MBR treatment of leachates originating from waste management facilities: a reference study of the design parameters for efficient treatment

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Abstract:

The main objective of the study was to define the interaction between the solid retention time (SRT) and the contaminant loading rate on a membrane bioreactor's efficacy in removing contaminants frequently detected (chemical oxygen demand (COD), NH_4^+, total phosphorus and metals) above the discharge criteria in waste-originating leachates. The rates and coefficient calculated from this study's experimental data can be used for the design of membrane bioreactor treating wastewaters, even beyond the scope of this experiment. Over a period of 152 days, SRTs of 28 and 47 days and HRTs of 13, 25, 36 and 52 hours were studied using a real leachate with a constant composition. Results showed that membrane bioreactors can efficiently treat > 1 850 mg COD L^{-1} d^{-1} of highly to moderately biodegradable COD, with the SRT having no significant impact on the removal of recalcitrant COD. Overall ammonium removal rates of > 740 mg NH_4-N L^{-1} d^{-1} can be achieved as long as a residual alkalinity of 200 mg CaCO3 L-1 and an adequate dissolved oxygen concentration (6-7 mg L^{-1}) are both maintained. Overall phosphorus removal rates are independent of the phosphorus loading rate. However, the highest overall phosphorus removal rate (39±2 mg P per g of total suspended solids) was obtained at the lowest SRT (28 days) due to an increased extracellular polymeric substance production. Finally, membrane bioreactor's metal removal capacity is mostly dependent on the metals' affinity to both the leachate's recalcitrant COD as well as sludge concentrations.

Keywords: membrane bioreactor, leachate treatment, nitrification, process design, extracellular polymeric substances

Declarations of interest: none

List of abbreviations

AOB	Ammonia oxidizing bacteria
BOD_5	Biochemical oxygen demand (measured over a period of 5 days)
CAS	Conventional activated sludge
COD	Chemical oxygen demand
DO	Dissolved oxygen
EPS	Extracellular polymeric substances
HB	Heterotrophic bacteria
HRT	Hydraulic retention time
MBR	Membrane bioreactor
MVLSS	Mixed liquor volatile suspended solids
MW	Molecular weight
NLR	Ammonia loading rate
OLR	Organic loading rate
PLR	Phosphorus loading rate
SRT	Solid retention time
TSS	Total suspended solids

1. Introduction

Despite the increase in environmental concerns regarding the global generation of waste, trends still forecast a growth over the next decades. Currently, sanitary landfills remain the preferred option compared to other waste management strategies such as incineration or composting, since they are the most economical option in terms of capital and exploitation costs [1].

Even though waste management improvements are being made, the amount of leachates originating from these wastes will continue to increase. Waste-originating leachates are considered highly complex wastewaters and are generally characterized by high concentrations of ammonium (NH_4^+) and recalcitrant dissolved organic substances (e.g. fulvic- and humic-like acids) [2]. Depending on the type of waste from which the leachates originate, they can also contain hazardous concentrations of heavy metals, phosphorus, [1, 3] and micropollutants [4]. For example, Fromme et *al.* (2002) reported concentrations of 17 – 26 µg Bis(2-ethylhexyl) phtalate L-1 and 25 – 146 µg Bisphenol-A L-1 in mixed municipal solid waste composting leachate [5]. These leachates therefore have potential adverse effects on the environment, and thus need to be properly collected and treated prior to disposal [6-8].

Conventional approaches for the waste-originating leachates consists in their transportation to the municipal sewage treatment system (co-treatment), or their on-site treatment by a conventional activated sludge (CAS) process or aerated lagoons. With the co-treatment approach, studies have reported significant biological treatment inhibition caused by high ammonium loading rates from the leachate [9, 10]. With the CAS or the aerated lagoons processes, the extensive hydraulic retention times (HRT) (generally a few days to a few weeks), combined with the large amount of chemical products required to meet the more stringent new regulations, makes these option less enticing. The co-composting facility, from which the leachates used in this study originates, operates an aerated lagoon with a HRT of 22 days and uses up to 195 g Ferric sulfate m^{-3} of treated leachate. An emerging process that addresses these issues and has gained interest over the last decade is the membrane bioreactor (MBR). By replacing the secondary clarifier from the CAS by a membrane, MBRs have the advantage of producing better effluent quality, and have process stability, independent control of the HRT and the solid retention time (SRT), and increased biomass or mixed liquor suspended solids [1, 11]. The increased biomass in MBRs generally leads to significantly increased contaminants removal rates when compared to CAS or lagoons systems, and consequently, a smaller footprint [8]. Furthermore, high SRTs allow the enrichment of slow-growing bacteria such as autotrophic nitrifiers, which are beneficial to leachates containing high concentrations of ammonium [12]. In a recent study, Vuono et *al.* (2016) suggested that high SRTs can also lead to higher rRNA expressions for rare taxa and increase the removal of micropollutants [13].

Several previous studies reported promising leachate treatment results with the application of the MBR process. There results are compared to those obtained in this study in Table 1. However, it is challenging to extrapolate results from one study to another due the lack of information regarding the operating parameters (e.g. dissolved oxygen (DO), HRT, SRT, and mixed liquor volatile suspended solids (MLVSS)) and their effect on the biokinetic parameters. This lack of information is evidenced in Table 1 by the work of Garcia-Lopez et *al.* (2014)[14] and Xue et.*al.* (2015) [15] that published results on MBR treating leachates without sufficient data to calculate the contaminants removal rates. Then Sadri et *al.* (2008) studied three different combinations of HRT and SRT, but their results were not sufficient to conclude on their specific effect on the MBR's treatment capacities [16]. Similarly, Babatsouli et *al.* (2015) studied 4 combinations of HRT and SRT. Due to compositional changes in their feed, all contaminant loading rates remained in the same range, preventing any strong conclusion of the effect of the HRT and SRT on process performance [17].

Furthermore, a majority of the previous studies on leachate-treating MBRs reported contaminant removal efficiencies in terms of percentage notwithstanding their loading rates, which can lead to a misinterpretation of the process capacities. For example, Duan et *al.* (2009) qualified a 87 – 90% ammonia removal efficacy as very good; however, this merely represented an ammonia removal rate of approximately 47 mg NH_4-N L^{-1} d^{-1} [11]. For comparison, Sadri et *al.* (2008), reported an ammonia removal rate of 670 mg NH_4-N L^{-1} d^{-1}, with a corresponding 99% removal [16]. Another current practice leading to misinterpretation is the expression of loading rates or removal rates per MLVSS concentration, while the leachate contains multiple contaminants at high concentrations. For example, Hasar et *al.* (2009) expressed the ammonium loading rates in terms of gN g $MLVSS^{-1}$ d^{-1}, while the mixed liquor is a combination of heterotrophic and autotrophic microorganisms whose concentration depends on both the ammonium loading rate (NLR) and the organic loading rate (OLR) [18]. These examples highlight the need for a standardized methodology for reporting treatment efficiencies when working with leachates in order to provide replicable data for MBR process design.

To fill the current gap of knowledge regarding the performance of leachate-treating MBRs, the main objective of this study was to define the effect of the SRT and the contaminant loading rate on the MBR's efficacy of contaminant removal. Experiments were conducted at SRTs of 28 and 47 days and HRTs of 13, 25, 36 and 52 hours using a real leachate with a constant composition. Due to their frequent detection above the standard limits in leachates, the following contaminants were studied: chemical oxygen demand (COD), biochemical oxygen demand (BOD_5), NH_4^+, total phosphorus, and dissolved metals. Furthermore, this study aims to provide a standardized methodology to analyze and report contaminant removal efficacies. A standardized methodology will facilitate the comparison of future leachate treatment studies from one to another. Finally, the study will be a reference for scientists and engineers undertaking the design of leachate-treating MBRs by providing useful tools and data to predict removal efficacies.

