



AMERICAN UNIVERSITY OF BEIRUT

IMPACT OF SRT ON THE PERFORMANCE OF  
HF AND FS MBRS FOR THE TREATMENT OF LANDFILL  
LEACHATE WITH MICROBIAL CORRELATION

by  
FATIMA AHMAD SLEEM

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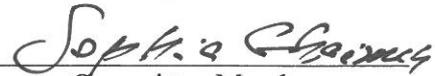
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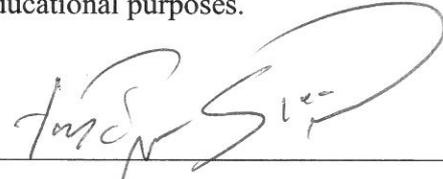
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# AN ABSTRACT OF THE THESIS OF

Fatima Ahmad Sleem for Master of Science in Environmental Sciences  
Major: Environmental Technology

Title: Impact of SRT on the performance of HF and FS MBRs for the treatment of landfill leachate with microbial correlation

This study examines the performance of flat sheet (FS) and hollow fiber (HF) membrane bioreactors (MBRs) for the treatment of landfill leachate (COD = 5010-6900 mg/L, BOD<sub>5</sub> = 406-1228 mg/L; Total Phosphorous (TP) = 14-36 mg/L; Total Nitrogen (TN) = 2300-3400 mg/L) under varying solid retention times (SRT = 5 to 20 days) and a constant hydraulic retention time (HRT = 100 hours). Mixed-liquor bacterial communities were examined over time using 16S rRNA gene sequence analysis in an attempt to define linkages between systems' performance and microbial community composition. Similarly, biofilm samples were collected at the end of each SRT to characterize the microbial communities that evolved on the surface of the FS and HF membranes. The comparative assessment of the FS-MBR and the HF-MBR showed that both membranes exhibited comparable removal efficiencies for BOD<sub>5</sub> (92% in FS vs. 90% in HF), TP (72% in FS vs. 66% in HF), PO<sub>4</sub><sup>3-</sup> (74% in FS vs. 75% in HF) and COD (51% in FS vs. 43% in HF) at SRT = 20 days. The performance of both systems degraded at shorter SRTs, with the FS system exhibiting better overall nitrogen removal. The statistical analysis showed that the removal efficiencies of TP, PO<sub>4</sub><sup>3-</sup>, COD, and BOD<sub>5</sub> were a function of SRT for both membrane bioreactors (p-value <0.05); yet TN removal was found to be independent of SRT.

The biokinetic coefficients governing FS and HF values were estimated with the half-saturation constant (K<sub>s</sub>) (mg COD/L) (1123.63 and 1160.30 for the FS-MBR and HF-MBR, respectively) and the maximum specific growth rate (μ<sub>m</sub>) (day<sup>-1</sup>) (1.42 and 1.72 for the FS and HF, respectively), falling within reported ranges for activated sludge processes (ASP) and MBR applications treating wastewater; yet both the maximum cell yield (Y) (mg VSS/mg COD) (1.04 and 1.87 for the FS-MBR and HF-MBR, respectively) and the endogenous decay coefficient (k<sub>d</sub>) (day<sup>-1</sup>) (0.99 and 1.2487 for the FS-MBR and HF-MBR, respectively) were higher than literature reported values. The fouling assessment showed that the steady-state fouling rate of both membranes increased linearly with the decrease in SRT for the FS-MBR and HF-MBR. The increase in the steady-state fouling rate was estimated at a factor of 1.1 and 1.2, respectively, when the SRT was reduced from 15 to 10 days and from 10 to 5 days.

The microbial analysis showed that similar dominant phyla were detected in both MBRs with *Proteobacteria*, *Bacteroidetes*, and *Firmicutes* constituting around 80% of the oxic and anoxic mixed liquor community. *Bacteroidetes* and *Proteobacteria* were also the dominant groups in the biofilms, forming together more than 60% of the bacterial community at the surface of both membranes. Noticeably, ammonia oxidizing bacteria and nitrite oxidizing bacteria were

not detected at the tested SRTs. It is suggested that nitrifiers were not enriched due to the high organic content of the leachate. Hierarchical clustering and non-metric multidimensional scaling (NMDS) revealed that the mixed liquor communities in the FS-MBR and HF-MBR were clustered separately. Similarly, the biofilm communities on the FS and HF membranes were different. The bacterial community structure was found to be dynamic, especially for the FS MBR. Overall, the mixed liquor community appeared to be more dynamic than the biofilm community.

The above findings suggest that membrane bioreactors for leachate treatment cannot be operated at low solid retention times (< 20 days) without degrading the removal efficiencies of carbon and nutrients. Moreover, the effect of high nitrogen concentrations in the leachate requires pretreatment or post treatment as high nitrogen removals are challenging with MBRs alone and at high organic loadings.

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## ABBREVIATIONS

AC	Activated Carbon
Aer	Aerobic
An	Anaerobic
anMBR	Anaerobic Membrane Bioreactor
AP	Aerobic Processes
ASP	Activated Sludge Process
BF-MBR	Biofilm-Membrane Bioreactor
Biodeg.	Biodegradation
Biof.	Biofilm
BPAC	Biological powdered activated carbon
BOD	Biochemical Oxygen Demand
Cap	Capillary
CAS	Conventional Activated Sludge
CF-ASP	Cross Flow Activated Sludge Process
CF-MBR	Cross flow membrane bioreactor
COD	Chemical Oxygen Demand
d	Day
Den.	Denitrifying
DNA	Deoxyribonucleic acid
EPS	Extracellular Polymeric Substances
Ext	External
F <sub>E</sub>	Effluent Flux
F <sub>S</sub>	Specific flux
FR <sub>ss</sub>	Steady-state membrane fouling rate
FS	Flat Sheet
F/M	Food-to-microorganism ratio
GAC	Granular active carbon
GE	General Electric
h	Hour
HF	Hollow Fiber
HMBR	Hybrid Membrane Bioreactor
HR	Hydrolytic Reactor
HRT	Hydraulic Retention Time
IMBR	Immersed Membrane Bioreactor
k <sub>d</sub>	Endogenous decay coefficient
K <sub>s</sub>	Half-saturation constant
KW	Kruskal-Wallis statistical test
LFL	Landfill Leachate
LMH	Liters per square Meter per Hour
Inf.	Infinite
(M)	Medium
MABR	Membrane Aerated Biofilm Reactor
MAP	magnesium ammonium phosphate
MBR	Membrane Bioreactor
MBBR	moving-bed biofilm reactor
MF	Microfiltration
MLSS	Mixed Liquor Suspended Solids
MLVSS	Mixed Liquor Volatile Suspended Solids
MSBR	Membrane Sequencing Batch Reactor
MW	Mann-Whitney non-parametric U test
N	Nitrogen
NH <sub>3</sub>	Ammonia
Nit.	Nitrifying

NF	Nanofiltration
NaOCl	Sodium Hypochlorite
NMDS	Non-Metric Multidimensional Scaling
(O)	Old
P	Phosphorous
PBS	Phosphate-Buffered Saline
PCR	Polymerase Chain Reaction
PO <sub>4</sub> <sup>3-</sup>	Phosphates
Q	Influent flow rate
OTU	Operational taxonomic unit
RO	Reverse Osmosis
rRNA	Ribosomal Ribonucleic acid
rpm	Revolutions per Minute
S <sub>o</sub>	Influent substrate concentration
S	Effluent substrate concentration
SAMBR	Submerged Anaerobic Membrane Bioreactor
SBR	Sequencing Batch Reactor
SHARON	single reactor for high activity ammonia removal over nitrite
SMBR	Submerged Membrane Bioreactor
SMP	Soluble Microbial Products
SRT	Solid Retention Time
SS	Steady-state
Sub (FS)	Submerged Flat Sheet
Sub (HF)	Submerged Hollow Fiber
T	Time
TMP	Trans-Membrane Pressure
TN	Total Nitrogen
TP	Total Phosphorus
TS	Total Solids
TSS	Total Suspended Solids
Tub	Tubular
UASB	Upflow Anaerobic Sludge Blanket
U	Specific substrate utilization rate
UF	Ultrafiltration
μ	Specific growth rate
μ <sub>m</sub>	Maximum specific growth rate
V	Volume
VSS	Volatile Suspended Solids
X	MLVSS concentration
WW	Wastewater
Y	Maximum cell yield
ZW	ZeeWeed

This thesis is dedicated to my family.

## INTRODUCTION

Landfilling remains a common element, and at times the only element, of a waste disposal strategy in many developing economies due to its relative simplicity and low cost as compared to other waste processing/disposal schemes. However, landfilling is associated with the inevitable formation of heavily polluted leachate generated as a result of biochemical processes and rainwater percolation within landfills, threatening the receiving environment if not properly treated (Guinness and Walpole (2012); (Kurniawan et al. 2006); (El-Fadel et al. 1997)). In this context, various biological and physical/chemical technologies have been developed ((Renou et al. 2008)). Biological technologies, 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 while physical/chemical methods are usually adopted as pre/post treatment or to remove specific recalcitrant pollutants. Stringent discharge standards have been developed to protect ground and surface water resources. As a result, improved combinations of biodegradation and physical separation have been developed. The Membrane Bioreactor (MBR) system is increasingly being recognized as the process of choice for the treatment of high-strength wastewater, containing complex and recalcitrant compounds ((Sutherland 2010); (Bilad et al. 2011); (Yang et al. 2006)). Leachate treatment by MBRs have generally employed hollow fiber (HF) membrane modules with a fewer number adopting the flat sheet (FS) membrane modules ((Cui et al. 2003); (Le-Clech et al. 2006)).

An MBR can be considered as a Conventional Activated Sludge (CAS) system with efficient membrane filtration that holds small particles (size < 0.1  $\mu\text{m}$ ) ((Santos et al. 2011)). 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. MBR leachate treatment has shown high five day- Biochemical Oxygen Demand (BOD<sub>5</sub>) removal rates (90-99%), irrespective of experimental conditions and leachate

maturity ((Hashisho et al. 2016)). 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); (Puszczalo et al. 2010); (Aloui et al. 2009)), depending on the level and type of recalcitrant compounds. Data on phosphorus removal achieved by MBR treating stabilized leachate is scarce, with limited to no data on removal achieved by FS MBRs ((Hashisho et al. 2016)).

The optimal operational solid retention time (SRT) for MBRs remains debatable. Long SRTs are desirable for higher biomass retention, leading to improved treatment efficiency and dominance of slowly growing microorganisms that are able to consume macro-molecules such as polysaccharides, carbohydrates, and protein ((Masse et al. 2006); (Ahmed et al. 2007)). However, excessively long SRTs can lead to low wasting frequency and accumulation of dead and inactive microorganisms, resulting in reduced sludge activity ((Huang et al. 2001); (Han et al. 2005)). Additionally high SRTs (50, 70 and 100 d) promotes microbial lysis, which generates soluble microbial products (SMP) or proteinaceous Extracellular Polymeric Substances (EPS) that can contribute towards fouling ((Han et al. 2005)) and hindering phosphorus removal ((Ersu 2006)). Accordingly, while early MBRs operated at SRTs as high as 100 days, with mixed liquor suspended solids (MLSS) up to 30,000 mg/L ((Hai and Yamamoto 2011)), (recent trends are towards operating under moderate SRTs (10–20 days) with lower MLSS levels (10,000–15,000 mg/L) so as to decrease fouling and reduce the frequency of membrane cleaning ((Le-Clech et al. 2006)).

While the literature on the use of MBR in wastewater treatment is relatively rich particularly in using HF membranes, studies examining the efficacy of various membrane types (i.e. particularly FS membranes) under varying SRT for the treatment of high strength stabilized landfill leachate are limited to non-existent. Therefore, in this study, the two most common membrane modules, HF and FS, were tested in an MBR system to assess their effectiveness in

treating stabilized high strength landfill leachate under varying SRTs (5–20 days) with the objective of defining guidelines for a pilot/full scale plant. During the process, several indicators were monitored at different compartments (oxic and anoxic) of the MBRs. Concurrently, bacterial communities in the mixed liquor (oxic and anoxic) were characterized using 16S rRNA gene sequencing in an attempt to establish linkages between systems' performance and microbial community composition.

## MATERIALS AND METHODS

### 1. MBR construction and operation

The experimental setup (Figure 1) consisted of two 10 L-Plexiglas anoxic tanks (D), stirrer mixers (C) to prevent settlement of solids, and two 50 L-oxic tanks (E, L) also made of Plexiglas with one equipped with FS (L) membranes and the other with HF (E) membranes (Table 1). A blower (Figure 1: 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. Peristaltic pumps (Master Flex 07528-10 and 7550- 22) (Figure 1: 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 multi-channel peristaltic pumps (Figure 1: A, B). The systems were connected to the drain (Figure 1: H) to allow sludge wastage and hence control the SRT. Two pressure sensors (Figure 1: F) (Omega DPG 1000ADA or DAR) connected to a digital display were used to trace variations in membrane pressure. The latter was displayed on the screens of the pressure sensors and the effluent flux was calculated as the flow per unit area. Operating conditions are summarized in Table 2.

