	3.5
85	56					5.8	7.8
88	23	13	15	8.5	10	6.5	1.5
93	34.5						6.2
95	37	21	21.5	17	18		4.8
99	41						6.2
109	45.5	18.5	40.5	21.5	12	8.8	4.3
113	38					8.5	25
116	40.5	20	33	26	25	2	5
120	43.5					12.5	29.5
127	23.5	16.5	15	18.5	9	3.8	3.7



### C.1.5 Total Nitrogen variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
1	1800	1150	1600	866.666667	700	350	400
8	1750					628.5714286	728.5714286
11	1550	1250	1450	833.333333	700	640	880
15	1800					750	720
18	1900	2300	1800	1400	1200	910	870
32	2000	1150	900	1400	1200	910	870
37	5200					650	800
39	2500	2400	1850	750	1600	350	1180
43	1500					280	580
46	2150	400	600	750	1000	370	650
50	2250					370	590
53	3350	1450	1400	600	1100	580	710
57	2550					720	770
60	2350	2250	2350	1500	2350	1340	1120
68	2300	2150	2200	1600	2300	1420	1100
71	3700					1178	1424
75	3300	2450	2550	1900	2650	1820	2000
78	2600					800	740
81	3300	2450	2550	1900	2650	1820	2000
85	3100					2100	2200
88	2400	2200	2400	1850	2450	150	2000
93	2350						2100
95	2400	2700	3050	2050	3050		2180
99	2600						2360
109	2550	2600	2650	2050	2000	1530	1540
113	2950					900	980
116	1900	1450	1750	950	1850	1450	1400
120	2300					1010	1330
127	3350	2950	3300	2800	3300	2600	2450

### C.1.6 Ammonia variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
1	1845	1250	1760	470.00	446.67	383.33	431.67
8	1945					612.86	687.14
11	1890	1600	1570	766.67	610.00	636.00	682.00
15	1910					693.00	642.00
18	1830	1820	1700	930	1250	630	500
32	2460	1915	2380	680	640	495	725
37	4410					405	300
39	2345	2245	1625	525	1315	152	500
43	2215					301	868
46	2230	870	400	495	615	152	998
50	2310					216	990
53	1770	1550	1405	510	785	517	688
57	2580					550	505
60	2440	2635	2585	1735	2620	1082	1058
68	2265	2210	2360	1490	2080	1290	2015
71	1790					1014	1182
75	2350	2185	2630	1825	2250	1750	1930
78	2830					1955	2030
81	2350	2185	2630	1825	2250	1750	1930
85	2870					944	950
88	2535	760	2695	2155	2435	60	1880
93	2380						2000
95	2305	1315	1045	1650	2725		1800
99	2535						1900
109	2685	1955	2435	1500	1640	1026	1660
113	2375					985	1900
116	2700	2240	2645	1875	2720	1725	1930
120	3060					1715	1835
127	4257	3265	4257	2850	3280	2930	2690

### C.1.7 BOD variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
32	1536.67	495.33	692.67	446.67	263.33	35.97	84.58
37	1016.67					58.19	68.47
39	956.25	705.83	637.08	355.83	901.25	42.89	101.95
43	830.00					96.83	96.67
46	576.67	317.17	204.00	153.33	214.00	45.70	45.90
50	885.17					213.40	80.00
53	866.83	461.17	517.17	189.83	936.67	29.70	42.90
57	663.67					64.92	64.00
60	718.50	219.67	531.67	173.83	215.67	19.50	47.35
68	590.31	450.98	496.64	199.60	335.44	37.49	67.88
71	444.50					16.35	25.13
75	609.40	311.75	351.99	381.77	373.79	19.40	40.71
78	682.50					28.35	46.20
81	439.74	318.80	326.21	239.53	430.77	5.54	8.23
88	688.00	529.83	658.25	446.08	681.83	80.00	75.46
93	496.15						36.92
95	534.00	427.58	475.17	252.50	559.17		60.00
99	601.71						51.58
109	490.33	319.67	331.17	312.58	483.08	11.52	24.17
113	629.77					112.45	84.53
116	599.50	370.17	325.50	310.50	497.25	13.40	11.17
127	443.17	271.83	384.92	100.50	380.00	22.00	39.42