Table 1 Comparison between Previous studies' treatment and this study's performance of membrane bioreactors treating waste-originating leachate

					COD		NH4		
MBR configuration	Wastewater	SRT	HRT	MLVSS	(OLR)	Removal	NLR	removal	
		(d)	(hr)	(g L^{-1})	(mg COD L^{-1} d^{-1})	(mg COD L^{-1} d^{-1})	(mg NH4-N L^{-1} d^{-1})	(mg NH4-N L^{-1} d^{-1})	Authors
Literature data									
Submerged	-	-	6.0 - 12.9	-	900 – 4 000	2 800	160 - 240	238	[6]
Submerged (thermophilic)	Medium aged LL	-	24	9 000	12 000	9 480	1000 - 1800	1080	[19]
Submerged	Old LL	60	84	4920	900	600	180	180	[16]
		60	48	9900	2 040	1590	320	320	
		30	24	5300	2 740	1480	670	670	
Submerged	Synthetic solution	10	16	822	270	260	53	47	[11]
		5	16	480	270	258	53	46	
		3	16	260	270	257	53	47	
Submerged	Composting leachate	-	2 280	-	1 200	1195	29	29	[8]
Submerged	Synthetic solution	30	8	7600	3 000	2940	150	136	[20]
		30	8	8300	3 000	2910	300	268	
External	Composting leachate	-	-	-	-	-	-	-	[14]
Submerged	Pre-treated Leachate	-	-	-	-	-	-	-	[15]
Side-stream	Pre-treated Leachate	-	-	-	-	-	-	-	[15]
Results from this study									

Submerged	Composting leachate	28	52	2 680	1 515	683	191	189	
		27	36	3 200	1 935	853	269	268	
		29	25	4 080	2 475	883	397	397	
		27	13	6 990	5 020	1846	742	740	
		46	25	6 790	2 340	806	382	381	
		47	13	8 780	3 970	923	740	736	-

2. Materials and methods

2.1. Lab-scale submerged membrane bioreactor

The lab-scale submerged MBR designed for this experiment is shown schematically in Figure 1.

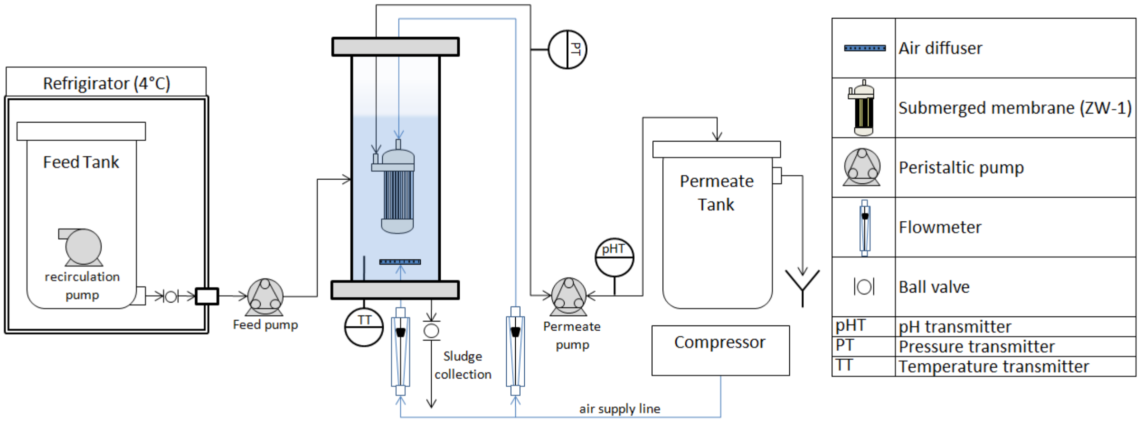


Figure 1 Schema of the lab-scale submerged membrane bioreactor

The leachate was kept in a 50 L polyethylene tank placed in a refrigerator at 4°C in order to avoid any biological activity that could alter its composition. The 10 L reactor was made of a 146.33 mm I.D. clear PVC tube (schedule 80). The leachate was fed into the reactor through a side port using a peristaltic pump (Masterflex, model #7528-10). An ultrafiltration hollow-fiber membrane module (Zee-Weed, ZW-1) was used. The ZW-1's specifications are presented in Table 2.

Table 2 Zee-Weed ZW-1 membrane module specifications

Nominal pore diameter (µm)	0.04
Fiber diameter (mm)	2
Membrane surface area (m^2)	0.047
Maximum transmembrane flux ($L\ m^{-2}\ h^{-1}$)	32

The permeate flow-rate (filtration: 300 s, flux 7.4 $L\ m^{-2}\ h^{-1}$; backwash: 20 s, flux 46.4 $L\ m^{-2}\ h^{-1}$) was maintained constant during each experiment by controlling the vacuum applied at the membrane (5 to 50 kPa) with a peristaltic pump (Masterflex, model #7528-10). To prevent the accumulation of a cake layer at the membrane surface filtration/backwash cycles were applied and air was introduced between the membrane fibers through a perforated pipe built in the module (2.5 L air min^{-1}). Membrane module cleaning was conducted when the transmembrane pressure reached -50 kPa. The cleaning procedure was the following: 1) 3 h of dynamic oxidative cleaning (in a close loop recirculation) with a 1000 mg L^{-1} active chlorine solution (NaOCl), 2) rinsing for 5 min with distilled water,

3) 1.5 h of dynamic acidic cleaning (in a close loop recirculation) with a 3 g L^{-1} citric acid solution, and 4) rinsing for 5 min with distilled water.

The sMBR was operated in oxic conditions (D.O. = 6 - 7 mg O_2 L^{-1}) at room temperature (20±1°C). Compressed air was introduced through a perforated tube placed at the bottom in the reactor (2.5 L min^{-1}) and arranged in such way as to create a homogeneous internal mixing of the reactor from the rising bubbles. SRT was controlled by collected mixed liquor samples on a daily basis.

2.2. Experimental procedure

Over the course of the experiment's 152-day duration, 6 different combinations of 2 SRTs (approx. 28 and 47 days) and 4 HRTs (approx. 13, 25, 36, and 52 hours) were tested. The details of the experimental conditions are presented in Table 3.

Table 3 Operating parameters of the sMBR (average values)

Parameters	Condition #1	Condition #2	Condition #3	Condition #4	Condition #5	Condition #6
Period length (d)	50	21	22	20	21	18
SRT (d)	28 ± 4	27 ± 4	29 ± 4	27 ± 5	46 ± 2	47 ± 3
HRT (hr)	52 ± 1	36 ± 3	25 ± 0.3	13 ± 1	25 ± 1	13 ± 0.3
TSS (mg L^{-1})	3 580 ± 170	4 210 ± 95	5 520 ± 254	11 087 ± 232	11 105 ± 294	12 890 ± 156
MLVSS (mg L^{-1})	2 680 ± 112	3 200 ± 90	4 080 ± 204	6 990 ± 303	6 790 ± 246	8 780 ± 28
MLVSS/TSS	0.75	0.76	0.74	0.63	0.61	0.68
NLR (g NH_4-N m^{-3} d^{-1})	191	269	397	742	382	740
OLR (g COD m^{-3} d^{-1})	1 514	1 933	2 477	5 022	2 342	3 969
PLR (g P m^{-3} d^{-1})	6.7	10.1	14.6	27.5	13.1	19.1

The duration of each experimental condition was at least 18 days in order to reach steady-state conditions and to provide an adequate duration for sample collection. Once steady-state was achieved (14 days), feed, mixed liquor, and permeate samples were collected for analysis on a daily basis.