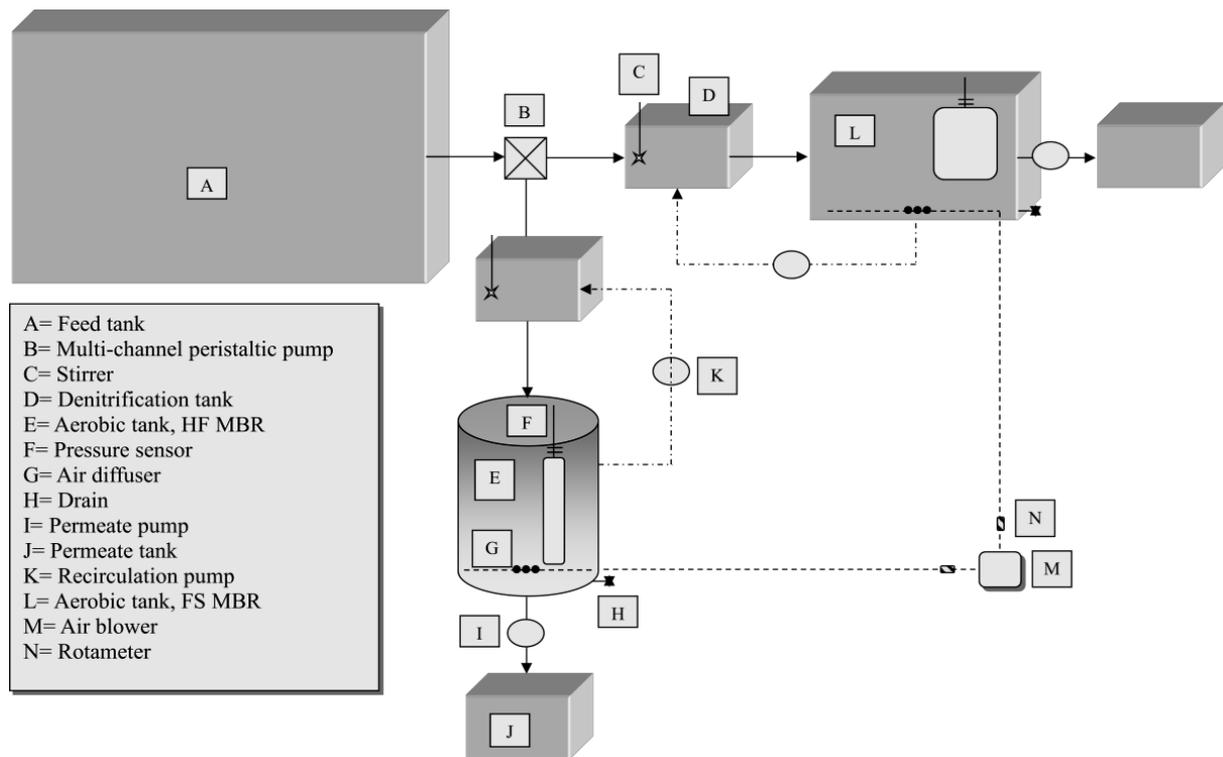


Figure 1 Experimental Setup

Table 1 Properties of membrane modules used

Indicator	Flat Sheet (ACWA)	Hollow Fiber (Zenon)
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

Table 2 Operating conditions of experimental program

Membrane Type	Surface Area ( $\text{m}^2$ )	Permeate Flow ( $\text{mL}/\text{min}$ )	Effluent Flux ( $\text{LMH}$ )	Hydraulic Retention Time (days)	Solid Retention Time (days)
Flat Sheet	0.1	9	5.4	4.16	20
					15
					10
					5
					20
Hollow Fiber	0.93	9	0.58	4.16	15
					10
					5

Leachate was collected weekly from a local 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 oxic tanks, and turning on the mixers in the anoxic tanks at low speed ( $\approx 150$  rpm). The flow rate was increased gradually until reaching an HRT of 100 hours (as adopted by (Alvarez-Vazquez *et al.* 2004) for a full-scale leachate MBR). An anti-foaming agent (Sigma Aldrich) was added (few drops twice per week during the first month, then once every two weeks afterwards) to control foaming in the membrane tanks. When needed, the HF membrane was cleaned using Sodium Hypochlorite (NaOCl) solution, while the FS membrane was cleaned by gentle scraping of solids.

## 2. Sampling and biochemical analysis

Samples were collected twice a week from the feed tank and permeate and once a week from all other tanks. They were analyzed for several indicators including pH, BOD<sub>5</sub>, Total Nitrogen (TN), ammonia (NH<sub>3</sub>), COD, Total Phosphorous (TP), Phosphate (PO<sub>4</sub><sup>3-</sup>), Total Suspended Solids (TSS) and Volatile Suspended Solids (VSS). Pollutant removal data were determined and compared with literature reported values. Additionally, the impact of the adopted SRTs on the removal efficiency of the two membranes was assessed using the non-parametric Kruskal-Wallis (KW) statistical test. The Mann-Whitney (MW) non-parametric U test was used to conduct a post hoc multiple comparisons between the different SRTs with the application of Holm's adjustment. The confidence level for all statistical tests was set to 95% (a significance level of 0.05). All statistical analyses were conducted using the R 3.0 statistical software (R Core (Team 2013)).

### *Biokinetic coefficients*

While the MLVSS is an indicator of microbial abundance in the system ((Macomber *et al.* 2005)), the Monod model (Equations 1 and 2) is widely used to define relationships between

microbial growth and substrate utilization (Al-Malack 2006). The model allows for the estimation of the biokinetic coefficients of biological systems such as the half-saturation constant ( $K_s$ , mg COD.L<sup>-1</sup>), the maximum specific growth rate ( $\mu_m$ , d<sup>-1</sup>), the maximum cell yield ( $Y$ , g<sup>-1</sup>VSSg<sup>-1</sup> COD), and the endogenous decay coefficient ( $k_d$ , d<sup>-1</sup>). Equations 1 and 2 can be rearranged to define the specific substrate utilization rate ( $U$ , mg COD.mg<sup>-1</sup> VSS.d<sup>-1</sup>) as expressed in Equation 4. The specific growth rate ( $\mu$ ) can be expressed as the reciprocal of SRT (Equation 3) (Snider-Nevin et al. 2013).  $Y$  and  $k_d$  can be determined from the slope and the intercept of the regression line representing  $\mu$  versus  $U$ . Similarly, the  $\mu_m$  and  $K_s$ , can be determined from the slope and the intercept of the regression line representing  $SRT/[1 + (SRT + k_d)]$  versus  $1/S$  in Equation 5.

$$\frac{Q}{VX}(S_0 - S) = \frac{1}{Y} \frac{1}{SRT} + \frac{k_d}{Y} \quad (1)$$

$$U = \frac{Q}{VX}(S_0 - S) = \frac{(S_0 - S)}{X \times HRT} \quad (2)$$

$$\mu = \frac{1}{SRT} = YU - k_d \quad (3)$$

$$U = \frac{1}{Y} \mu + \frac{k_d}{Y} \quad (4)$$

$$\frac{SRT}{1 + (SRT + k_d)} = \frac{K_s}{\mu_m} \left( \frac{1}{S} \right) + \frac{1}{\mu_m} \quad (5)$$

Where  $Q$  is the influent flow rate (L.day<sup>-1</sup>);  $S_0$  and  $S$  are the influent and effluent substrate concentration, respectively (mg/L); and  $X$  is the MLVSS concentration (mg/L).

### 3. Fouling analysis

Fouling behavior was assessed in terms of the steady-state fouling rate ( $FR_{ss}$ ; (Liter/m<sup>2</sup>/h.bar<sup>-1</sup>.d<sup>-1</sup>) (LMH.bar<sup>-1</sup>. d<sup>-1</sup>), which is defined as the ratio of effluent flux ( $F_E$ ; LMH) to trans-membrane pressure (TMP; bar) per unit time ( $T$ ; days) (Equation 6).  $F_E$  was maintained constant at 5.4 and 0.58 LMH for the FS and HF membranes, respectively. The specific flux ( $F_S$ ; (LMH.bar<sup>-1</sup>) was plotted against the days of operation, and the corresponding  $FR_{ss}$  was

determined as the slope of the best-fit linear regression line. The effect of SRT on the fouling performance was assessed by examining  $FR_{ss}$  as a function of SRT.

$$FR_{SS} = F_S/T = F_E/(TMP \times T) \quad (6)$$

#### **4. DNA extraction, PCR, 16S rRNA gene pyrosequencing and microbial analysis**

Bacterial 16S rRNA gene sequencing was conducted to characterize the communities in the FS-MBR and HF-MBR operated at an SRT of 20, 15, 10, and 5 days. For each SRT tested, 1.5 mL of mixed liquor samples were collected from each oxic and anoxic tank on a weekly basis. The mixed liquor samples were centrifuged at 10,000 rpm for 10 min to concentrate cells. The supernatant was discarded, and the pellets were stored at -80 °C for subsequent molecular analysis. Biofilm samples on the membrane surface were collected and analyzed at the end of each SRT. The biofilm samples were collected by scrubbing the biofilm attached to the surface of the FS membrane and from one fiber of the HF membrane after cutting it. The FS membrane was rinsed with a phosphate-buffer saline (PBS) solution prior to scrubbing and the HF was rinsed one time slowly by submerging it into the PBS solution. The biofilm samples were collected in 1.5 mL sterile tubes and were stored at -80 °C for subsequent molecular analysis. Genomic DNA was extracted using the PowerSoil DNA extraction kit (MO BIO Laboratories, Inc., Carlsbad, CA), according to the manufacturer's instructions. PCR reactions (triplicate) were performed for each extracted DNA sample in a 25  $\mu$ L reaction volume using the HotStarTaq Plus Master Mix (QIAGEN, Valencia, CA), 0.25  $\mu$ M of each primer and 20 ng of template DNA. Bacterial 16S rRNA genes were amplified using the bacteria-specific forward primer 341F (5'-Adaptor A-Barcode-CA Linker- CCTACGGGNGGCWGCAG-3') and reverse primer 805R (5'-Adaptor B-TC Linker- GACTACHVGGGTATCTAATCC-3') ((Klindworth et al. 2012)). PCR was performed using life technologies veritus thermocycler with the following PCR conditions: 94°C for 3 minutes, 28 cycles of 94°C for 30 seconds; 53°C for 40 seconds and 72°C for 1

minute; and a final elongation step at 72°C for 5 minutes. Following PCR, all amplicon products from different samples were mixed in equal concentrations, purified using Agencourt Ampure beads (Agencourt Bioscience Corporation, MA, USA), and pyrosequenced on the Roche 454 FLX Titanium genome sequencer (Roche, Indianapolis, IN) according to the manufacturer's instructions. The 16S rRNA gene sequences were processed using a proprietary analysis pipeline (MR DNA, Shallowater, TX, [www.mrdnalab.com](http://www.mrdnalab.com)). Raw reads were first demultiplexed to trim the barcodes and primers and then low quality sequence reads defined as outside the bounds of 200 and 1000 bp, sequences containing more than 6 ambiguous base or 6 homopolymers, and sequences with quality score less than 25 were removed. Sequences were then denoised and chimeras removed. Sequences were clustered into operational taxonomic units (OTUs) after removal of singletons with a 97% sequence identity threshold. A representative sequence from each OTU was phylogenetically assigned to a taxonomic identity using BLASTn against a curated Green Genes database ((DeSantis et al. 2006)).

For beta diversity measures, the Bray–Curtis dissimilarity index ((Bray and Curtis 1957)) (Bray and Curtis 1957) was used to determine the percent dissimilarity in microbial community structure between the various samples at the OTU level. The agglomerative hierarchical clustering ((Ward Jr 1963)) was used to generate a dendrogram from the Bray–Curtis dissimilarity matrix using the Community Analysis Package 4.0 (PISCES Conservation Ltd, UK). Also samples were compared by Nonmetric Multidimensional Scaling (NMDS) using the statistical software PRIMER 6 (version 6.1.13) and the PERMANOVA+ add on (version 1.0.3). NMDS ordination was generated based on Bray–Curtis dissimilarity index.

# RESULTS AND DISCUSSION

## 1. Leachate Characterization

The BOD<sub>5</sub>/COD ratio of the feed was relatively low (0.08-0.22, Table 3), which indicates the presence of a considerable portion of slowly- or non-biodegradable COD, such as humic and fulvic acids ((Johansen and Carlson 1976); (Miller and Clesceri 2003); (Harmsen 1983)). This is typical of old landfills, where the leachate has attained a certain degree of maturity ((Kewu and Wenqi 2008)). While the COD, BOD<sub>5</sub>, and pH of the leachate fell within reported ranges for old leachate (Table 4), the TP and NH<sub>3</sub> levels of 15–37mg/L and 2200- 3200 mg/L, respectively, were higher than commonly reported ranges of 1–20 mg/L and 39–2000 mg/L, respectively, for old leachate ((Tatsi and Zouboulis 2002); (Xie et al. 2010)). The high N and P content of the leachate can be attributed to uncontrolled discharge of high nutrient waste such as detergents and agricultural waste, typical in developing economies ((Chen 1996); (Robinson 2007)).

Table 3 Feed BOD<sub>5</sub>and COD under varied SRT

<i>SRT (days)</i>	<i>Operating period (days)</i>	<i>BOD<sub>5</sub>(mg/L)</i>	<i>COD (mg/L)</i>	<i>Feed BOD<sub>5</sub>/COD</i>
20	59	724 ±179.83	6088±614.55	0.12±0.02
15	26	750± 202.94	5399± 161.81	0.17± 0.02
10	29	947±78.36	5253±65.19	0.18±0.02
5	15	1003±17.28	5218±62.38	0.19 ±0.00

Table 4 Composition of feed leachate compared with reported ranges from the literature

<i>Parameter</i>	<i>Unit</i>	<i>Present Study (SRT, days)</i>				<i>Leachate</i>	
		<i>20</i>	<i>15</i>	<i>10</i>	<i>5</i>	<i>Old*</i>	<i>Young*</i>
pH		8.31-8.49	8.32-8.45	8.2-8.44	8.17-8.25	7.3 – 8.8	4.9 – 6.7
BOD <sub>5</sub>	mg/L	406-1020	742-1228	790-1022	988-1024	50-4200	9500-80795
COD	mg/L	5010-6900	5110-5600	5150-5330	5150-5300	685-15000	44000-115000
BOD <sub>5</sub> /COD**		0.08-0.16	0.14-0.22	0.15-0.19	0.19-0.20	0.05-0.64	0.14-0.97
NH <sub>3</sub>	mg N/L	2200-3035	2730-3140	2810-3200	3100-3200	39-1750	1400 -10250
TN	mg N/L	2300-3350	2800- 3200	2900-3400	3250-3400	370-1800	2023-10558
TP	mg P/L	14.5-36.5	23-36	19.5-32.5	31.5-34	1.27-19.9	1.6-65
PO <sub>4</sub> <sup>3-</sup>	mg P/L	12-27	20-28	17-28	29-31	0.12-10	-
TSS	mg/L	510-1390	250-790	470-2830	530-690	10-5900	400-1900
VSS	mg/L	220-710	160-690	200-230	100-120	130-11000	8500-51000

\* (Tatsi and Zouboulis 2002); \*\*BOD<sub>5</sub>/COD of the feed

## 2. Performance Assessment

Generally, all indicators except for TN exhibited a drop in removal efficiencies as the SRT decreased from 20 to 5 days (Table 5 and Figure 2) consistent with reported performance degradation under reduced SRT in MBRs treating wastewater ((Tan et al. 2008); (Jadhao and Dawande 2013)). Many studies examined the performance of the MBR process for leachate removal coupled at times with pre- and/or post-treatment, Table 6 presents removal efficiencies pertaining to the MBR alone for comparative purposes. The achieved removal efficiencies were in agreement with reported values and at times better when taking into consideration the strength of the leachate in this study.

Both membranes achieved comparable COD removal efficiencies across SRTs. Efficiencies were found to statistically decrease as a function of SRT (KW chi squared =27.0 for HF and 29.0 for FS with 3 degrees of freedom; p-values < 0.05). On average, COD removal decreased from ~50% in FS-MBR and 43% in HF-MBR, at SRT=20 days, to 34% in FS and 19% in HF, at SRT=15 days, and to <20% in both MBRs at SRT=10 and 5 days (Figure 2a). Across both membranes, the COD removal rates achieved at SRT 20 were statistically higher than those observed at the lower SRTs (MW U-test p-value < 0.05) (Figure 3- a2 and b2). At SRT 15, only the FS-MBR had COD removal rates statistically better than those achieved at the two lower SRTs (MW U-test p-value < 0.05) (Figure 3- b2). Removal rates achieved under an SRT of 5 and 10 were found to be statistically similar to each other (MW U-test p-value > 0.05).