### C.1.8 COD variation with time

Day	Influent	FS Denitrification	HF Denitrification	FS Reactor	HF Reactor	FS Permeate	HF Permeate
0							
1	7800	8500	5150	9800	8975	4650	3900
8	7000					4750	4000.00
11	5700	9450	6800	8550	8775	3060	3000
15	5750					1750	2610
18	4700	6500	6300	6250	9400	2540	3380
32	6950	9050	5750	5850	10250	2180	5430
37	5150					2500	2000
39	5000	4800	5150	6100	7550	2250	3340
43	3900					890	1250
46	5610	4560	3720	6560	9780	2370	1940
50	5640					3300	1800
53	5480	4720	4670	5350	7710	1720	940
57	5480					1610	2340
60	5070	4980	5170	5380	5330	1600	1100
68	6370	6400	6810	5810	10150	1130	1170
71	6300					1020	1230
75	6700	6230	6660	5450	8720	910	1070
78	6770					720	820
81	6700	6230	6660	5450	8720	910	1070
83	6090					1600	750
85	5880					1500	1340
88	6000	5000	5500	8000	7500	1350	1250
89	6270					725	260
93	6330						120
95	5910	5480	5550	5080	6530		60
96	6100						100
99	5990						350
109	6210	5920	6130	5710	8790	1400	1550
113	6300					1680	1790
116	6240	5900	6080	5540	6690	1040	880
120	5870					1900	1940
127	6050	5610	5220	4710	5810	1260	1270