2.3. Leachate origin and characterization

The leachate used in this study was taken in the retention basin (RB1) of an open organic waste co-composting (67% green waste, 18% industrial sludge, 8% municipal sewage sludge and 7% paper mill sludge) facility located in the province of Quebec (Canada) during the month of October[21]. In order to maintain a constant feed composition for each different tested condition, a 1 m^3 leachate sample was collected and stored in a cold room at 4°C. Then, the sMBR's feed tank was filled from that 1 m^3 leachate sample. Table 4 summarizes the compositional ranges (minimum and maximum values) obtained from all the MBR's feed samples taken during the experiment. Table 4 also compares these compositional ranges to those reported in the literature [22] and to the acute exposure US-EPA criteria for aquatic life [23].

Table 4 Organic waste co-composting leachate composition

Parameter	**Units**	**US-EPA Criteria [23]**	**Literature (min. - max.) [22]**	**Co-composting leachate (min. - max.)**
Alkalinity	(mg $CaCO_3$ L^{-1})	20	35 – 7 289	3 062 – 3 365
Ammoniacal nitrogen	(mg NH_4-N L^{-1})	1.9	1.3 -21 180	398 - 413
BOD_5	(mg L^{-1})	-	30 - 72 000	51 - 950
BOD_5/COD	-	-	0.02 - 0.87	0.02 - 0.29
COD	(mg L^{-1})	-	81 - 185 000	2 150 - 3 280
Conductivity	(mS cm^{-1})	-	1.4 - 82.6	7.49 - 7.73
Nitrites/nitrates	(mg NO_3-N L^{-1})	-	-	0.2 - 3.5
Ortho-phosphate	(mg PO_4-P L^{-1})	-	1.4 - 119	12.4 - 14.3
pH	-	6.5 - 9.0	3.0 - 9.2	7.74 - 8.09
Total dissolved solids	(mg L^{-1})	-	-	4 451 - 5 057
Total nitrogen	(mg N L^{-1})	2.18	7.2 - 18 570	421 - 447
Total organic carbon (dissolved)	(mg C L^{-1})	-	-	698 - 844
Total phosphorus	(mg P L^{-1})	0.128	0.5 - 485	13.7 - 15.2
Total solids	(mg L^{-1})	-	-	4 680 – 5 264
Total suspended solids	(mg L^{-1})	N	90 - 33 700	100 - 280
Metals (total)				
Al	(mg L^{-1})	0.75	-	0.47 - 0.73
Cu	(mg L^{-1})	C	0.01 - 2.34	0.008 - 0.03
Cd	(mg L^{-1})	0.0018	0 – 1.58	ND
Cr	(mg L^{-1})	0.074	-	0.012 - 0.014
Ni	(mg L^{-1})	0.47	0 - 4.43	0.13
Pb	(mg L^{-1})	0.0065	0 - 0.52	<0.003 - 0.0046
Zn	(mg L^{-1})	0.12	0.01 - 37.5	0.05 - 0.1
Anions				
F	(mg L^{-1})	-		0.24 - 3.1
Cl	(mg L^{-1})	860	32 - 8 670	475 - 533
SO_4	(mg L^{-1})	-		26.4 - 94.4

N: narrative statement

C: calculated

2.4. Activated sludge growth and acclimation

Prior to this experiment, the MBR was operated with synthetic composting leachate in the context of a nitrification kinetic study [24]. In order to acclimate the activated sludge, the synthetic leachate fed to the MBR was gradually substituted with real leachate over a period of 35 days. During the first 14 days, 25% of the synthetic feed was replaced with real leachate. Then, every 7 days, 25% of the synthetic leachate was replaced with real leachate until its complete replacement. The operating conditions of the reactor over the course of the sludge adaptation were the following: SRT = 30±2 d, HRT = 48±4 h.

2.5. Mass balance equations

MBRs' contaminant removal capacities can be compared between different studies when mass balance equations are clearly defined. The lab-scale sMBR used in this study was assumed to be a perfectly agitated continuous stirred tank reactor with two exit streams: 1) the permeate obtained from membrane filtration, and 2) the mixed liquor removed from the reactor on a daily basis. The main parameters involved in assessing the MBRs' leachate treatment capacities in this study are presented in Figure 2.

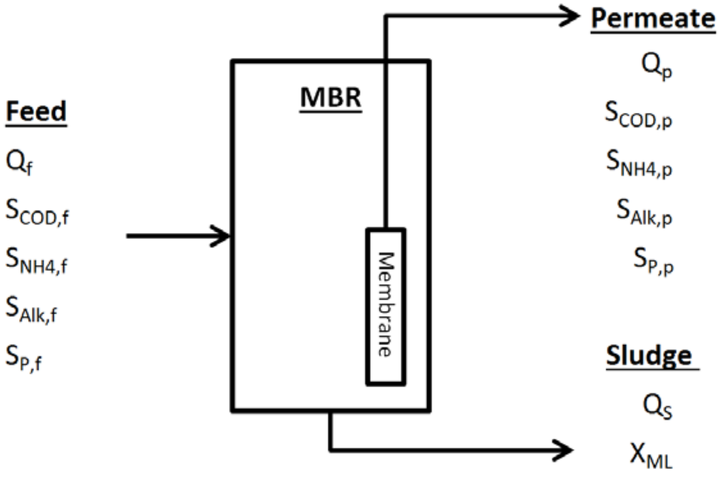


Figure 2 Experimentally measured variables

2.5.1. Mixed liquor biomass

Assuming a sterile feed, a constant volume, and a complete retention of the suspended solids by the submerged membrane, the biomass (mixed liquor) mass balance equation can be defined by the following equation (Eq. 1).

$$\frac{dX}{dt} = -\frac{X_{ML}}{SRT} + r_{X,ML} \quad \text{(Eq. 1)}$$

The apparent biomass growth rate ($r_{X,ML}$), which includes cells decay, is commonly expressed as (Eq.2):

$$r_{X,ML} = \mu X_{ML} \quad \text{(Eq. 2)}$$

The mixed liquor in the MBR is composed of a combination of both ammonia oxidizing (AOB) and heterotrophic (HB) bacteria, due to the presence of high concentrations of both ammoniacal nitrogen and COD in the leachates. Since these two groups of microorganisms have significantly different growth rates (to the order of h^{-1} for HB vs d^{-1} for AOB), the lumped mixed liquor generation rate ($r_{X,ML}$) must be expressed in terms of both AOB and HB apparent growth rates (Eq. 3).

$$\frac{dX}{dt} = -\frac{X_{ML}}{SRT} + r_{X,AOB} + r_{X,HB} \qquad \text{(Eq . 3)}$$

2.5.2. Biodegradable soluble carbon

In the case of biodegradable soluble carbon, the contaminants are the substrate for growing HB in the mixed liquor. Generally, this parameter is expressed in the form of COD concentration (mg COD L^{-1}). Assuming a constant volume, an excess of DO, and a growth rate limited reaction, change in biodegradable soluble carbon concentration in the MBR is expressed by the following equation (Eq. 4):

$$\frac{dS_{COD}}{dt} = \frac{(S_{COD,f} - S_{COD,p})}{HRT} - r_{S,COD} \qquad \text{(Eq. 4)}$$

The overall growth yield, which is defined as the ratio of biomass growth rate ($r_{X,HB}$) to substrate removal rate ($r_{S,COD}$), can be combined with Eq. 4 in order to obtain the following equation (Eq. 5).