Literature reported removal efficiencies of COD vary significantly from as high as 99% ((Brown et al. 2013)) at a COD feed of 116,000 mg/L to as low as 23% at a COD feed of 1400-2800 ((Jakopović et al. 2008)). Moreover, at an influent COD concentration of 2200 mg/L (three folds lower than the current study) equally low COD removal (<30%) was reported by (Svojitka et al. 2009) at an SRT of 100 days. High COD removals of old leachate have been attained with combined processes; (Papadopoulos et al. 1998) demonstrated high COD removals (87%) of old

leachate (BOD/COD =0.1–0.17) when primary aerobic treatment was followed by combined chemical and biological oxidation with chemical precipitation as the final step. In addition, (Alvarez-Vazquez et al. 2004) reported that the combination of MBR with nano-filtration (NF) and activated carbon (AC) permits 92–93% COD removal from old leachate at feed COD concentrations up to 5000 mg/L. Moreover, (Jia et al. 2009) reached a rate as high as 98% at a COD feed of 40,000-75000 mg/L using an MBR system coupled with upflow anaerobic sludge blanket (UASB) unit.

Similar to COD removals, BOD<sub>5</sub> removal efficiency tended to increase as a function of SRT for both membranes. For both membranes, removal rates were found to statistically increase as SRT was prolonged (KW chi squared =27.4 for HF and 29.8 for FS with 3 degrees of freedom; p-values < 0.05) (Figure 3- a1 & a2). Both MBRs achieved their highest BOD<sub>5</sub> removal rates (>90%) at SRT=20 days (Figure 2b). As SRT decreased, the BOD<sub>5</sub> removal rates decreased reaching moderate (<70% at SRT=15 days) to low (<65% at SRT=10 and 5 days) values (Figure 2b). In general, high BOD<sub>5</sub> removals (90-99%) were reported in the literature at high SRTs (>20 days) at a BOD<sub>5</sub> feed of (440-45000 mg/L) using FS and HF MBRs, with the exception of a recent study that reported an efficiency ranging between 75-99% ((Akgul et al. 2013)) although the efficiency was mostly greater than 90%.

The removal of phosphorus compounds was comparable in both MBRs and TP removal efficiencies were found to increase as a function of SRT for both membranes (KW chi squared =12.0 for HF and 21.4 for FS with 3 degrees of freedom; p-values < 0.05). For the HF-MBR, the Mann-Whitney post-hoc U-test with Holm's correction showed that removal rates achieved at SRT 20 days were statistically similar to the rates achieved at SRT 15 (MW U-test p-value = 0.536). Removal rate were statically better than those achieved at the lower SRTs (MW U-test p-value = 0.036 with SRT 10; and MW U-test p-value = 0.05 with SRT 5). Moreover, no statistically significant differences were found between the removal efficiency observed at SRT

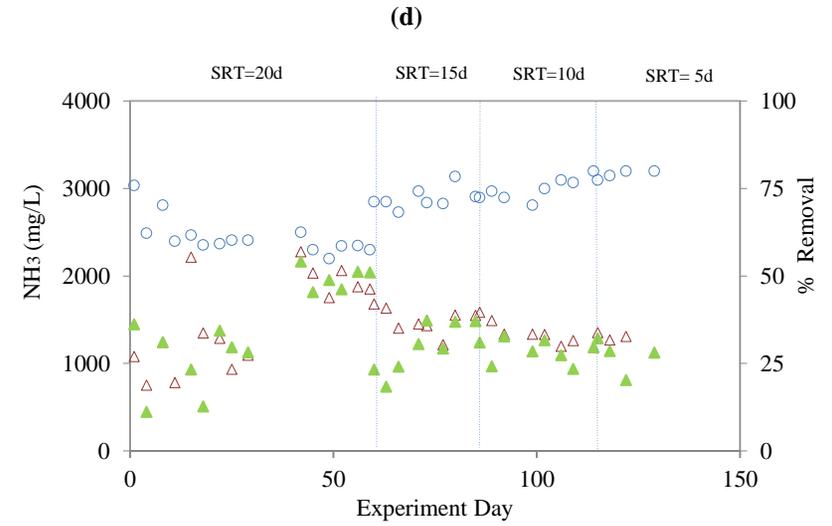
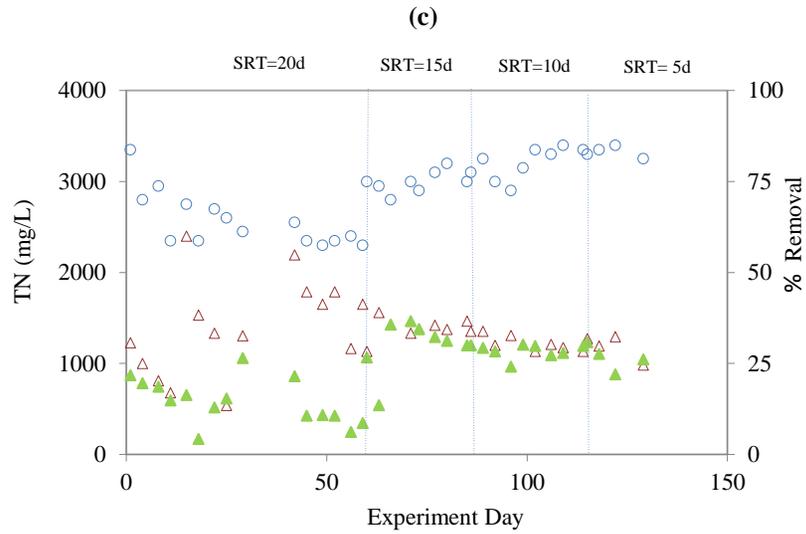
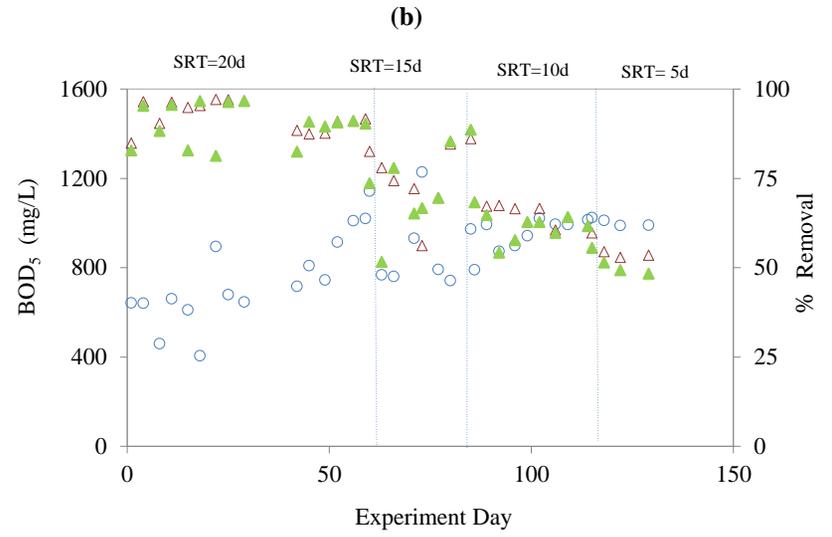
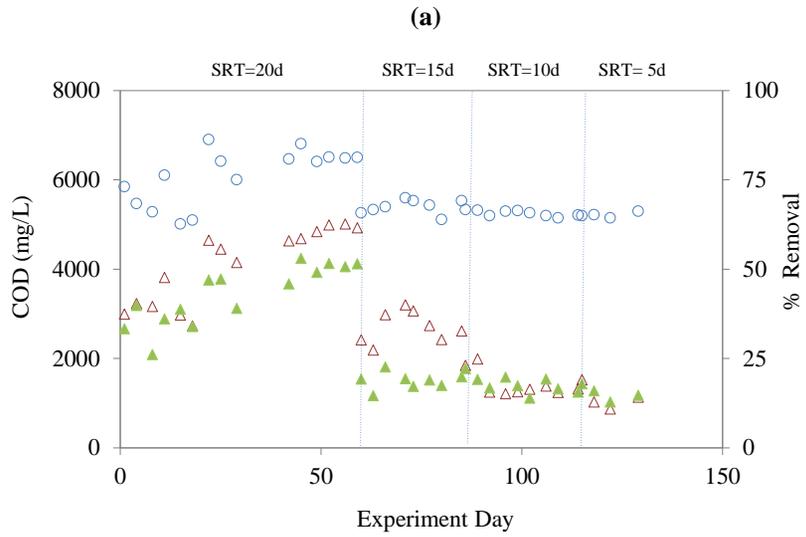
5, 10, or 15 (MW U-test p-value > 0.05 across all comparisons) (Figure 3-a6). As for the FS-MBR, the post-hoc test showed statistically significant differences in the removal efficiencies at all SRT, except those at SRT 10 and 15 days (MW U-test p-value p-value 0.31) (Figure 3-b6). Similarly, the removal efficiency of  $\text{PO}_4^{3-}$  was found to vary significantly by SRT for both membranes (KW chi squared =14.8 for HF and 16.7 for FS with 3 degrees of freedom; p-values < 0.05), with higher SRTs associated with higher removals (Figure 3- b5).  $\text{PO}_4^{3-}$  removal rate was relatively similar to that of TP at SRT=20 days: 74 and 75% for FS-MBR and HF-MBR, respectively, for  $\text{PO}_4^{3-}$ (Figure 2f); and 72 and 66% for FS-MBR and HF-MBR, respectively, for TP (Figure 2e). For both membranes, the post-hoc test using the Mann-Whitney U-test with Holm's correction showed statistically significant differences between SRT 20 on one hand and SRT 5 and 10 on the other, but no difference was found with the removal rates achieved at SRT 15. For FS-MBR, statistically significant differences in the  $\text{PO}_4^{3-}$  removal rates were also found between the rates achieved at SRT 5 and 10 (MW U-test p-value 0 0.035) (Figure 3-a5). Seo et al. (2000), reported 66% TP removal using a submerged MBR for the treatment of domestic wastewater (COD = 216-327 mg/L, TP = 3.8-8.1 mg/L or  $\text{PO}_4^{3-}$  = 2.4-3.5 mg/L) at SRT 25 days. Likewise, Ujang et al. (2002) reported 71%  $\text{PO}_4^{3-}$  removal efficiency at a laboratory-scale MBR fed with synthetic wastewater (COD = 625-678 mg/L,  $\text{PO}_4^{3-}$  = 10.9-12 mg/L) at SRT = 25 days. Lower  $\text{PO}_4^{3-}$  removal efficiency (40-50%) were reported by (Isma et al. 2014) at SRT 4 days for the treatment of synthetic wastewater with influent COD = 1496 mg/L,  $\text{PO}_4^{3-}$  9 mg/L.

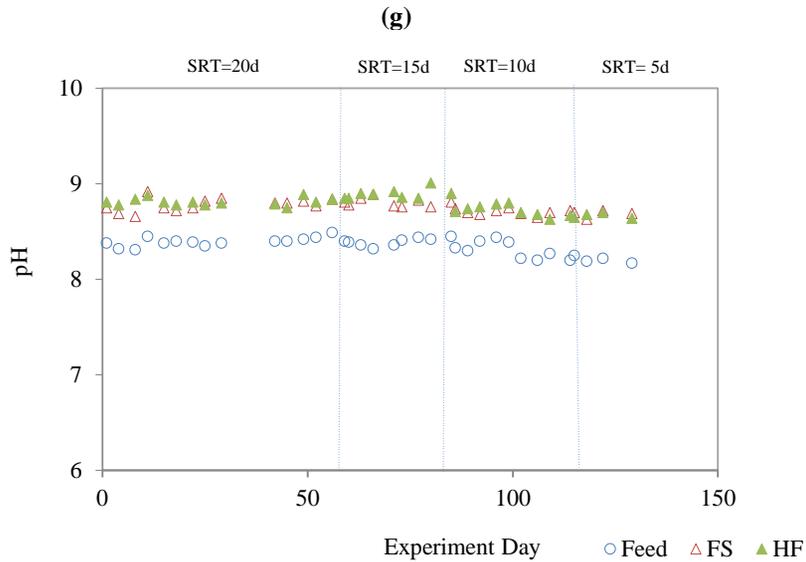
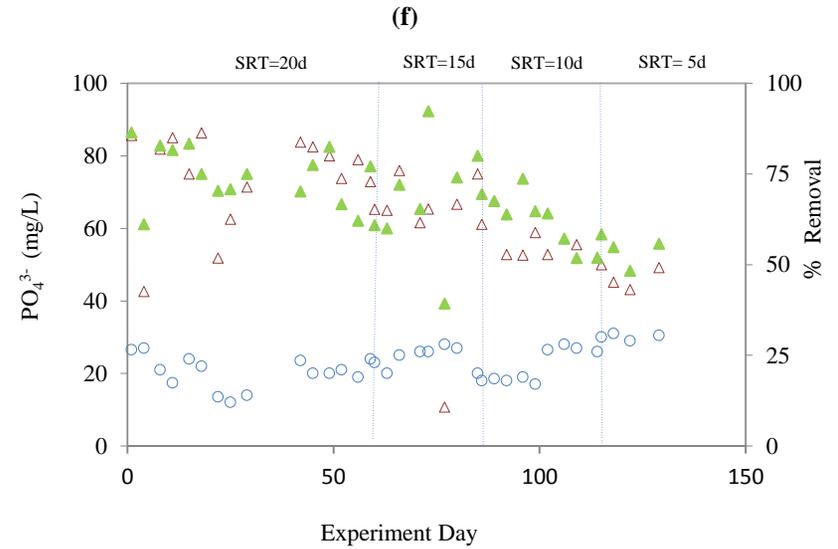
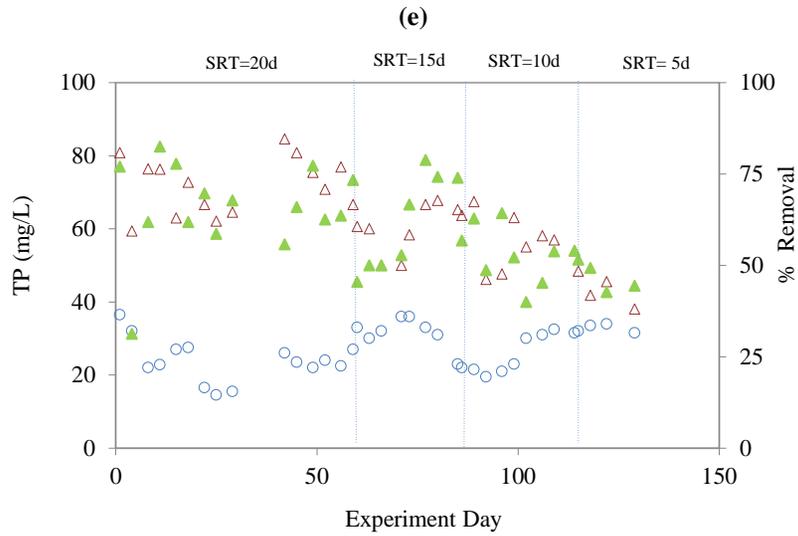
As for TN, both membranes achieved low removal rates. TN removal rates ranged between 30 and 35% in the FS-MBR and between 15 and 30% in the HF-MBR across the tested SRTs (Figure 2c). No statistically significant differences in removal efficiency were found between the different SRTs for FS MBR (Kruskal-Wallis non-parametric test p-value > 0.05) (Figure 3-b4); yet for the HF removal efficiencies at SRT 20 were exceptionally lower than those recorded at lower SRTs. Regarding the removal of  $\text{NH}_3$ , both membranes achieved comparable