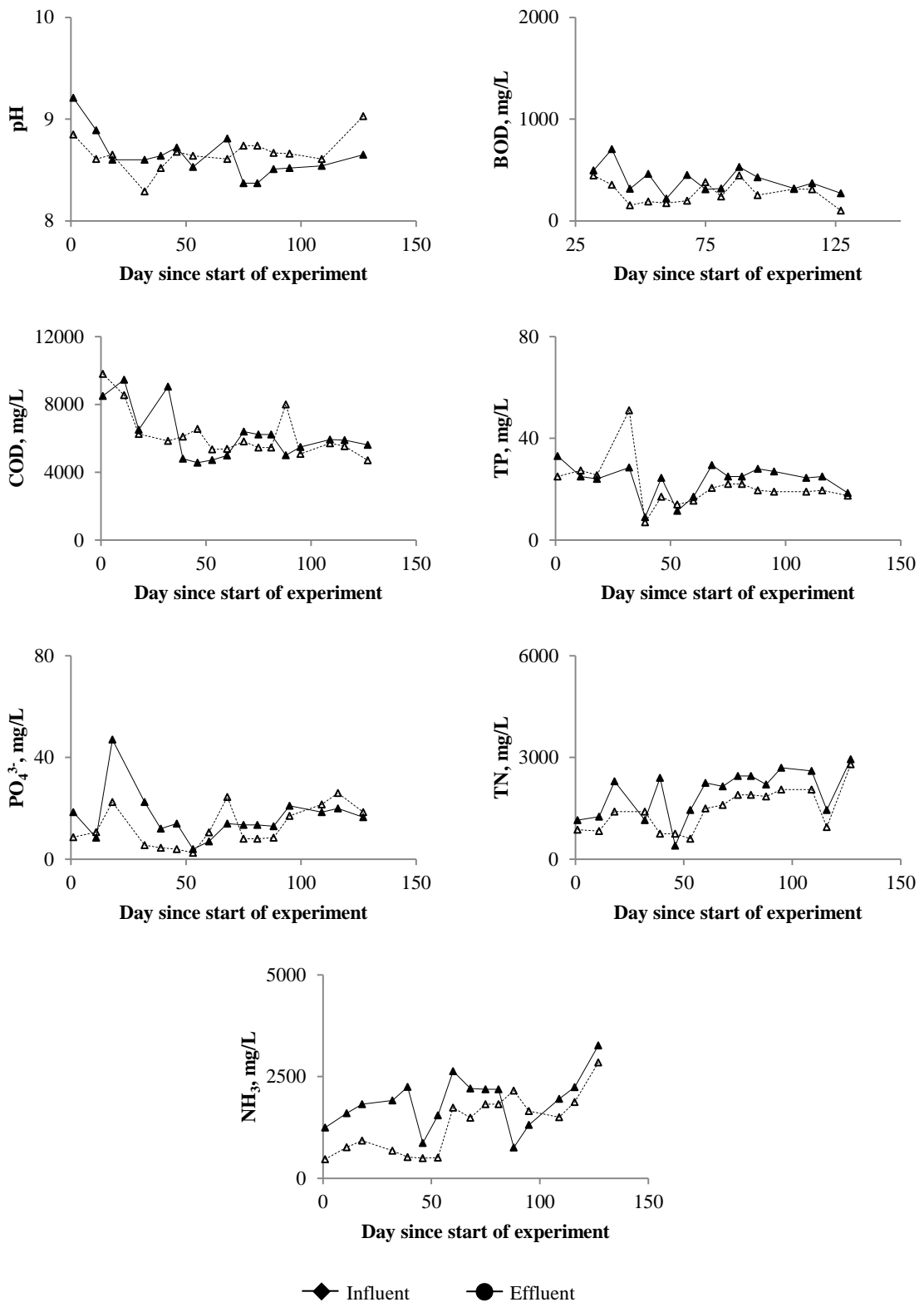


Figure C-1: Temporal variation of main performance indicators: Flat Sheet membrane

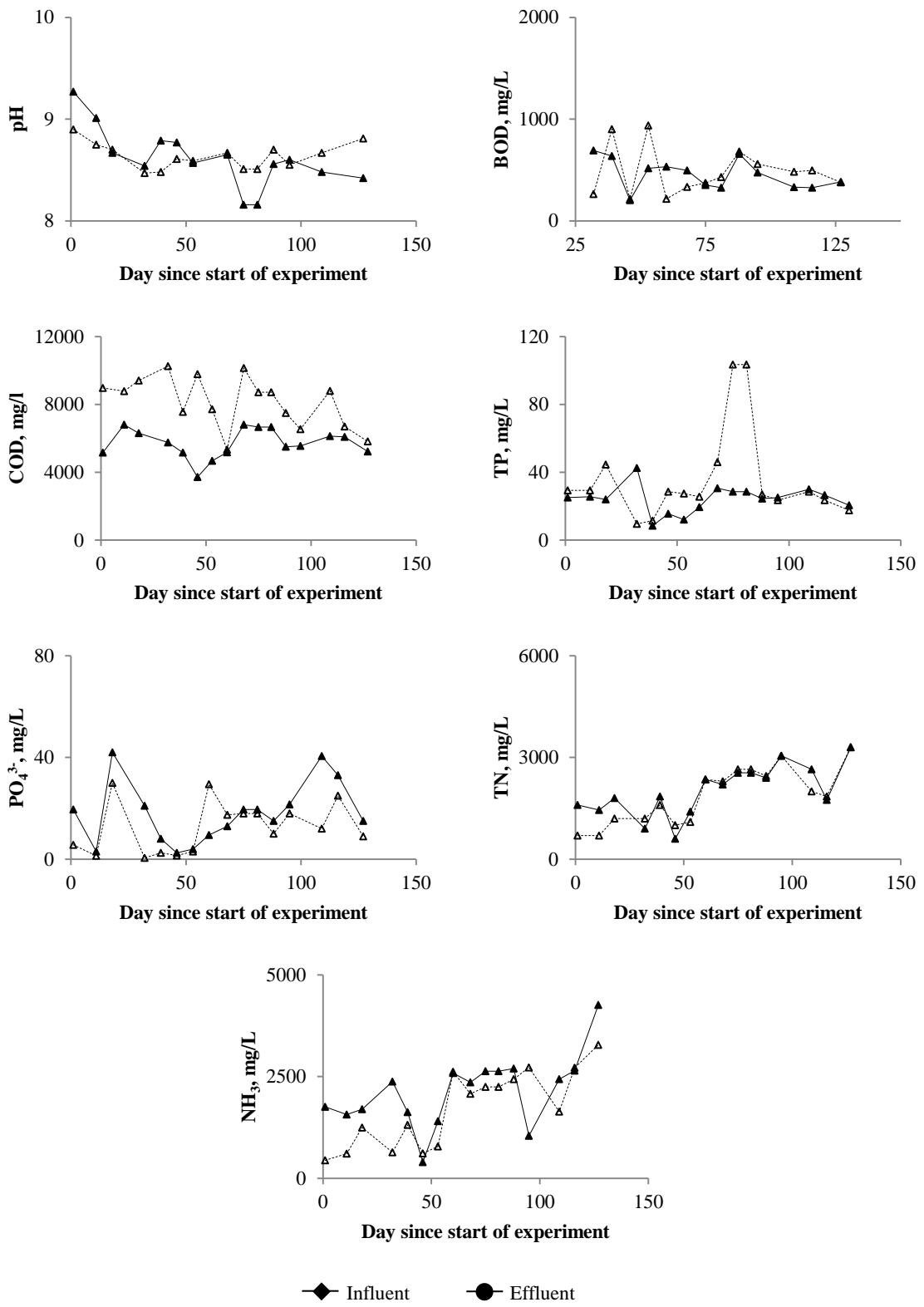


Figure C-2: Temporal variation of main performance indicators: Hollow Fiber membrane