$$\frac{dS_{COD}}{dt} = \frac{(S_{COD,f} - S_{COD,p})}{HRT} - \frac{r_{X,HB}}{Y_{X/S,COD}} \qquad \text{(Eq. 5)}$$

2.5.3. Ammoniacal nitrogen

Similarly to the biodegradable soluble carbon, ammoniacal nitrogen is the substrate for growing autotrophic AOB in the mixed liquor. Assuming a constant volume and an excess of DO (diffusion rate in microbial flocs > consumption rate), change in the ammoniacal nitrogen concentration in the MBR is expressed by the following equation (Eq. 6).

$$\frac{dS_{NH4}}{dt} = \frac{(S_{NH4,f} - S_{NH4,p})}{HRT} - r_{S,NH4} * \left(1 - \frac{S^*_{Alk,p}}{S_{Alk,p}}\right)^n \qquad \text{(Eq. 6)}$$

The last term on the right-hand side of the equation is added to account for the inhibitory effect of the acidic pH on the nitrification process. $S^*_{Alk,p}$ is the alkalinity concentration (expressed in mg eq. $CaCO_3$ L^{-1}) at which all nitrification metabolism ceases and n is an empirical constant.

2.5.4. Total soluble phosphorus

Assuming a constant volume, the MBR mass balance equation for total phosphorus is expressed by the following equation (Eq. 7).

$$\frac{dS_P}{dt} = \frac{(S_{P,f} - S_{P,p})}{HRT} - \frac{X_{ML} * Y_{P/X,ML}}{SRT} \quad \text{(Eq. 7)}$$

Where $Y_{P/X,ML}$ is the amount of phosphorus accumulated in the mixed liquor. Phosphorus can either be assimilated by the microorganisms, or chemically precipitated by the coagulating (Fe^{3+}, Al^{3+}) and flocculating agents (extracellular polymeric substance) present in the mixed liquor. Therefore, $Y_{P/X,ML}$ can take the form of (Eq. 8):

$$Y_{P/X,ML} = Y_{P,Bio/X,ML} + Y_{P,Coa/X,ML} \quad \text{(Eq. 8)}$$

2.6. Analytical methods

Water samples (feed and permeate) were analyzed for pH (Mettler Toledo SevenEasy), electrical conductivity (Mettler Toledo SevenCompact Conductivity), alkalinity (bromocresol green titration), solid content (total, dissolved, and volatile)(EPA Method 160.2), chemical oxygen demand (CEAEQ MA. 315 – DCO 1.1, Potassium dichromate), biological oxygen demand (5 days)(CEAEQ MA. 315 – DBO 1.1,), dissolved ammoniacal nitrogen (QuickChem Method 10-107-06-2-O, salicylate – nitroprusside colorimetric method), dissolved ortho-phosphate (QuickChem Method 10-115-01-1-B), dissolved nitrites/nitrates (QuickChem Method 10-107-04-2-B), dissolved organic carbon and total nitrogen (Shimadzu VCPH), and total metals (22 metals) and phosphorus (Varian Vista AX ICP-AES). Total metal and phosphorus contents were determined after preliminary sample digestion (15% trace metals grade HNO_3 and 5% H_2O_2 at 95°C for 2 hours).

Mixed liquor samples were analyzed for solid content (total, dissolved, and volatile) (EPA Method 160.2) and soluble extracellular polymeric substances (EPS) following the method described by More et *al.* (2016) [25].

3. Results and discussion

3.1. Organic contamination (BOD_5, COD, TOC)

Figure 3 presents the feed and the permeate COD concentrations, as well as the biodegradability of the organic contamination in the feed (BOD_5/COD). The “blank” condition was obtained by filtering the leachate without biotreatment in order to assess the COD removal rate associated with the micro-filtration membrane.

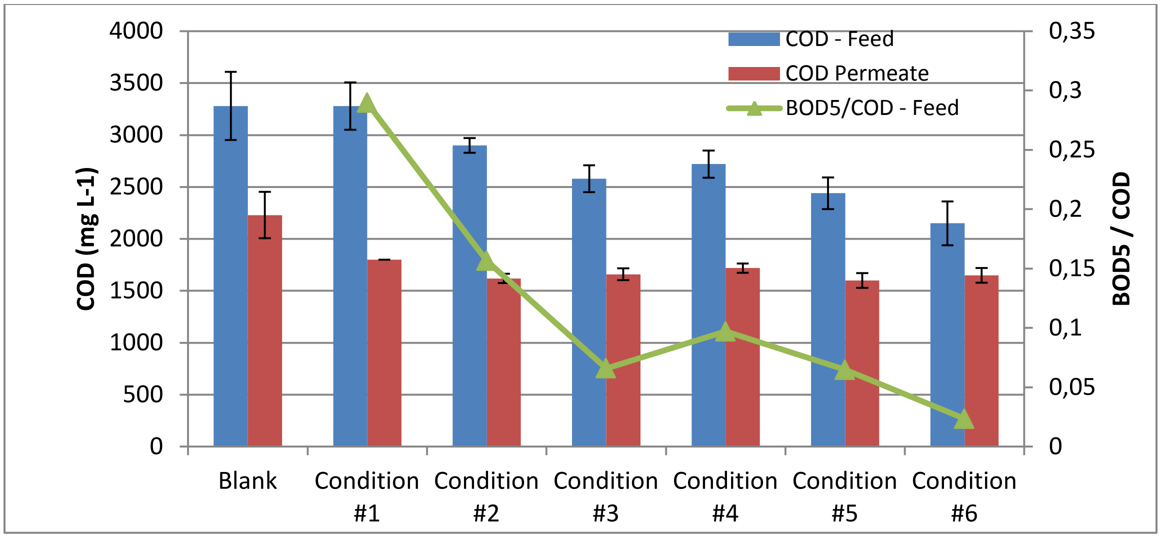


Figure 3 COD concentrations and leachate organic contamination biodegradability (BOD$_5$/COD)

The BOD$_5$/COD of the feed (fresh leachate) decreased from 0.28 to 0.03 between Condition #1 (day 0) and Condition #6 (day 148). The feed COD concentration also decreased from 3 280 to 2 150 mg L-1 over that same period. Despite the preservation efforts to maintain the leachates' composition constant, a biodegradation occurred at 4°C, leading to a significant decrease in biodegradable organic carbon.

By simply filtering the leachate, a 1050 mg L^{-1} removal of COD was obtained, thus indicating that approximately 32% of the dissolved organic molecules have molecular weight (MW) above 100 kDa. Campagna et *al.* (2013) reported a similar MW distribution from landfill leachate in which the proportion of COD > 100 kDa accounted for 37% of the total COD [26]. Therefore, notwithstanding the operational conditions (HRT and SRT), MBRs can separate a significant fraction of organic contamination, depending on the MW distribution of the contamination as well as the membrane MW cut-off.

For each tested condition (Figure 3), the permeate's COD concentration remained constant at 1675 ± 75 mg L^{-1}, despite the changes in the feed's BOD$_5$/COD. These results indicate the presence of refractory organic compounds with MW < 100 kDa. According to a previous study conducted on leachates originating from the same facility as those treated in this study, the remaining COD fraction is mostly comprised of humic- and fulvic-like and neutral hydrophobic substances with MW of approximately 1.5 kDa [21]. These highly aromatic molecules have various functional groups, including carboxylic and hydroxyl, and frameworks of randomly condensed aromatic rings [27], both of which explains their low biodegradability. Results from Figure 3 also indicate that increasing both the OLR from 1500 to 5000 mg COD L^{-1} d^{-1} and the SRT from 28 to 47 days have no significant impact on the removal efficacy of recalcitrant organic contamination. This observation is in accordance with the results of a previous study on landfill leachate by Chen and Liu which reported no significant impact of the OLR on the COD removal rate in an OLR range of 1000 to 4000 mg COD L^{-1} d^{-1} [6].