low removal efficiencies (between 27 and 38%) with no statistical difference observed in removal rates across the different SRTs (Kruskal-Wallis non-parametric test p-values of 0.5 and 0.3 for the HF and FS respectively) (Figure 3- a3 & b3). The low removal rates may be attributed to the inhibiting effect that the high  $\text{NH}_4^+$  concentrations in the feed (>1,000 mg/L) may have had on nitrifiers (Ahmed and Lan 2012; Ahn et al. 2002; An et al. 2006; Ince et al. 2013; Wichitsathian et al. 2004)). High levels of  $\text{NH}_4^+$  in leachate are a result of the lack of a mechanism capable of removing it under the landfills' methanogenic conditions, leading to its accumulation (Kaczorek and Ledakowicz 2006; Kurniawan et al. 2010). Pre-treatment through ammonia-stripping (An et al. 2006; Hasar et al. 2009b; Wichitsathian et al. 2004), biological fluidized bed with granular activated carbon (Horan et al. 1997) fluidized beds (Imai et al. 1993), or a combination of different technologies such as biological treatment, electro-beam radiation and chemical oxidation (Bae et al. 1997; Lin and Chang 2000) can achieve removal rates in excess of 80 to 85%. As such, establishing pre-treatment prior to MBR treatment is imperative (Berge et al. 2006) so as to limit the discharge of high nitrogenous compounds into surface water bodies causing the release of nitrous oxide into the atmosphere and promoting eutrophication and aquatic toxicity (Philips et al. 2002). A summary of the achieved removal efficiencies are shown in Table 5.

Table 5 Removal efficiencies (%) of various indicators at different SRTs (days) Values reported are the mean removal efficiency  $\pm$  1 standard deviation  
P-values are reported for the non-parametric Kruskal Wallis tests

SRT	% removal											
	BOD <sub>5</sub>		COD		NH <sub>3</sub>		TN		TP		PO <sub>4</sub> <sup>3-</sup>	
	FS	HF	FS	HF	FS	HF	FS	HF	FS	HF	FS	HF
20	92.4 $\pm$ 4.1	90.0 $\pm$ 5.5	51.0 $\pm$ 10.6	42.9 $\pm$ 8.2	37.7 $\pm$ 13.2	36.00 $\pm$ 14.13	35.1 $\pm$ 13.3	14.6 $\pm$ 6.3	71.8 $\pm$ 7.8	65.8 $\pm$ 12.4	74.3 $\pm$ 12.9	74.3 $\pm$ 12.9
15	75.4 $\pm$ 9.8	72.3 $\pm$ 11.9	33.8 $\pm$ 4.4	18.6 $\pm$ 2.3	37.3 $\pm$ 3.7	29.7 $\pm$ 7.2	34.7 $\pm$ 3.1	30.1 $\pm$ 7.4	59.8 $\pm$ 6.9	61.5 $\pm$ 13.3	60.7 $\pm$ 20.8	68.0 $\pm$ 15.7
10	65.0 $\pm$ 2.8	61.8 $\pm$ 4.1	17.8 $\pm$ 3.6	17.8 $\pm$ 2.5	33.6 $\pm$ 3.4	28.6 $\pm$ 3.4	30.8 $\pm$ 2.2	28.5 $\pm$ 1.9	56.9 $\pm$ 7.2	53.1 $\pm$ 7.8	56.7 $\pm$ 5.2	62.7 $\pm$ 7.6
5	55.1 $\pm$ 3.2	51.2 $\pm$ 3.2	14.2 $\pm$ 3.5	15.3 $\pm$ 2.1	31.6 $\pm$ 2.5	27.3 $\pm$ 5.0	29.7 $\pm$ 3.5	26.7 $\pm$ 3.7	43.5 $\pm$ 4.5	47.0 $\pm$ 4.1	46.9 $\pm$ 3.3	54.3 $\pm$ 4.3
KW test p-value	<0.05	<0.05	<0.05	<0.05	0.3	0.5	0.3	<0.05	<0.05	<0.05	<0.05	<0.05





- Feed Concentration (mg/L)
- ▲ HF Removal Efficiency (%)
- △ FS Removal Efficiency (%)
- SRT Solid Retention Time
- COD Chemical Oxygen Demand
- BOD Biochemical Oxygen Demand
- TN Total Nitrogen
- $NH_3$  Ammonia
- TP Total Phosphorus
- $PO_4^{3-}$  Phosphates

Figure 2 Temporal variation of feed concentrations and removal efficiencies of various indicators:  
 (a) COD, (b) BOD, (c) TN, (d)  $NH_3$ , (e) TP, (f)  $PO_4^{3-}$ , (g) pH

Table 6: 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> %
This study	Lebanon	Lab	MBR	Sub (FS)	6088	0.12 (O)	2450	4.16	20	51.0	92.4	37.7
				Sub (HF)	6088	0.12 (O)	2450	4.16	20	42.9	90.0	36.00
				Sub (FS)	5399	0.17 (O)	2890	4.16	15	33.8	75.4	37.3
				Sub (HF)	5399	0.17 (O)	2890	4.16	15	18.6	72.3	29.7
				Sub (FS)	5253	0.18 (O)	2994	4.16	10	17.8	65.0	33.6
				Sub (HF)	5253	0.18 (O)	2994	4.16	10	17.8	61.8	28.6
				Sub (FS)	5218	0.19 (O)	3163	4.16	5	14.2	55.1	31.6
				Sub (HF)	5218	0.19 (O)	3163	4.16	5	15.3	51.2	27.3
(Hashisho et al. 2016)	Lebanon	Lab	MBR	Sub (HF)	5,978	0.12 (O)	2,464	4.16	30	71.4	92.2	47.8
				Sub (FS)	5,978	0.12 (O)	2,464	4.16	30	68.5	93.2	63.4
(Trzcinski and Stuckey 2016)	United Kingdom		anMBR	Sub				10		90		
(Hashemi et al. (2016). )	Iran	Pilot	MBR (compost leachate) <sup>a</sup>	(HF)	50-2200	0.04-0.2				97	98.4	
				(FS)						99	99.7	
(Syron and Casey 2008)	Ireland	Pilot	Membrane Aerated Biofilm Reactor (MABR)	(HF) diluted leachate	1000 to 3000	<0.2	500 to 2500	4.7-7.5		17.5		80-99
(Xue et al. 2015)	China	Lab	MBR	Sub (HF)	783.21	0.32-0.39	41.53			93.61		97.66
(Xue et al. 2015)	China	Lab	MBR	Recirculated (HF)	783.21	0.32-0.39	41.53			93.85		97.86
(Wang et al. 2014)	China	Pilot	Anoxic/Aerobic-MBR	Sub (HF)	3134.88	0.14	434.76			81.5		93.2
(Wang et al. 2014)	China	Pilot	Anoxic/Aerobic-GAC-MBR	Sub (HF)	3134.88	0.14	434.76			89.5		80.5
(Akgul et al. 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 et al. 2013)	Canada	Lab	MBR (compost leachate)	Sub (HF)	116,000		2720	95	-	99.7		≈100
(Campagna et al. 2013)	Turkey	Full	MBR+NF	Ext (UF)	16360		>400	-	-	84.4		88 <sup>d</sup>
(Ince et al. 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 et al. 2013) <sup>c</sup>	Turkey	Lab	MBR+ NF+RO	Sub (UF)	18,685		1245		30	89		83.5
(Sanguanpak et al. 2013)	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9240	0.629 (Y)	--	1	Inf.	87	99	
(Thanh et al. 2013)	Vietnam	Lab	MBR	Sub	1200–1400 <sup>a</sup>		68 ± 26	3.5-14.6 h	30	Up to 97.5		≤92.0 ± 1.5
(Zhang et al. 2013)	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 et al. 2012a)	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9,389	0.746	105-174	1	Inf.	87	97	83-91

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> %
(Boonyaroj et al. 2012b)	Thailand	Pilot	Two-stage MBR (An-Aer)	Sub (HF)	9306	0.72 (Y+O)	138	1	Inf.	87	97	90
(Brito et al. 2012)	Brazil	Lab	MBR <sup>a</sup>	Sub(HF)	3942	0.06	1529			74		
(Syron and Casey 2008)	Ireland	Pilot	MBR <sup>a</sup>	Sub(HF)	1100-1600							600-700
(Coban et al. 2012)	Turkey	Full	Ammonia stripping+MBR+NF	Ext (UF)	24,000	0.33	2313	-	-	93.75		98
(Hua and Zhang 2012)	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 et al. 2012)	Greece	Pilot	SMBR (SBR) Mixture of LFL+Synthetic WW 1:1	Sub (FS)	1772	(O)	269	9	-	95		98.2-99.2
(Mahmoudkhani et al. 2012) <sup>c</sup>	Iran	Lab	MBR+RO	Sub (HF)	68250±8000	0.65	1470	15	55	97	99	99.45
(Lv et al. 2012)	China	Pilot	MBR	Sub (HF)	3600-9700	0.31-0.65	200-620 <sup>a</sup>	4		95	100	61.7
(Santinelli et al. 2012)	Italy	Pilot	MBR	Sub (HF)	802		--	-	-			
(Bai et al. 2011)	China	Lab	Anoxic-oxic hybrid MBR Diluted leachate	Ext	500-4500	-	150-1400	-	-	Up to 90		≤60
(Chiemchaisri et al. 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 et al. 2010)	Turkey	Lab	UASB+MBR+MAP	Sub	4250a	(M)	2315.4 <sup>a</sup>	-	-	10-70		35
(Trzcinski and Stuckey 2016)	UK	Lab	3-stage (HR-SAMBR-MBR)	Sub (FS)	150-1300		--	Variable	300	30-90		
(Li et al. 2010)	China	Pilot	Anaerobic pretreatment/ air-lift bioreactor	Ext(UF/Tub)	4670–6700	(Y)	820–960	-	-	87		100
(Puszczało et al. 2010) <sup>c</sup>	Poland	Lab	Mixture of 10% LFL+ synthetic WW/SBR	Sub (MF/Cap)	757-1708	0.06(O)	53.5-510	2-3	15	89	>98	>95
(Aloui et al. 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 et al. 2009) <sup>c</sup>	Tunisia	Lab	MBR/electrochemical oxidation	Ext(Tub)	6500-8000	0.09 (O)	1500	-	-	61	100	72.8
(Hasar et al. 2009a)	Turkey	Bench	Mixture of LFL+ domestic WW	Sub (HF)	8500–14200 +750–2400a	0.4–0.67 (Y)	1100-2150	3.6–6.0 h	5–30	72-99	-	
(Hasar et al. 2009b) <sup>c</sup>	Turkey	Lab	Ammonia stripping+ coagulation/ flocculation pretreatment + aer/an-MBR+RO	Sub (HF)	8500–19200 ~7300a	0.4–0.7 (Y)	200-1000a	3.6–16.4 h	10–50	60-90		87-98
(Jia et al. 2009)	China	Pilot	UASB/MBR	Sub(HF)	40,000-75,000 1440-25600a	0.42-0.52 (Y)	380-1800	0.5	14	98	99	≈100
(Ratanatamskul and Nilthong 2009)	Thailand	Lab	BPAC-MBR	Sub (HF)	5000–6000 1000a	~0.1 (O)	--	1	Inf.	83		
(Svojitka et al. 2009)	Germany	Bench	Compartmentalized activated sludge tank	Ext (UF/Tub)	2200	<0.05	1200	70–170 h	100	≈30	91	90-99
(Jakopović et al. 2008) <sup>c</sup>	Croatia	Pilot	Stirred tank reactor	Sub (HF)	1400–2800	0.46	--	8 h	-	23		

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> %
(Sadri et al. 2008)	Canada	Lab	Stirred tank reactor	Sub (HF)	2737–4079	0.11–0.18 (O)	662±176	1–3.5	30, 60	54–78	>97	>99
(Tsilogeorgis et al. 2008)	Greece	Bench	MSBR	Sub (UF/HF)	1391–3977	(O)	200-279	10	infinite	40–60		≈100
(Xu et al. 2008)	China	Pilot	Combined anaerobic pre-treatment and MBR	Air-lift Ext (UF/HF)	10,084 9357a, b	0.71 (Y)	--	9.5	-	89	>99	
(Judd et al. 2006)	UK	Lab	MBR	Ext (Tub)	2701	(O)	21.77-588	5	30			73-99
(Robinson 2007)	UK	Full	3 aerobic biological tanks in series	Ext (UF/Tub)	5000	0.05	2000	-	-	76	>96	≈100
(Sang et al. 2007)	Vietnam	Lab	Stirred tank reactor	Sub (MF)	4000–39,600	>0.68 (Y)	--	50 – infinite	-	84–97		
(Visvanathan et al. 2007)	Thailand	Lab	Thermophilic MBR	Sub	12000±1000	0.39–0.65 (M)	1000-1700	1	-	62–79	>97	60-75
(An et al. 2006)	China	Pilot	Anoxic +aerobic zone in tank	Sub (MF)	1500	-	500	8.5	-	75	-	80-99
(Bodzek et al. 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 et al. 2006)	Italy	Pilot	MBR+MBBR	Sub (Tub)	6,316	0.3 (O)	1000-1500	-	>45	Up to 75		≥90
(Chen and 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 et al. 2006)	finnish	Pilot	Dual-tank MBR (SBR +MBR)	Sub (HF)	2200±230	(Y)	210±90	2–5	35–60	>80	>97	>97
(Chaturapruek et al. 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 et al. 2004) <sup>d</sup>	Germany	Full	2 reactors in series (denitrification+nitrification)+AC filter	MF	136–1980	~0.2	120	-	-	65	95	97
(Wichitsathian et al. 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 <sup>b</sup>	94–98	
(Setiadi and Fairus 2003)	Indonesia	Lab	Stirred tank reactor	Ext(MF/HF)	1800	0.15–0.17	114.8	1	32	31.3	98	66
(Ahn et al. 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