Table C-1: Leachate parameters and corresponding standard methods of analysis

<b>Parameter</b>	<b>Reference method</b>
pH	SM* 4500-H+ B
Total phosphorus	SM 4500 P B (5), E
COD	SM 5220 D
BOD5	SM 5210 B
Ammonia	Hach 8155
Total nitrogen	Hach 10072
Ortho-phosphates	SM 4500-P-E

\*SM: Standard Methods (APHA 2005)

## C.2. FIGURES OF EXPERIMENTAL SETUP

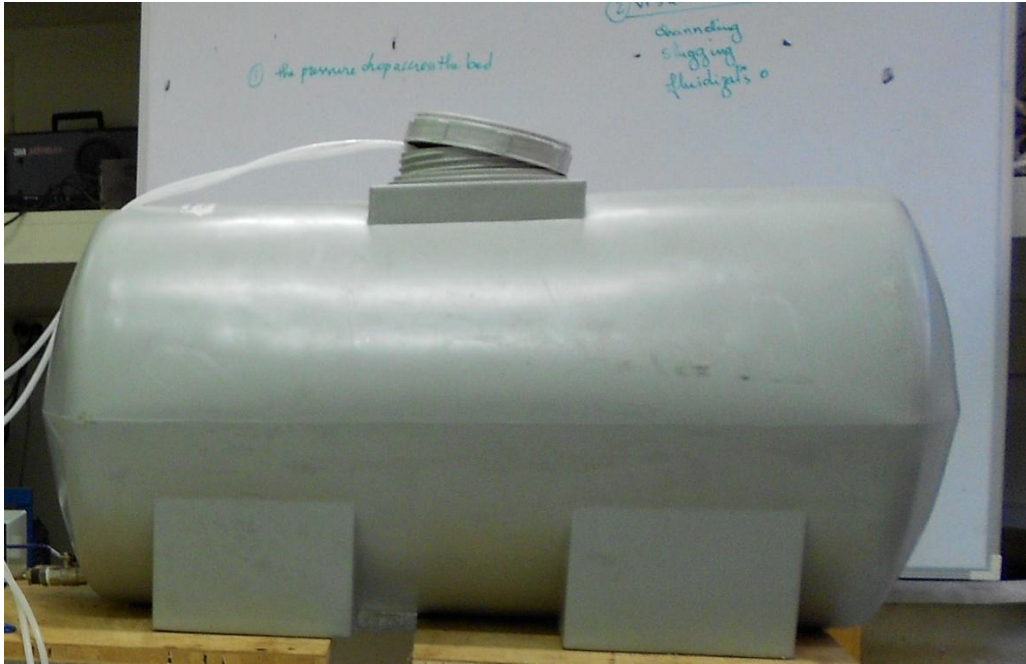


Figure C-3: Feed tank



Figure C-4: Peristaltic pumps



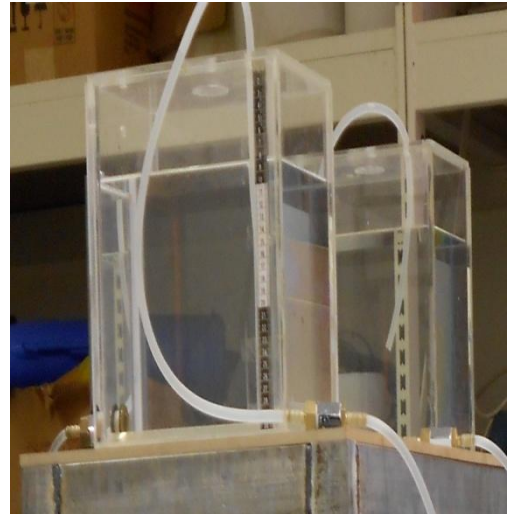


Figure C-5: Denitrification tanks



Figure C-6: Flat sheet MBR (left), hollow fiber MBR (right)



Figure C-7: Pressure sensor (left), rotameter (right)



Figure C-8: Permeate tank



Figure C-9: Experimental setup (during construction)



Figure C-10: Experimental setup (during operation)