Specific COD removal rates ($r_{X,ML}$) and mixed liquor yield ($Y_{X/S}$) calculated from Eq. 1, Eq. 4, and Eq. 5 solved at steady-state conditions (d/dt = 0), are presented in Table 5.

Table 5 Organic contaminant removal percentage and calculated removal rates and yields

	BOD$_5$		TOC		COD		Substrate utilization	
	OLR	Removal	OLR	Removal	OLR	Removal	Removal rate	$Y_{(X/S)}$
	(mg L^{-1} d^{-1})	(%)	(mg L^{-1} d^{-1})	(%)	(mg L^{-1} d^{-1})	(%)	(mg COD L^{-1} d^{-1})	(mg MLVSS mg COD $^{-1}$)
Blank	370	14	375	5.3	1430	28.0	-	-
Condition #1	440	> 99,1	375	19,9	1515	45,1	683	0,14
Condition #2	300	> 99	518	20,2	1933	44,1	853	0,14
Condition #3	160	> 97,6	701	16,2	2477	35,7	883	0,16
Condition #4	490	> 98,1	1433	15,7	5022	36,8	1846	0,14
Condition #5	150	> 97	675	16,6	2342	34,4	806	0,18
Condition #6	95	> 91,9	1289	14,2	3969	23,3	923	0,20

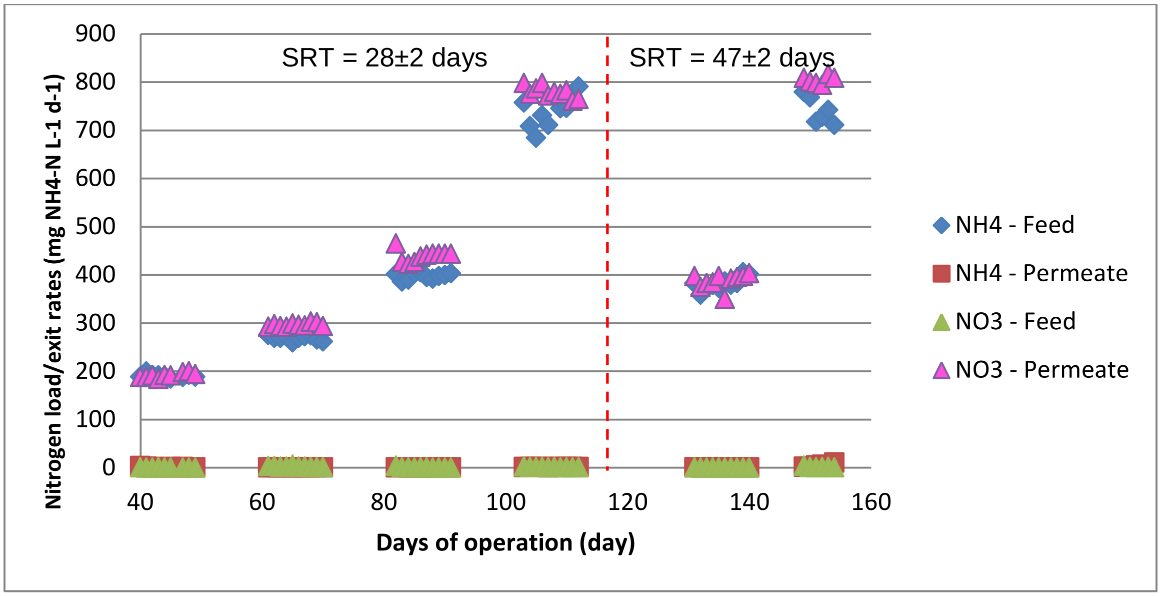
The variation in organic contaminant removal percentages from Condition #1 to #6 are mostly related to the decrease in BOD$_5$/COD in the raw leachate (feed). In terms of COD removal rates, the highest value (1 846 mg COD L^{-1} d^{-1}) was obtained at Condition #4 (HRT = 13h, SRT = 27d). This particular removal rate is comparable to those reported by Brown et *al.* (2013) (1 196 mg COD L^{-1} d^{-1}) and Sadri et *al.* (2008) (1 590 mg COD L^{-1} d^{-1}) [8, 16]. According to those results, COD removal rate values of up to 1 850 mg COD L^{-1} d^{-1} for easily to moderately biodegradable organic contamination can be used to design MBRs in leachate treatment applications.

In order to evaluate the effect of both OLR and SRT on the microorganisms' capacity to metabolize the organic carbon, the specific biomass/substrate yield ($Y_{X/S}$) was calculated. According to the results presented in Table 5, the $Y_{X/S}$ mostly depends on the SRT instead of the OLR. From Condition #1 to #4 (SRT = 28 d), the $Y_{X/S}$ remained almost constant at 0.14, and from Condition #5 to #6 (SRT = 47 d), the $Y_{X/S}$ was 0.19. It is hypothesized that the increase in $Y_{X/S}$ is due to a shift in microbial population caused by the change in their retention time within the MBR. With a SRT of 47 days, slow-growing autotrophic microorganisms such as nitrosomonas can proliferate and compete against fast-growing HB for DO and nutrients. Previous studies have reported net yields of 0.34 mg AOB mg N^{-1} [28] and 0.67 mg HB mg COD^{-1} [29] for autotrophic and heterotrophic microorganisms, respectively, treating leachates. Since both strains of microorganisms have different growth rates and yields [30], the resulting overall yield changed according to the microbial population structure in the mixed liquor. Such a shift is the results of two or more sources of substrate present in significant concentrations in the wastewater to be treated. Therefore, according to the results from this experiment, when treating leachates using a MBR, higher biomass production will occur with a SRT of 47 days as opposed to one of 28 days.

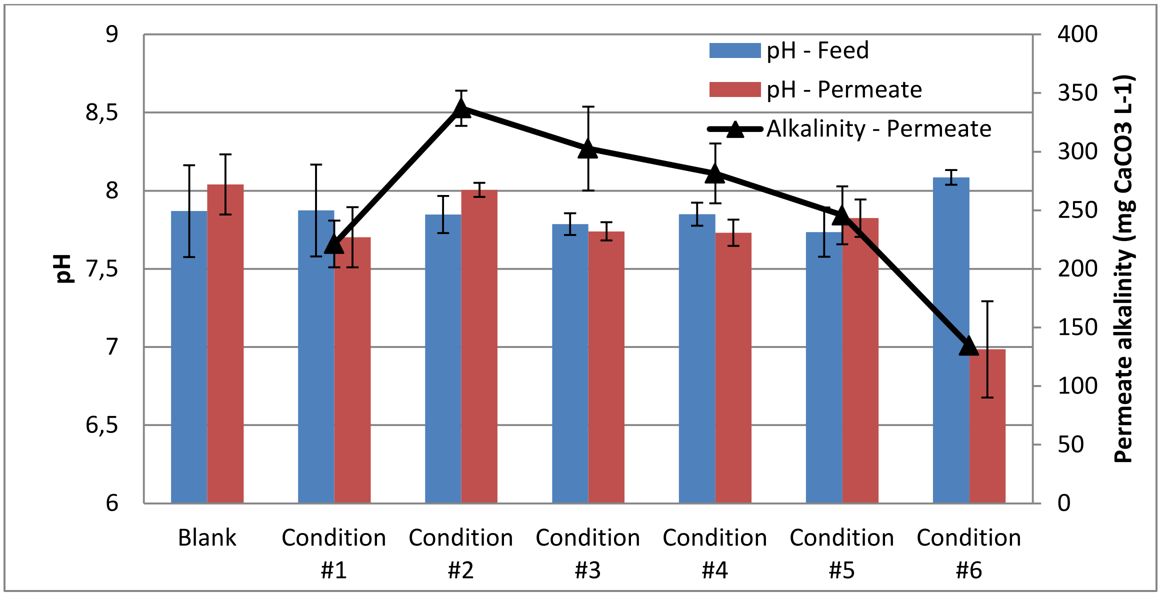
Between Condition #1 and #4, increasing the OLR from 1 515 to 5 022 mg COD L^{-1} d^{-1} resulted in an increase in MLVSS concentration from 2 680 to 6 990 mg L^{-1}. However, the respective $Y_{X/S}$ calculated for each of these conditions remained constant at 0.15 ± 0.01. The same trend was observed with Condition #5 and #6, where the $Y_{X/S}$ remained constant at 0.19 ± 0.1 despite an increase in OLR from 2 342 to 3 969 mg COD L^{-1} d^{-1}. The HB concentration in the reactor is thus directly proportional to the amount of available carbon fed to the reactor. Therefore, the MBR's efficacy in removing COD remained stable over the range of OLR tested, and is mainly limited to the amount of bioavailable carbon and DO [6, 17].