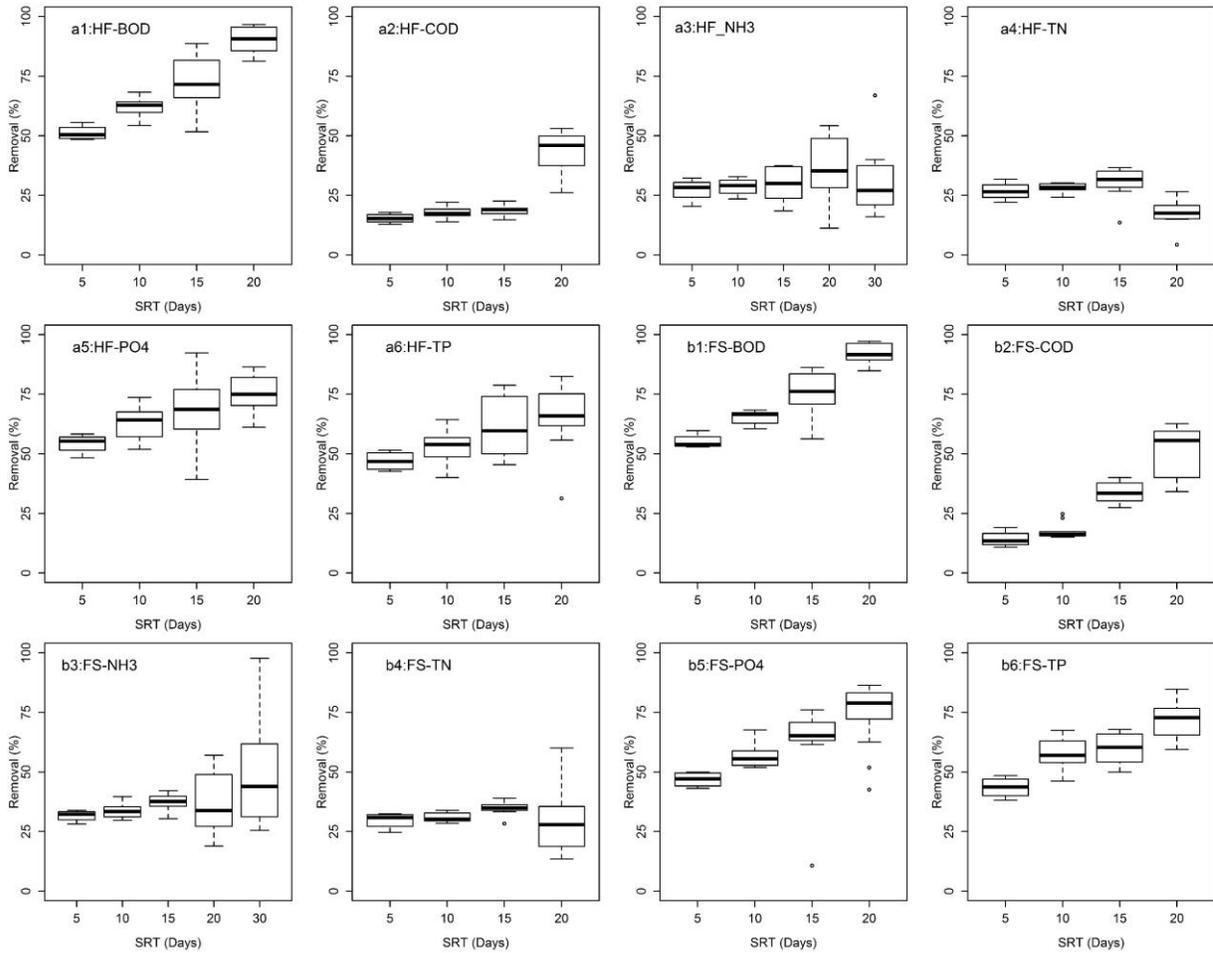


Figure 3 Removal (%) as a function of SRT and MBR type

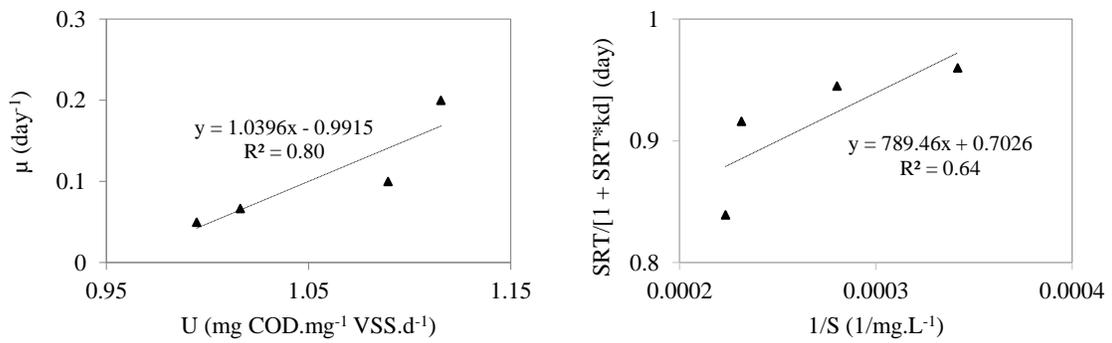
### *Biokinetic Coefficients*

The biokinetic coefficients governing the FS and HF MBRs were obtained using Equations 1 to 5 with corresponding operating parameters summarized in Table 7. The value of the coefficients were estimated using Figure 4 and were compared with literature reported values for various wastewater streams and treatment systems (Table 8). While the observed  $K_s$  and  $\mu_m$  fell within reported ranges for activated sludge processes (ASP) and MBR applications treating wastewater, both  $Y$  and  $k_d$  are higher than reported values (Table 8). Commonly, higher ranges of  $Y$  and  $k_d$  have been correlated with shorter sludge ages (Chaize and Huyard 1991; Huang et al. 2001) similar to SRTs of 5 to 15 days. Also, the higher decay rate ( $k_d$ ) can be attributed to the high shear

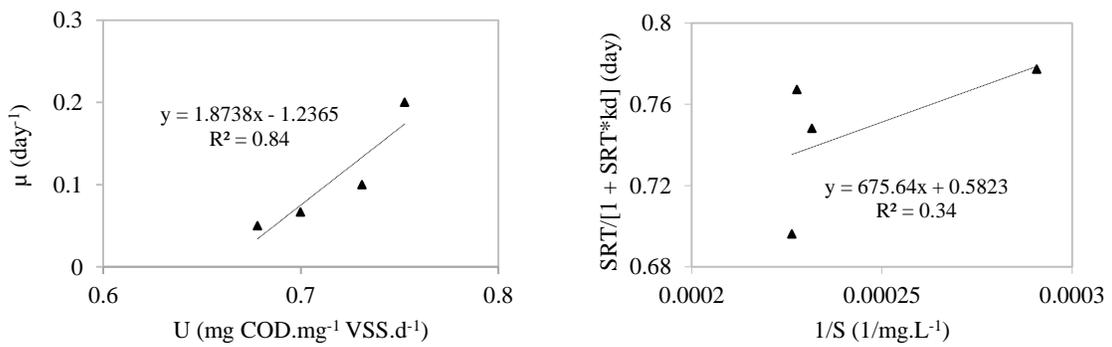
and pressure which the biomass is subjected to under high suspended solids concentrations in the leachate (Rahman and Al-Malack 2012) or unfavorable environment leading to low microbial activity (Cicek 1999).

Table 7 Operating conditions and leachate characteristics (HRT = 100 hrs)

SRT (days)	$\mu$ (/day)	So (mg/L)	FS			HF		
			S (mg/L)	X (mg/L)	U (mg COD / mgVSS/d)	S (mg/L)	X (mg/L)	U (mg COD /mgVSS/d)
20	0.05	6088 ± 6154	2927± 369	764 ± 39	0.99	3440 ± 295	939± 22	0.68
15	0.067	5399±162	3569± 179	433± 92	1.02	4391 ± 157	346± 43	0.70
10	0.1	5253± 65	4319±167	206 ± 48	1.09	4316.67 ±114	308 ± 53	0.73
5	0.2	5218 ± 62	4475 ±178	160 ± 114	1.12	4418± 113	256± 21	0.75



(a) FS MBR



(b) HF-MBR

Figure 4 Estimation of biokinetic coefficients

Table 8 Biokinetic Coefficients for the FS and HF membrane bioreactors in comparison with literature reported values for various wastewater streams and treatment systems

Wastewater	$Y$ (mg VSS/mg COD)	$k_a$ (day <sup>-1</sup> )	$\mu_m$ (day <sup>-1</sup> )	$K_s$ (mg COD/L)	System	Reference
Synthetic	0.49-0.58	0.037-0.151	1.28–6.46	289-2933	IMBR-ASP	(Al-Malack 2006) <sup>a</sup>
Refinery	0.22-0.28	0.07-0.09	0.653-1.2	397-660	CF-MBR	(Rahman and Al-Malack 2012) <sup>b</sup>
Synthetic	0.67	0.056	1.86	6.65	MBR	(Naghizadehi et al. 2014) <sup>c</sup>
Rendering	0.20	0.14	4.11	806	SM-MBR	(Kurian et al. 2006) <sup>d</sup>
Dairy	0.2281	0.1383	1.69	174	MSBR	(Kaewsuk et al. 2010) <sup>e</sup>
Landfill leachate	-	-	1.18	96.4	SBR	(Choudhary 2005) <sup>f</sup>
Dairy	0.714	0.038	-	534	ASP	(Lateef et al. 2013) <sup>g</sup>
Pharmaceutical	0.3-0.72	0.045	0.77	2980.5	ASP	(Raj and Anjaneyulu 2005) <sup>h</sup>
Raw Wastewater	0.5	-	0.87	6691	ASP	(Nakhla et al. 2006) <sup>i</sup>
Municipal	0.48-0.8	0.0189- 0.026	0.95-0.98	52-71	ASP	(Mardani et al. 2011) <sup>j</sup>
Dairy	-	0.015	2.3325	867.76	ASP	(Venkatesan et al. 2004) <sup>k</sup>
Industrial	0.23	0.32	0.26	77.5	UASB	(Jawad et al. 2015) <sup>l</sup>
Brewery	0.357	0.083	0.117	-	UASB	(Enitan and Adeyemo 2014) <sup>m</sup>
Synthetic	0.485-0.622	0.047-0.057	-	149.64-364.81	SBR	(Kundu et al. 2013) <sup>n</sup>
Synthetic	0.13-0.41	0.035	-	-	MBR	(Holakoo et al. 2007) <sup>o</sup>
Synthetic	-	-	8.856	18	MBR	(Thuy and Visvanathan 2006) <sup>p</sup>
Synthetic	-	-	1.86	651	Biodeg.	(Jeswani and Mukherji 2013) <sup>q</sup>
Saline	0.45	0.025	-	195	SBR	(Taheri et al. 2012) <sup>r</sup>
Municipal	0.235-0.286	0.065-0.069	-	-	SBR	(Chen et al. 2008) <sup>s</sup>
Dairy	0.26	0.032	0.44	141	AP	(Carta-Escobar et al. 2005) <sup>t</sup>
Food Processing	0.35	0.13	-	-	ASP	(Hao and Li 1987) <sup>u</sup>
Landfill leachate	1.04	0.99	1.42	1123.63	FS MBR	This study
Landfill leachate	1.87	1.24	1.72	1160.30	HB MBR	This study

<sup>a</sup> (Al-Malack 2006); IMBRs- ASP: Immersed Membrane Bioreactors- Activated Sludge Processes; HRT: 12-16 h; SRT: 2.11- 31.25 d; MLSS: 3000-15000 mg/L

<sup>b</sup> (Rahman and Al-Malack 2012); CF-MBR: Cross Flow- Membrane Bioreactor; SRT: 9.05-36.25 d; COD: 396.62-659.45 mg/L; MLSS: 3000-5000 mg/L

<sup>c</sup> (Naghizadehi et al. 2014); MBR: Membrane Bioreactor; HRT: 4-12.8 h; SRT: 3-17 d; MLSS: 6.8-7.8 g.L<sup>-1</sup>; MLVSS: 5.3-5.8 g.L<sup>-1</sup>

<sup>d</sup> (Kurian et al. 2006); MBR: Membrane Bioreactor; HRT: 5-10 d; COD: 15900-18700 mg/L; BOD<sub>5</sub>: 7700-9050 mg/L; TSS: 1600-1750 mg/L; VSS: 1300-1400 mg/L

<sup>e</sup> (Kaewsuk et al. 2010); MSBR: Membrane Sequencing Batch Reactor; pH: 5.1-6.9; COD: 1700–4000 mg/L; BOD<sub>5</sub>: 2000–2900 mg/L; TP: 2.4–3.6 mg/L; TN: 21–49 mg/L; TSS: 430–750 mg/L

<sup>f</sup> (Choudhary 2005); SBR: Sequencing Batch Reactors; HRT: 24 h; COD: 8,000 mg/L; pH: 6.8-7.0; MLSS: 6,000 mg/L

<sup>g</sup> (Lateef et al. 2013); ASP: Activated Sludge Process; HRT: 2-12 days; BOD<sub>5</sub>: 1400-1900 mg/L; COD: 1500-3500 mg/L; TSS: 400-600 mg/L

<sup>h</sup> (Raj and Anjaneyulu 2005); ASP: Activated Sludge Process; HRT: 4.5 days; pH 7.9; TS: 35886mg/L; TSS: 7131.8mg/L; COD: 12378.4mg/L; BOD<sub>5</sub>: 5992 mg/L

(Nakhla et al. 2006); ASP: Activated Sludge Process; bTSS: 14,470 mg/L; VSS: 13,870 mg/L; COD: 77,300 mg/L; BOD<sub>5</sub>: 77,800 mg/L; NH<sub>3</sub>: 1350 mg/L; PO<sub>4</sub><sup>3-</sup>: 290 mg/L

<sup>j</sup> (Mardani et al. 2011); ASP: Activated Sludge Process; BOD<sub>5</sub>: 240 mg/L; COD: 575 mg/L; TSS: 226 mg/L; pH:7.64

<sup>k</sup> (Venkatesan et al. 2004); ASP: Activated Sludge Process; pH:7.2; TSS: 820 mg/L; BOD<sub>5</sub>: 1050 mg/L; COD: 3600 mg/L

<sup>l</sup> (Jawad et al. 2015); UASB: Upflow Anaerobic Sludge Blanket; TSS: 59 mg/L; VSS: 41 mg/L; HRT: 9-38 h

<sup>m</sup> (Enitan and Adeyemo 2014); UASB: Upflow Anaerobic Sludge Blanket; pH: 6.9; COD: 2005.73 mg/L; BOD<sub>5</sub>:1877.09 mg/L; TSS: 2449.40 mg/L