3.2. Ammoniacal nitrogen oxidation

Nitrogen is considered one of the major contaminants in leachates originating from wastes [31]. Nitrogen is found as: 1) proteinaceous compounds, 2) amines integrated into fulvic- and humic-like substances, and 3) ammonium salts [7, 32]. In this study, the leachate treated had an average ammonium concentration of 406 mg NH_4-N L^{-1}, which accounted for 93% of the total nitrogen. The pH, alkalinity, ammonium and nitrate concentrations values measured in each condition are presented in Figure 4a and Figure 4b.



(a)



(b)

Figure 4 (a) Concentration of ammonia and nitrates in feed and permeate samples at different NLRs (dashed line = change in SRT) and HRTs, and (b) pH and alkalinity measurements at different NLRs and HRTs

Figure 4a shows that all the ammonium (NH_4-N) fed to the MBR has been converted into nitrates (NO_3-N) through biological nitrification. The slightly higher concentrations of nitrates in the permeate are attributed to the conversion of organic ammonium to ionic ammonium by HB, followed by the conversion of ionic ammonium to nitrates by nitrifying bacteria. At each tested condition, the constant concentrations of ammonium and nitrates measured in the permeate indicated that a steady state was reached. Figure 4b shows that feed pH remained in the range of 7.74 to 8.09. The Blank experiment permeate's alkalinity remained unchanged (approx. 3 250 mg $CaCO_3$ L^{-1}) and, therefore, is not presented in Figure 4 in order to maintain the Y axis scale from 0 to 400 mg $CaCO_3$ L^{-1}. A drop in permeate pH and residual alkalinity was observed at Condition #6. A similar difference in alkalinity was observed in the untreated leachates (3 060 mg $CaCO_3$ L^{-1} at Condition #6 vs. an average of 3 300 $CaCO_3$ L^{-1} for the other conditions). This change in alkalinity in the untreated leachates is assumed to be caused by the biological activity evidenced by the COD consumption that occurred in the 1 m^3 leachate sample that was preserved at 4°C for the duration of the experiment.

The average ammonium concentration measured in the feed and the permeate for Condition #1 to #6, as well as the overall ammonium removal rates ($r_{S,NH4}$) calculated by solving Eq. 6 at steady-state conditions, ($dS_{NH4}/dt = 0$) are presented in Table 6.

Table 6 Ammonium concentrations, removal percentage, and calculated removal rates

NLR	Feed concentration	Permeate concentration	Removal	$r_{NH,i}$
(g NH_4-N m^{-3} d^{-1})	(mg NH_4-N L^{-1})	(mg NH_4-N L^{-1})	(%)	(mg NH_4-N L^{-1} d^{-1})

Blank	191	413 ± 9	380 ± 10	8.0	-
Condition #1	191	413 ± 9	2.8 ± 1.8	99.3	189 ± 4
Condition #2	269	403 ± 8	0.9 ± 0.2	99.8	268 ± 5
Condition #3	397	414 ± 7	0.4 ± 0.1	99.9	397 ± 7
Condition #4	742	402 ± 18	0.6 ± 0.1	99.8	740 ± 33
Condition #5	382	398 ± 14	0.6 ± 0.1	99.9	381 ± 14
Condition #6	741	401 ± 15	2.7 ± 1.7	99.3	736 ± 30

In Table 6, the “blank” condition was obtained by filtering the leachate without biotreatment in order to assess the ammonium removal associated with the micro-filtration membrane. Just 8% of the ammonium was removed, which might be attributed to the ammonium found as proteinaceous compounds or amines integrated into fulvic- and humic-like substances that have MW > 100 kDa.

At a SRT of 28 days, the ammonium concentration in the permeate remained in the range of 0.4 to 2.8 mg NH_4-N L^{-1} for NLRs ranging from 191 to 742 mg NH_4-N L^{-1} d^{-1}, corresponding to removal percentages of >99.3%. Similarly, at a SRT of 47 days, ammonium concentrations of 0.6 and 2.7 mg NH_4-N L^{-1} for NLRs ranging from 382 and 741 NH_4-N L^{-1} d^{-1}. Ammonium concentration in the permeate exceeded 1 mg NH_4-N L^{-1} in two conditions: Condition #1 and Condition #6. The highest ammonium concentration was measured at Condition #6, with a value of 5.60 mg NH_4-N L^{-1}. This lower ammonium removal efficacy can be correlated with the pH of the reactor. The MBR’s pH remained above 7.5 during the whole experiment with the exception of Condition #6, where it reached an average of 6.98. Furthermore, the drop in ammonium removal efficacy correlates with Condition #6’s drop in residual alkalinity to concentrations below 200 mg $CaCO_3$ L^{-1} (Figure 4b). According to the stoichiometry of the nitrification reaction, 7.14 mg of alkalinity (as $CaCO_3$) is consumed for every milligram of ammonium oxidized. While this ratio was respected for each condition, the results clearly demonstrated that a residual alkalinity of > 200 mg $CaCO_3$ L^{-1} is required in order to maintain optimal ammonium removal efficacy.

Overall ammonium oxidation rates calculated in this study are not sufficient to conclude on the effect of the SRT (in the range of 28 to 47 days) and the NLR on MBRs’ nitrification performance. Since no significant residual ammonium concentration was measured in the permeate for each condition, it can be concluded that the maximum overall ammonium oxidation rate ($r_{S,NH4}$) of leachate-treating MBRs is equal to or higher than the highest rate calculated during the experiment, which was 740 mg NH_4-N L^{-1} d^{-1}. This value was obtained when adequate residual alkalinity and DO concentration were maintained.

The proportional increase in both the NLR and the $r_{S,NH4}$ also indicate that the nitrifying bacteria population can easily adapt to NLR increments of 75 to 345 mg NH_4-N L^{-1} d^{-1}. This observation is valid for both SRTs tested. Therefore, limiting factors for nitrification in MBRs are the alkalinity, the maximum total suspended solids concentration (TSS) achievable (which will not cause major membrane fouling), and the oxygen transfer rate to the nitrifying bacteria. According to Ahmed et *al.* (2012), very low SRT (< 10 days) can also have adverse effects on nitrification due to the depletion of nitrifying bacteria population [1].