<sup>n</sup> (Kundu et al. 2013); SBR: Sequencing Batch Reactors; pH: 8-8.5; TSS: 10120-14225 mg/L; BOD<sub>5</sub>:3000- 35000 mg/L; COD: 6185- 6840 mg/L; TN: 1050-1200 mg/L; NH<sub>3</sub>: 650-735 mg/L

<sup>o</sup> (Holakoo et al. 2007); MBR: Membrane Bioreactor; SRT: 20 & 40 d; HRT: 4h; TSS: 10700-15200 mg/L; VSS: 9400- 14700 mg/L; COD: 286-301 mg/L; NH<sub>3</sub>: 25.6-28.5 mg/L; PO<sub>4</sub><sup>3-</sup>: 4.9-6 mg/L

<sup>p</sup> (Thuy and Visvanathan 2006); MBR: Membrane Bioreactor; HRT: 5-16 h; pH: 7; MLSS: 8000 mg/L

<sup>q</sup> (Jeswani and Mukherji 2013); Biodeg.: Biodegradation; COD: 1326 mg/L; NH<sub>3</sub>: 500-1500 mg/L

<sup>r</sup> (Taheri et al. 2012); SBR: Sequencing Batch Reactors; pH: 7.6-8.4; COD: 75-8400 mg/L

<sup>s</sup> (Chen et al. 2008); SBR: Sequencing Batch Reactors; SRT: 3.65-13.29 d; TSS: 46900-283400 mg/L

<sup>t</sup> (Carta-Escobar et al. 2005); AP: Aerobic Processes; COD: 3745 mg/L; VSS: 936 mg/L

<sup>u</sup> (Hao and Li 1987); ASP: Activated Sludge Process; BOD<sub>5</sub>:3100-5040 mg/L; COD: 630-2200 mg/L

### 3. Fouling assessment

In order to investigate the fouling performance of the MBRs, the steady-state fouling rates were determined at each SRT (15, 10 and 5 days) upon reaching steady-state conditions, (i.e. over the last SRT period of each run). Fouling rates for SRT of 20 days were not reported due to an unexpected failure in the pressure sensor. The steady-state membrane fouling rate, defined as the decline in specific flux (or slope of the specific flux line), was determined under steady-state conditions at 20°C. At SRT= 15 days, the steady-state fouling rate of the FS membrane was 1.76 LMH.bar<sup>-1</sup>.d<sup>-1</sup>, compared to 0.18 LMH.bar<sup>-1</sup>.d<sup>-1</sup> for HF membrane. As the SRT decreased to 10 and 5 days, the steady-state fouling rate increased to 1.99 LMH.bar<sup>-1</sup>.d<sup>-1</sup> and 2.38 LMH.bar<sup>-1</sup>.d<sup>-1</sup>, respectively, in the FS membrane (Figure 5) and to 0.22 and 0.27 LMH.bar<sup>-1</sup>.d<sup>-1</sup>, respectively, in the HF membrane (Figure 6).

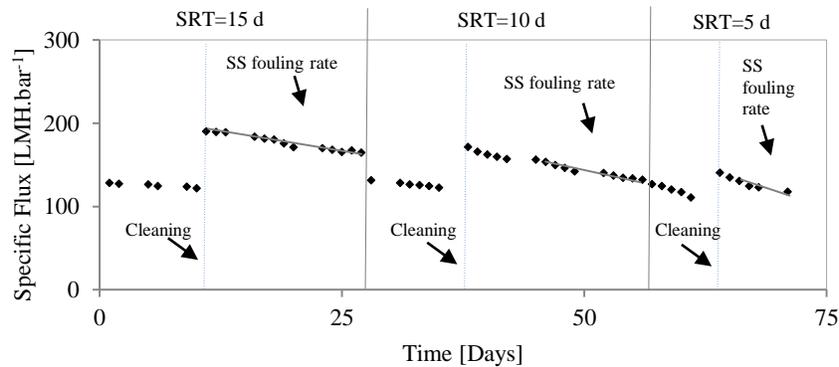


Figure 5 Flat Sheet Membrane fouling at SRT=15, 10, and 5 days

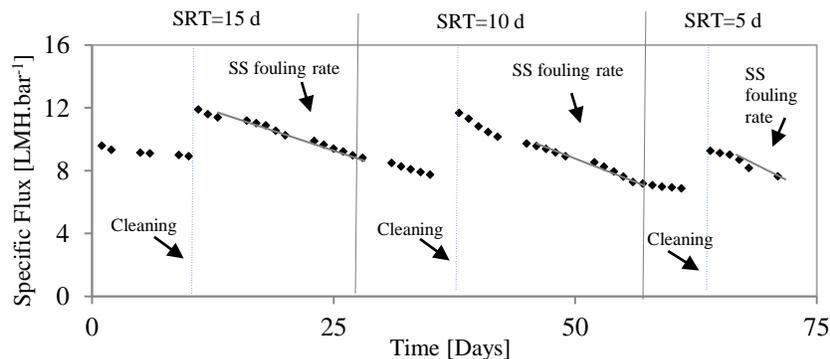


Figure 6 Hollow Fiber membrane fouling at SRT=15, 10, and 5 days

The steady-state fouling rate of both membranes increased almost linearly with decreasing SRTs (Figure 7). In general, a similar pattern was observed for both the FS and HF membranes with an increase in the steady-state fouling rate by a factor of 1.1 and 1.2, respectively, as the SRT decreased from 15 to 10 days and from 10 to 5 days respectively. These findings ascertain that SRT has a direct impact on the membrane fouling rate, consistent with reported studies (Trussel et al. 2006; Van den Broeck et al. 2012). In an experiment to investigate the effect of SRT on membrane fouling in MBR, Van den Broeck et al. (2012) reported higher membrane fouling rates 10.8 mbar.d<sup>-1</sup> at SRT 10 days as compared to 0.311 mbar.d<sup>-1</sup> at SRT 30 days and 0.187 mbar.d<sup>-1</sup> at SRT 50 days at 16 LMH membrane flux (Table 9). In addition, higher fouling rates were reported at SRT 30 days in comparison to SRT 50 days while operating at the same flux (3.37 mbar.d<sup>-1</sup> vs. 0.644 mbar.d<sup>-1</sup> at 22.5 LMH; 5.242 mbar.d<sup>-1</sup> vs. 0.821 mbar.d<sup>-1</sup> at 24 LMH; and 38.7 mbar.d<sup>-1</sup> vs. 11.15 mbar.d<sup>-1</sup> at 27 LMH at SRTs 30 days and 50 days, respectively). Similarly, Trussel et al. (2006) reported that the rate of membrane fouling in MBRs were higher at lower SRTs while operating at constant flux of 30.6 LMH (3.65 LMH.bar<sup>-1</sup>.d<sup>-1</sup> at SRT 2 days, 1.57 LMH.bar<sup>-1</sup>.d<sup>-1</sup> at SRT 3 days, 0.59 LMH.bar<sup>-1</sup>.d<sup>-1</sup> at SRT 4 days, 0.39 LMH.bar<sup>-1</sup>.d<sup>-1</sup> at SRT 5 days, and 0.18 LMH.bar<sup>-1</sup>.d<sup>-1</sup> at SRT 10 days) (Table 9). Extracellular polymeric substances (EPS) have been recognized as the main fouling components in MBRs and contribute to membrane biofouling ((Kimura et al. 2012), (Gao et al. 2013)). According to several studies, the higher the SRT, the lower the concentration of EPS as the biomass stays longer in the system, whereas at lower SRTs, the higher is the amount of EPS ((Ahmed et al. 2007), (Pan et al. 2010)). Thereby, fouling is more prominent at lower SRTs.

Table 9 Fouling rates for the FS and HF membrane bioreactors in comparison with literature reported values for various wastewater streams treated using an MBR system

Wastewater Type	Fouling rates ( $m^{-1}d^{-1}$ )	Permeate Flux (LMH)	COD influent (mg/L)	HRT (h)	SRT (d)	System	Reference
Municipal WW	$1.5 \times 10^{15}$	51	-	10	25	MBR	(Van der Marel 2009)
Synthetic WW	$0.5-10.5 \times 10^{12}$	15	700	-	45	MBR	(Bella et al. 2014)
Synthetic WW	0.03	0.60-0.75	1000	24	-	HMBR <sup>a</sup>	(Boonyungyuen and Wichitsathian 2014)
Synthetic WW	0.23					MBR	
Synthetic WW	9.14	0.60-0.75	1000	24	-	MBR	(Boonyungyuen et al. 2014)
	12.14	1-1.2		12			
	169.92	1.4-1.7		6			
Municipal WW	33.12-472.32	3-22	200	24	30	MBR	(Brookes et al. 2003)
Raw WW	230.4-3600	52	274.5	4		BF-MBR <sup>b</sup>	(Ivanovic and Leiknes 2008)
Synthetic WW	100.8-316.8	16-121	-	-	-	MBR	(Le-Clech et al. 2003)
Municipal WW	0.187	16	457	15-26	50	MBR	(Van den Broeck et al. 2012)
	0.625	20					
	0.644	22.5					
	0.821	24					
	11.15	27					
Municipal WW	0.311	16	457	15-26	30	MBR	(Van den Broeck et al. 2012)
	0.626	20					
	3.37	22.5					
	5.242	24					
	38.7	27					
Municipal WW	10.8	16	457	15-26	10	MBR	(Van den Broeck et al. 2012)
	<i>LMH.bar<sup>-1</sup>.d<sup>-1</sup></i>						
Municipal WW	0.18	30.6	345	-	10	SMBR <sup>c</sup>	(Trussel et al. 2006)
	0.39				5		
	0.59				4		
	1.57				3		
	3.65				2		
Landfill Leachate	1.76	5.4	5399	100	15	FS-MBR	This study
	1.99		5253		10		
	2.38		5218		5		
Landfill Leachate	0.18	0.58	5399	100	15	HF-MBR	This study
	0.22		5253		10		
	0.27		5218		5		

<sup>a</sup> Hybrid Membrane Bioreactor; <sup>b</sup> Biofilm Membrane Bioreactor; <sup>c</sup> Submerged Membrane Bioreactor

Yet, the steady-state fouling rate of the FS membrane was nearly 10 folds than that of the HF membrane for all tested SRTs (Figure 7), which can be attributed mainly to the higher flux in FS-MBR (5.4 LMH) compared to HF-MBR (0.58 LMH). Fouling is indeed expected to be higher for microfiltration-membranes (i.e. FS-MBR) than for ultrafiltration-membranes (i.e. HF-MBR) as a result of pore clogging by larger particles ((Choi et al. 2006)). Greater initial fouling for the larger pore-size membranes have equally been reported when smaller pore-size membranes were

used over an extended period of time (Gander et al. 2000). In addition, the hydrophobicity of the FS material (compared to the hydrophilic nature of the HF surface) can aggravate fouling. In fact, hydrophobic interaction between solutes and membrane material is one of the predominant fouling mechanisms (Maximous et al. 2009; Sun et al. 2006; Yu et al. 2005). As a result, the fast decline in the filtration rate with strong fouling are more often observed in hydrophobic than hydrophilic membranes (Fane and Fell 1987; Hilal et al. 2005; Kim et al. 1991; Kulkarni et al. 1992; Toyomoto and Higuchi 1992). However, changes in the membrane hydrophobicity often occur with modifications other than membrane properties such as pore size and morphology, which renders the correlation between membrane hydrophobicity and fouling more difficult to assess (Maximous et al. 2009; Pinnau and Freeman 2000). This necessitates more characterization of membrane fouling with a particular emphasis on the interaction of foulants with the membrane material (Maximous et al. 2009).

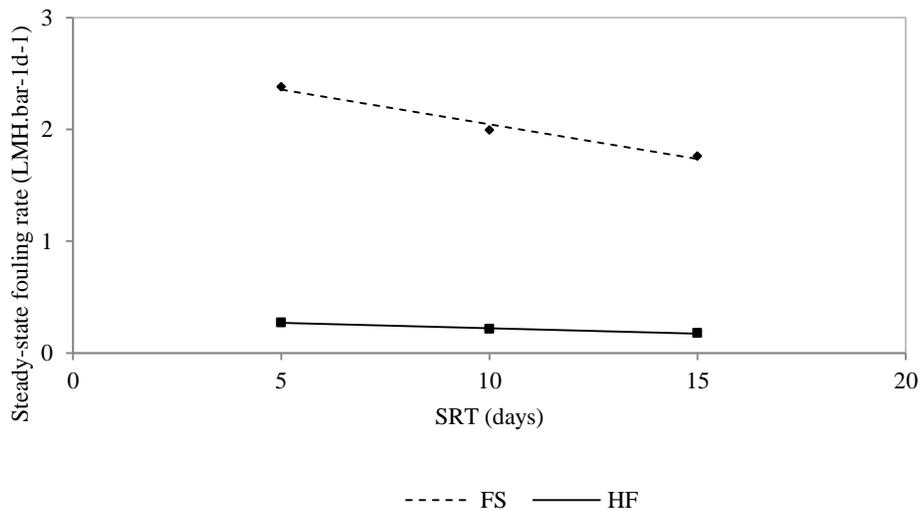


Figure 7 Effect of SRT on steady-state membrane fouling rate of FS and HF membranes

#### 4. Microbial characterization

##### *Bacterial community composition in the mixed liquor*

The results of the 16S rRNA gene sequencing revealed that *Proteobacteria* (42.6%), *Bacteroidetes* (23.7%), and *Firmicutes* (13.5%) were the dominant phyla detected in the mixed liquor of both MBRs (*i.e.* FS and HF) (Figures 8 and 9). The number in parentheses corresponds to the average relative abundance for the whole duration of the experiment and includes both the oxic and anoxic tanks. While no work has been reported on microbial composition in anoxic-oxic MBRs treating landfill leachate, studies on the treatment of municipal wastewater reported the dominance of *Proteobacteria* and *Bacteroidetes* in aerobic MBR systems operated at SRTs ranging between 3 and 60 days ((Su et al. 2011); (Wan et al. 2011); (Duan et al. 2009)). (Su et al. 2011) reported the dominance of *Proteobacteria* and *Bacteroidetes* in the suspension of MBR at SRT 10 and 60 days treating concentrated synthetic municipal wastewater of pH 7. Likewise, (Wan et al. 2011) reported the dominance of *Proteobacteria* and *Bacteroidetes* in the MBR treating municipal wastewater of influent concentrations: COD= 220 mg/L, TN= 26 mg/L, NH<sub>4</sub><sup>+</sup>-N= 19 mg/L, TP=3.9 mg/L. (Duan et al. 2009) also reported the dominance of *Proteobacteria* and *Bacteroidetes* in the MBR at SRT 3, 5 and 10 days treating synthetic wastewater with influent COD and ammonia concentrations of 182 and 35 mg/L, respectively.

The relative abundance of the dominant phyla in the mixed liquor, of the anoxic and oxic tanks, varied with respect to time and type of MBR (*i.e.* FS vs. HF). In addition, it is noticeable that the change in the evolution of each of these phyla in one MBR was very often accompanied by similar behavior in the other MBR. For instance, in the FS-MBR, the proportions of *Proteobacteria* increased from 38.3% to 46.3% when varying the SRT from 20 to 15 days; similar-wise, in the HF-MBR the proportions of *Proteobacteria* also increased from 43.6% to 53.2% at the

corresponding SRTs. In addition, the proportions of this phyla decreased, in the FS-MBR, from 46.2% to 40.3% when the SRT was varied from 15 to 10 days; likewise, in the HF-MBR, the proportions decreased from 53.2% to 39.2% at the corresponding SRTs. Moreover, the proportions of *Bacteroidetes* in the FS-MBR slightly increased from 19% to 22% when moving from SRT=20 days to SRT=15 days; likewise, the proportions of *Bacteroidetes* in the HF-MBR slightly increased from 15.8% to 16.2% at the same SRTs. Likewise, when the SRT was varied from SRT=15 days to SRT=10 days, the proportions of *Bacteroidetes* in the FS-MBR increased from 22% to 34.2% and in the HF-MBR from 16.2% to 23.2%. Furthermore, the proportions of *Firmicutes* increased from 11.2% to 14.2% when varying the SRT from SRT=15 days to SRT=10 days; similar-wise, in the HF-MBR the proportions of *Firmicutes* showed a slight increase from 20.4% to 21.3% at the corresponding SRTs. When the SRT was varied from SRT=10 to SRT=5 days, the proportions of *Firmicutes* slightly decreased from 14.2% to 12.9% in the FS-MBR; similarly, the proportions of this phyla decreased from 21.3% to 16.1% at the corresponding SRTs.

Comparing the proportions of these phyla between both MBRs, it is noticed that at SRT=15 days, *Proteobacteria* was at its highest among all the other SRTs with 46.3% and 53.2% in the FS-MBR and HF-MBR respectively. Likewise, at SRT=10 days, *Firmicutes* was at its highest in both MBRs among all the other SRTs with 14.2% and 21.3% in the FS-MBR and HF-MBR respectively. As for *Bacteroidetes*, the proportions of this phyla were at its lowest at SRT= 20 days with 19% and 15.8% in the FS-MBR and HF-MBR respectively. The similar behavior of these phyla in the mixed liquor of both MBRs and the fact that thier evolution was not consistent with the decrease in SRT drives us to conclude that the changing leachate composition is responsible for the phyla's behavior regardless of the SRT. At the class level, *Alphaproteobacteria* (5-23%), *Betaproteobacteria* (7-21%), *Gammaproteobacteria* (5-38%) and *Flavobacteria* (2-33%) were the

dominant classes in the oxic tanks of both systems, at all SRTs tested (Figure 8). The anoxic tanks were dominated by *Betaproteobacteria* (3-11%), *Gammaproteobacteria* (17-39%) and *Clostridia* (12-22%) (Figure 9). It was also noticeable that the change in the evolution of each of these classes in one MBR was very often co-occurring by comparable behavior in the other MBR. For instance, in the oxic tanks of the FS-MBR, the proportions of *Gammaproteobacteria* decreased from 13.5% to 10.2% to 8.9% when varying the SRT from SRT=15 days to SRT=10 days to SRT=5 days; similar-wise, in the HF-MBR compartment the proportions of *Gammaproteobacteria* also decreased from 32.2% to 23.1% to 22.6 at the corresponding SRTs. This decrease in the *Gammaproteobacteria* proportions was also noticeable in the anoxic tanks with a decrease from 25.2% to 23.5% for the FS-MBR and from 30.4% to 23% in the HF-MBR when SRT was varied from 15 to 10 days. In addition, the proportions of *Alphaproteobacteria* in the oxic tank of the FS-MBR increased from 10.3% to 22.7% when varying the SRT from 20 to 15 days; likewise, the proportions of this class showed an increase from 4.6% to 8.6% at the equivalent SRTs. As for the class *Flavobacteria*, the proportions in the FS compartment showed a sharp increase from 17.5% to 29.9% as SRT was varied from 15 to 10 days and similar-wise from 2.2% to 15.4% in the HF compartment at the corresponding SRTs. As for *Betaproteobacteria* in the anoxic tanks, the proportions of this class showed a slight increase from 8.5% to 9% in the FS-MBR and from 7.9% to 11.5% in the HF-MBR as SRT was varied from 20 to 15 days. The evolution of *Clostridia* was comparable across the anoxic tanks of both MBRs with a slight increase from 12.7% to 13% and from 21.4% to 22.1% across the FS-MBR and the HF-MBR, respectively as SRT varied from 15 to 10 days and with a slight decrease into 12% and 17% across the FS-MBR and the HF-MBR, respectively as SRT reached 5 days.

### *Bacterial community composition in the biofilm*

Concerning the biofilm samples, the results of the 16S rRNA gene sequencing showed that *Proteobacteria* and *Bacteroidetes* were the dominant phyla, representing more than 60% of the bacterial community at the surface of both membranes (Figure 10). Studies showed similar dominance of these phyla on the membrane surface of MBRs. (Huang et al. 2008) reported the dominance of *Proteobacteria* and *Bacteroidetes* in the biofilm communities of MBR treating municipal wastewater at SRTs 8 and 30 days. Likewise, (Lim et al. 2012) reported the dominance of *Proteobacteria* in the biofilm communities of MBR treating synthetic wastewater of influent COD concentration of 1200 mg/L at SRT 30 days. Specifically, it was suggested that *Bacteroidetes* are major contributors to the development of membrane biofilms and could have a competitive advantage over other colonizers ((Huang et al. 2008)). However, it was noticeable that the evolution of these dominant phyla was not accompanied by a similar behavior in the parallel MBR. For instance, *Bacteroidetes* proportions decreased steadily on the FS membrane with decreasing SRT (from 44.7% at SRT=20 days reaching 18.6% at SRT=5 days). In contrast, the *Bacteroidetes* proportions on the HF membrane remained in the narrow range of 20% to 24 %, irrespective of the adopted SRT. On the other hand, the *Proteobacteria* varied, in a fluctuating manner with respect to SRT, between 35.5% and 51% on the HF membrane and between 38% and 46% on the FS membrane. At the class level, *Alphaproteobacteria* (16 to 25%), *Gammaproteobacteria* (11 to 18%) and *Flavobacteria* (4 to 20%) were among the dominant classes of the biofilm microbial communities of both MBRs at all SRTs tested.

In general, the biofilm communities seem to be more stable to the change in SRT compared to the mixed liquor communities. (Huang et al. 2008) compared the biofouling communities of identical membranes operated under 15 and 30 LMH and different SRTs of 8 and 30 days and

concluded that the imposed membrane flux affected the community structure and composition of biofouling microorganisms. The low-flux (i.e. 15 LMH) biofilm communities from two MBRs operated at different SRTs were related whereas distinct biofilm communities developed on the high-flux MBRs operated at different SRTs. Also, the biofilm microbial communities were significantly different between the same SRT MBRs operated at different fluxes. (Huang et al. 2008) attributed this difference between the low and high-flux to the strong convective force that transports bacterial cells towards the membrane surface at higher permeate flux. This can explain the lack of a makeable differences in the composition of biofouling communities at different SRTs in our study, given the low fluxes (5.4 LMH for the FS and 0.58 LMH for the HF) used.

#### *Beta diversity measures*

The bacterial communities were compared using non-phylogenetic measures (Bray-Curtis distance). Beta diversity indicates the partitioning of microbial diversity; thus the relatedness of the bacterial communities in the mixed liquor and biofilm in the different MBRs (FS vs HF) were assessed at different SRTs. Despite the fact that both MBRs were operated under the same conditions and fed with the same leachate, hierarchical clustering and NMDS plot revealed that mixed liquor community from the FS-MBR were clustered separately from the mixed liquor community in the HF-MBR (Figures 11 and 12). Similarly, the biofilm community on the HF membranes was different from the biofilm community on the FS membranes. Also, the bacterial community structure was dynamic as can be seen by NMDS analysis with gradual succession away from initial conditions (i.e. SRT 20) (Figure 12). This dynamic was more pronounced in the HF-MBR than FS-MBR. Moreover, the mixed liquor community was more dynamic than the biofilm community as can be seen by their wide distribution in the NMDS plot (Figure 12). Similar to previous studies in lab-scale MBRs ((Lim et al. 2004); (Lee et al. 2014); (Zhang et al. 2006);

(Huang et al. 2008; Lim et al. 2012)) , the biofilm microbial community was distinct from the mixed liquor community where they formed a separate cluster in the dendrogram and NMDS plot (Figures 11 and 12).

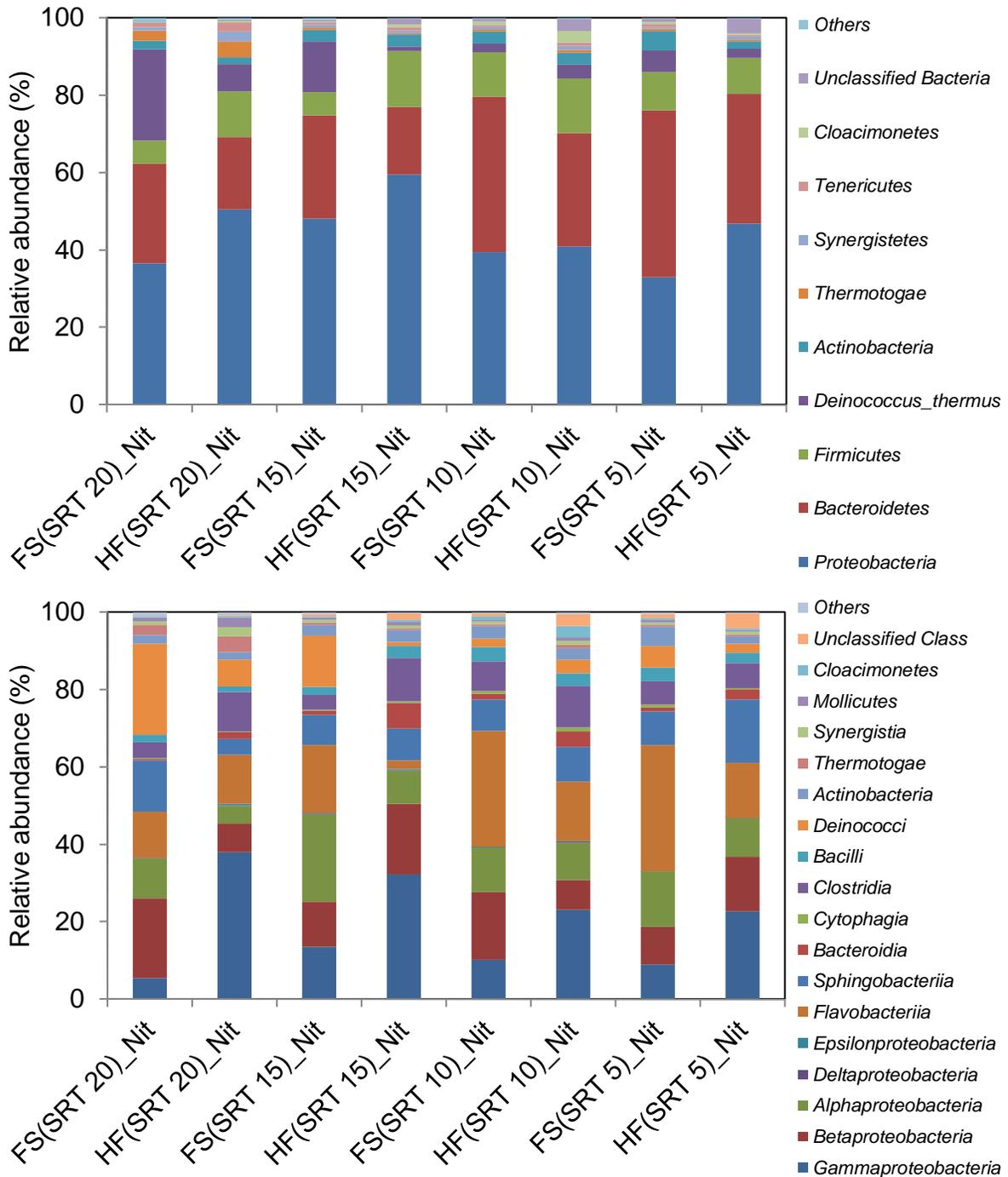


Figure 8 Relative abundance of bacterial reads retrieved from nitrifying (oxic) tank of the hollow fiber (HF) and flat sheet (FS) MBR classified at the (a) phylum and (b) class level. Taxa that represent less than 1% of the total bacterial community composition were classified as “others”.