3.3. Phosphorus

Phosphorus removal in a MBR is obtained through two possible ways: 1) bioaccumulation/assimilation, 2) chemical precipitation with coagulants naturally present in the wastewater (Fe^{3+}, Al^{3+}) and bioflocculants such as EPS. Bioflocculants are of interest in MBRs since they can improve phosphorus removal efficacy by increasing the size of colloidal particles that contain phosphorus, and thus prevent these particles from passing through the membrane. In order to evaluate the effect of the phosphorus loading rate (PLR) and the SRT on the MBR's removal efficacy, the calculated phosphorus mass balance and removal rates are presented in Table 7. This table also includes the ratios of phosphorus per TSS ($Y_{P/X,ML}$), calculated by solving Eq. 7 at steady state ($dS_P/dt = 0$) and the molar ratio of removed phosphorus per removed coagulant (Fe^{3+}, Al^{3+}).

Table 7 Phosphorus mass balance in MBR treating leachate

	Mass balance				Ratios	
	PLR	Bioaccumulation/ assimilation	Chemical precipitation	Overall Removal	P:TSS	P:Coagulant
	(mg P L^{-1} d^{-1})	(mg P L^{-1} d^{-1})	(mg P L^{-1} d^{-1})	(%)	(mg P:g TSS)	(mol P:mol Coa)
Blank	6,43	0	3,88	60%	-	1,47
Condition #1	6,72	1,92	3,37	79%	41	1,58
Condition #2	10,11	2,34	3,94	62%	39	1,64
Condition #3	14,63	2,86	4,28	49%	38	1,46
Condition #4	27,54	6,16	8,75	54%	38	1,99
Condition #5	13,12	3,62	2,93	50%	27	1,19
Condition #6	19,12	4,11	2,98	37%	26	1,11

An elementary composition analysis on the biomass found in the mixed liquor was performed first in order to estimate the amount of phosphorus removed by bioaccumulation and assimilation. On average, the biomass from the mixed liquor contained 1.5 ± 0.2% of phosphorus. Then, the remaining phosphorus removed from the feed was considered chemically precipitated. At Condition #1 to #4 (SRT = 28 days), increasing the PLR from 6.7 to 27.5 mg P L^{-1} d^{-1} decreased the removal efficacy from 79 to 54%. On average, 23% of the phosphorus was bioaccumulated, while 30 to 50% of the remaining phosphorus was chemically precipitated. Then, at Conditions #5 and #6 (SRT = 47 days), increasing the PLR from 13.1 to 19.1 mg P L^{-1} d^{-1} decreased the removal efficacy from 50 to 37%. On average, 24% of the phosphorus was bioaccumulated, while 16 to 22% of the remaining phosphorus was chemically precipitated.

At first glance, increasing the PLR seems to have decreased the phosphorus removal efficacy. However, considering that other factors, such as the biomass and coagulant concentrations, play a significant role in phosphorus removal (See Eq. 7 and Eq. 8), it is more accurate to use the $Y_{P/X,ML}$ to compare the different conditions on the same grounds. Results reported in Table 5 show that the $Y_{P/X,ML}$ remained constant at 39±2 and 26±1 mg P / g TSS at SRT of 28 and 47 days, respectively. Therefore, MBRs' overall phosphorus removal capacity is dependent on the SRT and the mixed liquor TSS concentration rather than on the PLR, as is commonly reported [1].

Phosphorus chemical precipitation in MBRs have previously been reported by Roy et *al.* (2019). Using synthetic composting leachate, they showed that phosphorus can either be precipitated in the form of struvite or by coagulation with multivalent cations present in the leachate, such as Fe and Al. Since the ammoniacal nitrogen had been completely oxidized to nitrates in this experiment, the struvite precipitation hypothesis was ignored. Phosphorus coagulation with multivalent metal ions is obtained through different reactions. Orthophosphate ions can either be attracted to metal oxide polymers by Van Der Waal force, or form ionic bonding directly with multivalent metal cations according to the following reactions:

$$M^{3+} + PO_4^{3-} \rightarrow MPO_4$$

$$3\ M^{2+} + 2\ PO_4^{3-} \rightarrow M_3(PO_4)_2$$

In each case, the highest achievable molar ratio of phosphorus:multivalent metal (P:M) is 1. In order to estimate the role played by the EPS in the phosphorus chemical precipitation, the P:M (Fe and Al) was calculated (Table 7) and EPS concentrations were measured at each tested condition and are presented in Figure 5 (note: the Y axis begins at 1.5 g L^{-1}). The average P:M was 1.67 and 1.15 for SRTs of 28 and 47 days, respectively. Ratios higher than 1 indicate that EPS had an impact in phosphorus removal, and the impact was greater at a SRT of 28 days compared to 47 days. This observation is in agreement with EPS concentration measurements that showed that EPS concentrations were 13% higher at a SRT of 28 days when compared to 47 days. This difference could be attributed to a shift in the microbial population responsible for the production of EPS between the two SRTs tested.

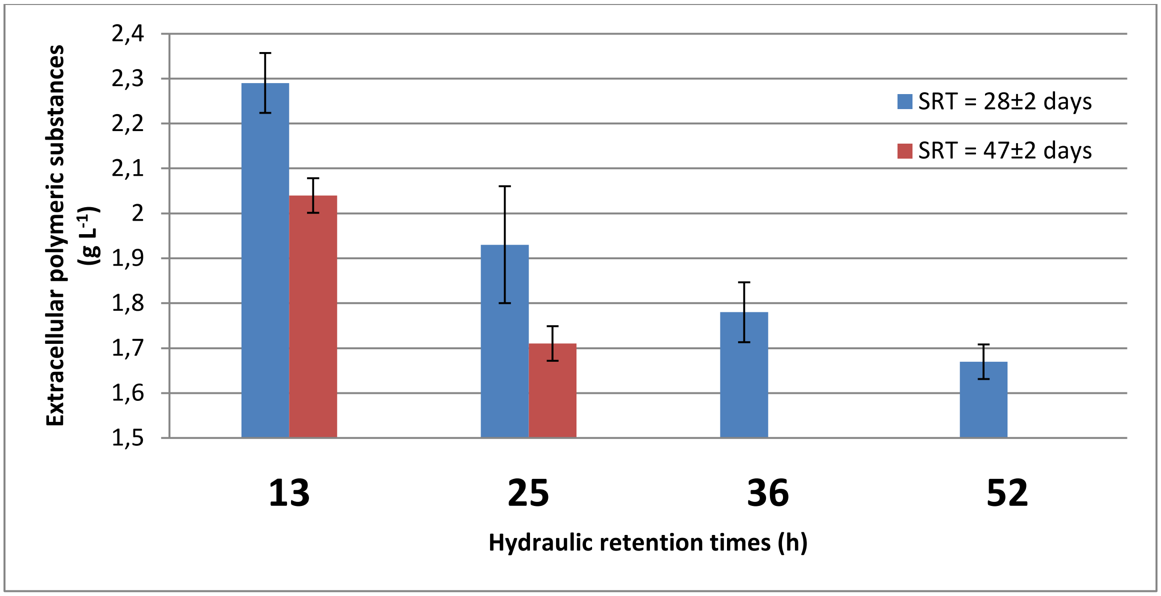


Figure 5 EPS concentrations in the mixed liquor measured at SRTs of 28±2 days (blue) and 47±2 days (red) and different HRTs

Globally, MBRs treating leachates have the ability to remove phosphorus at rates of 39±2 and 26±1 mg P / g TSS when operated at SRTs of 28 and 47 days, respectively, notwithstanding the PLR. Higher removal rates at lower SRTs are correlated with an increased production of EPS, which can bond soluble phosphorus.