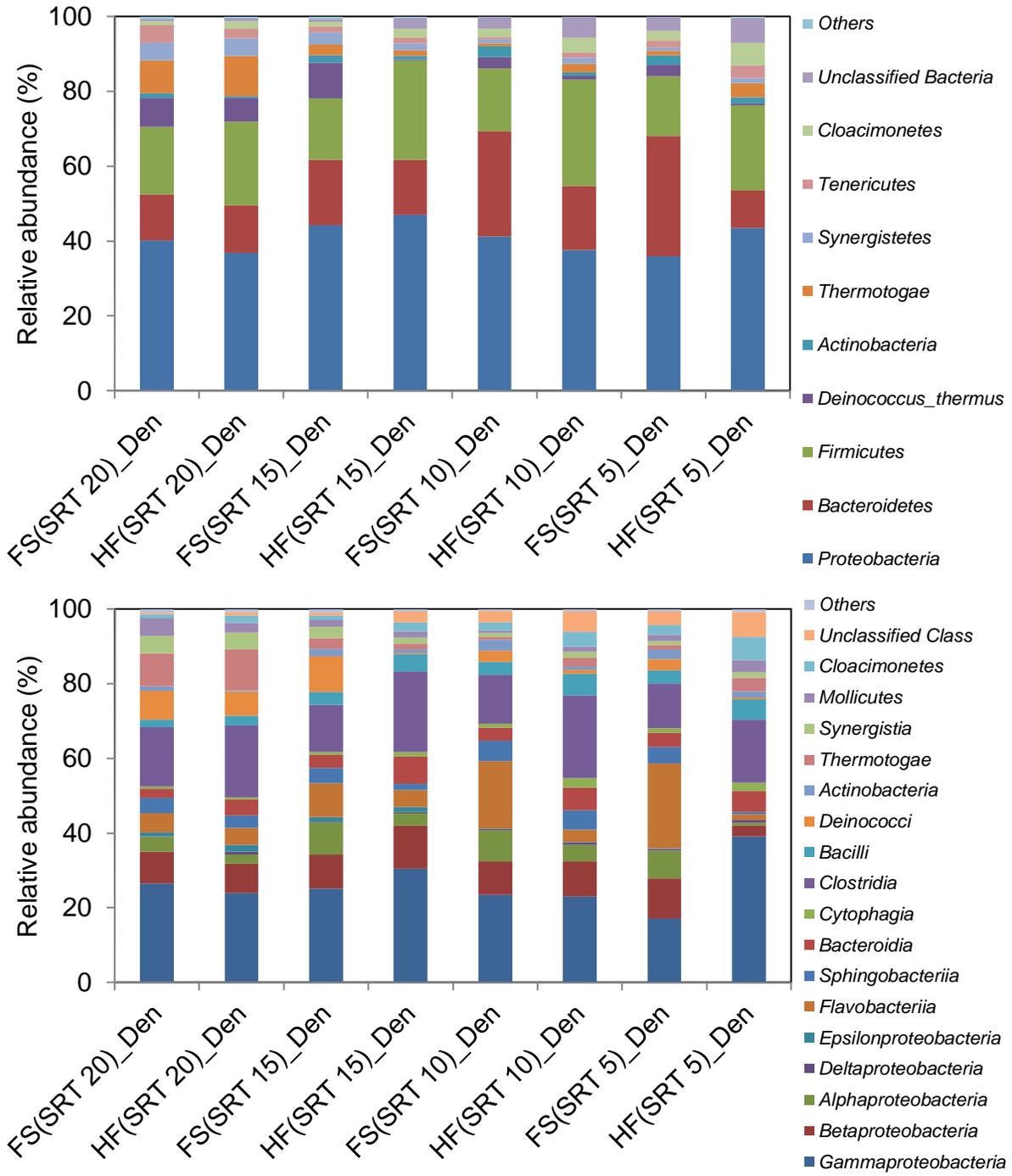


Figure 9 Relative abundance of bacterial reads retrieved from the denitrifying (anoxic) tank of the hollow fiber (HF) and flat sheet (FS) MBR classified at the (a) phylum and (b) class level. Taxa that represent less than 1% of the total bacterial community composition were classified as “others”.

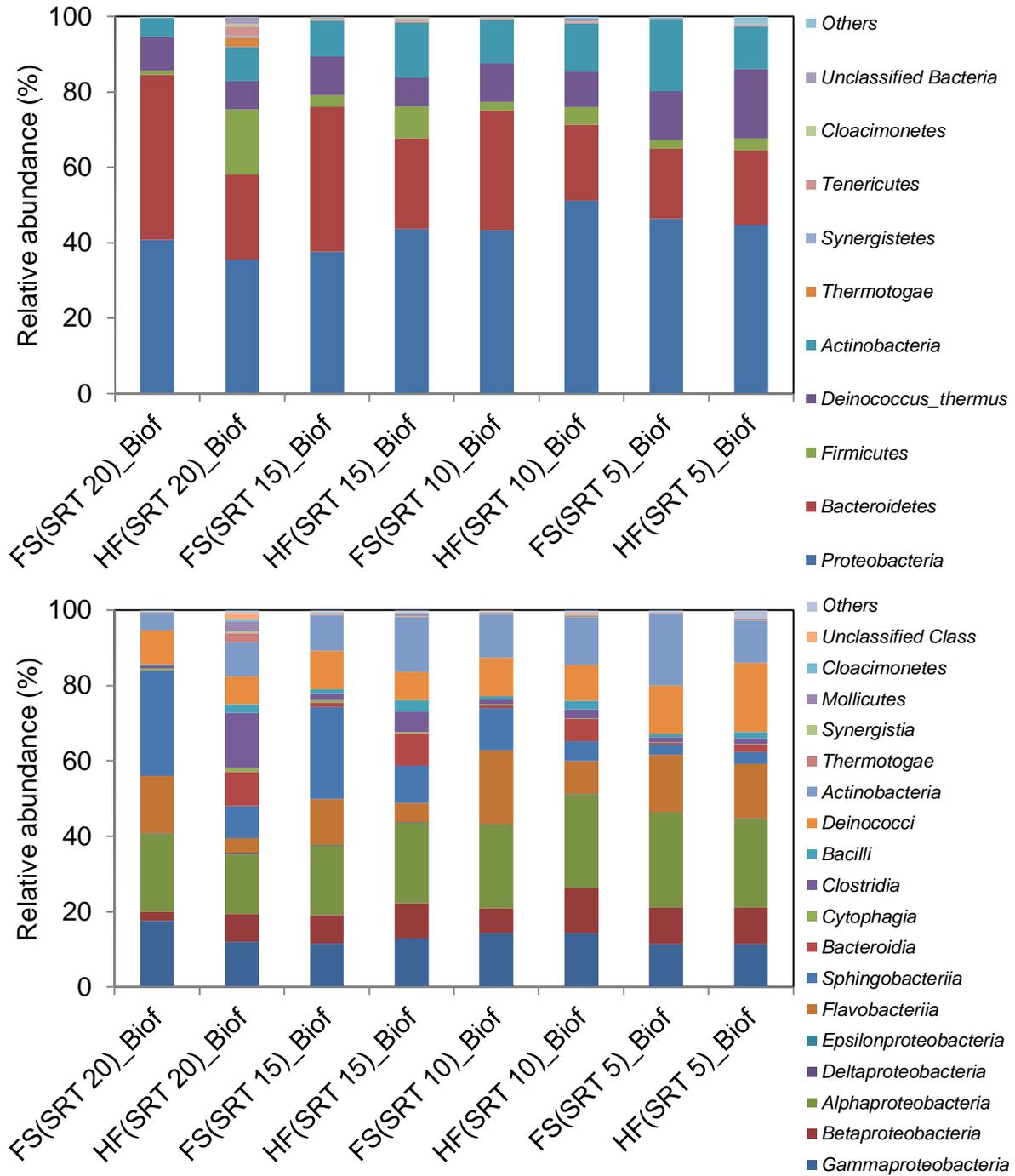


Figure 10 Relative abundance of bacterial reads retrieved from biofouled membranes classified at the (a) phylum and (b) class level. Flat sheet: FS; Hollow fiber: HF. Taxa that represent less than 1% of the total bacterial community composition were classified as “others”.

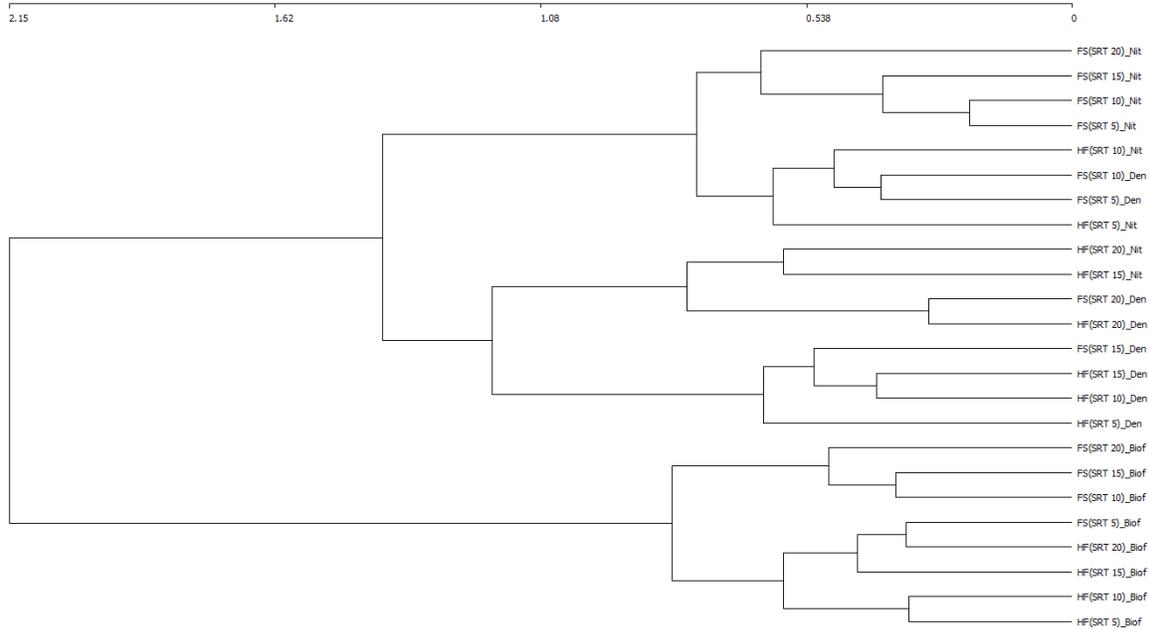


Figure 11 A dendrogram generated based on Bray-Curtis distance at 3% cutoff-OTU level showing the % dissimilarity in bacterial community structure between samples collected from the nitrifying (Nit) and denitrifying (Den) tanks, and membrane surface (Biof) of the hollow fiber (HF) and flat sheet (FS) MBR.

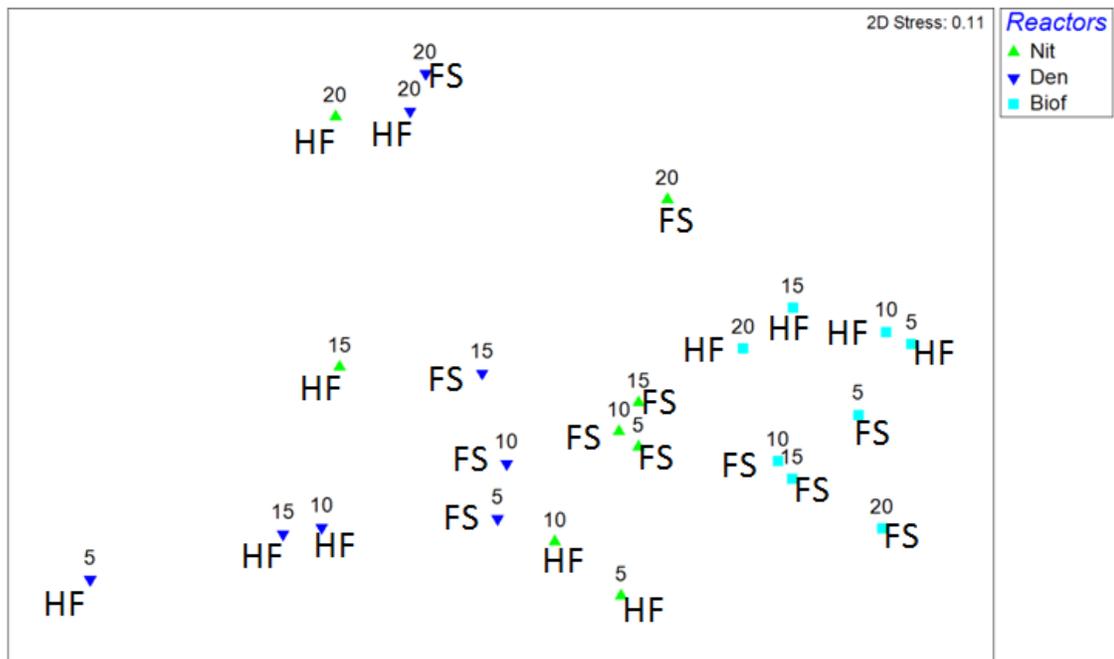


Figure 12 Nonmetric multidimensional scaling (NMDS) plot based on based on Bray-Curtis distance at 3% cutoff-OTU level for samples collected from the nitrifying (Nit) and denitrifying (Den) tanks, and membrane surface (Biof) of the hollow fiber (HF) and flat sheet (FS) MBR. The numbers on the symbols represent the different SRT values: 20, 15, 10 and 5 days, respectively.

## CONCLUSION

The comparative assessment of the FS-MBR and HF-MBR showed that both MBRs exhibited comparable removal efficiencies for BOD<sub>5</sub> (92% in FS vs. 90% in HF), TP (72% in FS vs. 66% in HF), PO<sub>4</sub><sup>3-</sup> (74% in FS vs. 75% in HF) and COD (51% in FS vs. 43% in HF) at SRT = 20 days. Better TN removal was achieved with the FS-MBR at SRT = 20 days (35% in FS vs. 15% in HF). The performance of both systems degraded at shorter SRTs, with the FS system exhibiting better overall nitrogen removal although both MBRs received similar influent feed and were operated under the same configuration with anoxic/oxic units and under the same HRT and SRT. The statistical analysis showed that the removal efficiencies of TP, PO<sub>4</sub><sup>3-</sup>, COD, and BOD<sub>5</sub> were a function of SRT for both membranes (p-value <0.05); the TN removal also varied as a function of SRT for the HF-MBR (p-value <0.05) but not for the FS-MBR (p-value=0.3).

The biokinetic coefficients governing FS and HF values were estimated with K<sub>s</sub> (mg COD/L) (1123.63 and 1160.30 for the FS-MBR and HF-MBR, respectively) and μ<sub>m</sub> (day<sup>-1</sup>) (1.42 and 1.72 for the FS and HF, respectively) falling within reported ranges for activated sludge processes (ASP) and MBR applications treating wastewater; yet both Y (mg VSS/mg COD) (1.04 and 1.87 for the FS-MBR and HF-MBR, respectively) and k<sub>d</sub> (day<sup>-1</sup>) (0.99 and 1.24 87 for the FS-MBR and HF-MBR, respectively) were higher than reported values.

The fouling assessment showed that the steady-state fouling rate of both membranes increased, almost linearly, with the decrease in SRT. A similar pattern was observed in the FS and HF membranes with an increase in the steady-state fouling rate at a factor of 1.1 and 1.2, respectively, when the SRT was reduced from 15 to 10 days and from 10 to 5 days.

The microbial analysis showed that similar dominant phyla were detected in both MBRs with *Proteobacteria*, *Bacteroidetes*, and *Firmicutes* constituting around 80% of the oxic and

anoxic mixed liquor community. *Bacterioidetes* and *Proteobacteria* were also the dominant groups in the biofilms, forming together more than 60% of the bacterial community at the surface of both membranes. Hierarchical clustering and NMDS plot revealed that the mixed liquor community in the FS-MBR were clustered separately from the mixed liquor community in the HF-MBR. Similarly, the biofilm community on the HF membranes was different from that on the FS membranes. The bacterial community structure was found to be dynamic, especially for the FS MBR. Overall, the mixed liquor community appeared to be more dynamic than the biofilm community.

The above findings suggest that membrane bioreactors cannot be conveniently operated at low solid retention times (< 20 days) without drawbacks in terms of removal efficiencies of carbon and nutrients. The absence of nitrifiers in the system resulted in low nitrogen removal. It is suggested that nitrifiers were not enriched due to the high organic content of the leachate. Moreover, the effect of high nitrogen concentrations in the leachate requires pretreatment or post treatment as high nitrogen removals are challenging with MBRs alone.

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