3.4. Metals

The MBRs' removal capacity was only studied for Al, Cr, and Ni because some MBR parts released Zn and Cu in the mixed liquor, resulting in higher concentrations in the permeate than in the feed. Ba and Sr were also studied, since they are commonly responsible for the irreversible chemical fouling in membrane filtration processes [33]. Figure 6 shows the residual metals concentration in the permeate in terms of C/Co, where C and Co are the permeate and the feed metals concentration, respectively.

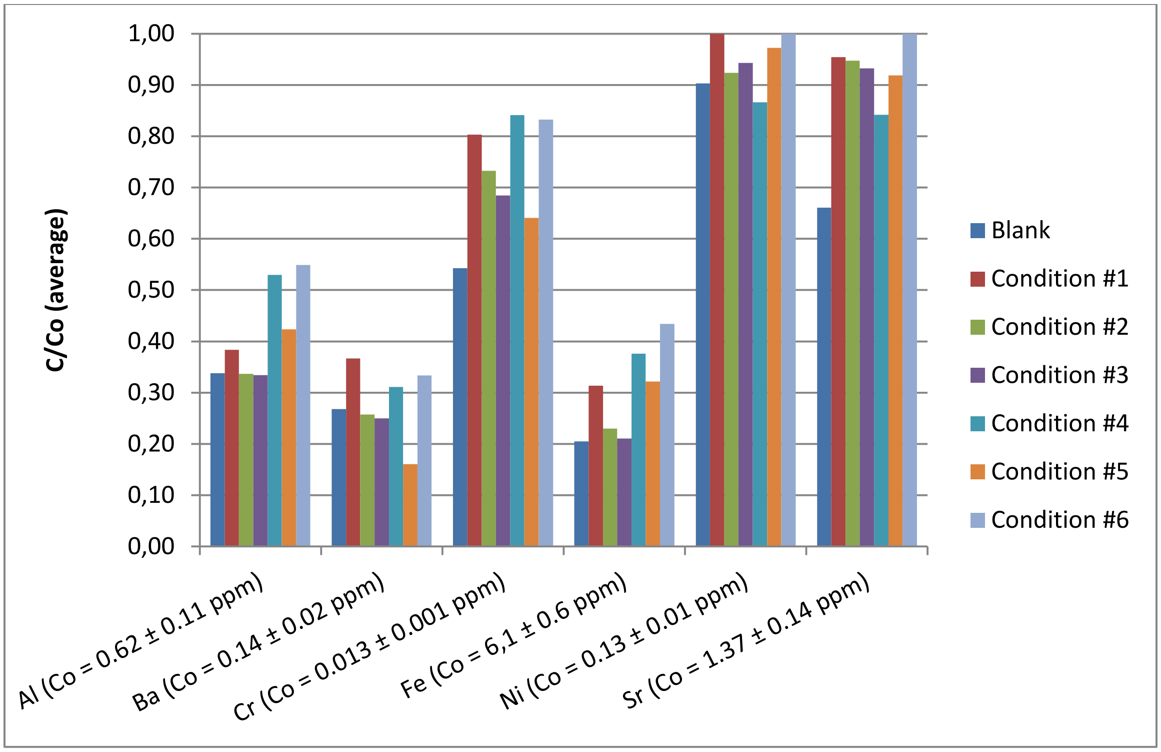


Figure 6 Residual metal concentrations measured in the permeate at the different tested conditions

While no direct correlation was observed between the operating conditions (HRT and SRT) and the observed removal rates, each metal individually followed a similar removal trend. In general, the MBR metal removal capacity showed the following results, in order of average removal percentage: Ba (72%) > Fe (69%) > Al (57%) > Cr (24%) > Sr (7%) > Ni (5%). In a previous study, Zolfaghari et *al.* (2017) showed that humic-like substances, with their three pKa values of 2.5, 6.1, and 8.8, possess negative sites which can interact with metal cations [34]. Consequently, humic-like substances are often associated with the transport of metals in aquatic environments. According to the results presented in Figure 4, the permeate contained a constant concentration of recalcitrant COD mainly composed of humic-like substances. At the permeate's pH (ranging from 6.98 to 8.5), the humic-substances were mostly deprotonated (pH > pKa), which increased their affinity for metals ions. Therefore, the presence of recalcitrant COD in the permeate, mostly comprised of humic-and fulvic-like substances [21], is presumed to be responsible for the presence of unwanted metals in the treated leachate, due to their strong ionic interactions [34].

Higher removal of Al and Fe is explained by stronger interactions between three valent cations and the negatively charged microbial cells surface [35]. Al and Fe also contribute to the formation of insoluble salts with orthophosphate. High removal rates of Ba can also be explained by the formation of insoluble salts such as barium sulfate or barium carbonates in the leachate, since their solubility is amongst the lowest of all sulfate and carbonate salts. Low removal rates of Ni were expected due to high concentrations of humic-like substances in the permeate. He et *al* previously reported that Ni exhibited a strong complexation capability with aliphatic structures, carbohydrates, and proteinaceous compounds [7]. Likewise, Zolfaghari et *al.* (2017) reported that Cr has more adsorption affinity for humic substances than for sludge, which explains its low removal in treated leachates containing high concentrations of recalcitrant COD.

In general, MBRs' metal removal capacity is mostly dependant on the chemical properties and concentrations of the recalcitrant COD found in the leachates, and, therefore, SRTs ranging from 28 to 47 days and HRTs ranging from 13 to 52 hours have no impact on MBRs' metal removal capacity.

4. Conclusion

The main objectives of this study were to define the effect of the SRT and the loading rates on MBRs' efficacy in removing contaminants, as well as to provide useful data for the design of MBRs treating leachates. Specific operating conditions were selected in order to remain in the range of economically feasible designs. Main conclusions for each of the contaminants studied are the following:

Organic contaminants (BOD_5 and COD):

- Based both on previously reported data as well as results from this study, MBRs can efficiently treat up to 1 850 mg COD L^{-1} d^{-1} of highly to moderately biodegradable COD.
- SRTs in the range of 28 to 47 days have no impact on the degradation of recalcitrant COD, notwithstanding the OLR.
- In multi–substrate wastewater such as leachate, increasing the SRT can lead to a higher sludge production due to the creation of favorable conditions for slow-growing microorganisms.

Ammoniacal nitrogen:

- Overall ammonium removal rates of >740 mg NH_4-N L^{-1} d^{-1}, corresponding to a >99.3% removal, can be achieved with MBRs treating leachates at both SRTs tested.
- Ammonium oxidation limiting factors are: sufficient alkalinity (residual concentration > 200 mg $CaCO_3$ L^{-1}), optimal pH (>7.5 in the permeate), MBRs' maximum achievable TSS, and the oxygen transfer rate to the nitrifying bacteria.

Total phosphorus:

- Overall phosphorus removal rates of 39±2 and 26±1 mg P g TSS^{-1} were calculated for SRTs of 28 and 47 days, respectively.
- PLR in the range of 6.7 to 27.5 mg P L^{-1} d^{-1} have no significant impact on MBRs' phosphorus removal capacity.
- Low SRTs showed higher removal rates due to the increased production of EPS.

Metals:

- MBRs’ metal removal capacity depends mostly on the metals’ affinity with both the leachate’s recalcitrant COD (humic-like substances) and sludge.
- Tested HRT and SRT showed no significant impact on MBRs’ metal removal capacity, since the concentration of recalcitrant COD in the permeate remained constant.

Acknowledgements

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