



Nanofiltration as post-treatment of MBR treating landfill leachate

Míriam Cristina Santos Amaral*, Wagner Guadagnin Moravia, Liséte Celina Lange, Mariana Moreira Zico Roberto, Natalie Cristine Magalhães, Túlio Luís dos Santos

Department of Sanitary and Environmental Engineering, Federal University of Minas Gerais, P.O. Box 1294, 30.270-901 Belo Horizonte, MG, Brazil, Tel. +5531 3409 3669; email: miriam@desa.ufmg.br (M.C.S. Amaral)

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ABSTRACT

Landfill leachate management has been a major environmental, economic, and social concern, and its treatment brings forth a challenge, especially regarding the high concentration of refractory organic matter, ammonia, and toxic compounds, which may vary considerably depending on the maturity, age, and biochemical reactions occurring in the landfill, besides the high variation on the volume generated. This study was aimed to investigate NF use as a post-treatment of landfill leachate that has been treated by membrane bioreactor (MBR). The experimental setup consisted of pilot plant comprised by an air stripping reactor, plus MBR and NF membranes. The pilot plant has a treatment capacity of 3 m³/d. The system has shown excellent leachate treatment performance, especially regarding the removal of chemical oxygen demand (80–96%), ammonia (85–95%), color (98–99.9%), and phosphorus (78–99.8%). The results have also shown the importance of NF to improve the effluent condition, and to produce treated effluents free of chemical substances and sludge. The membrane fouling was imputed to the adsorption of a humic substance on the membrane surface and the pore-blocking phenomena. By this route, treated landfill leachate may be reused at the landfill as water for dust arrestment, and also as earth work on construction sites.

Keywords: Landfill leachate; Nanofiltration; Membrane bioreactor

1. Introduction

Landfills are regarded as efficient, cost-effective, and technically effectual disposal systems for solid waste throughout the world. Up to 95% of solid waste generated worldwide has been currently disposed of at landfills [1,2]. However, the leachate generation remains a challenge to the landfill management.

Landfill leachate is the moisture produced by natural air humidity plus the water content present in organic wastes that allows for the biological degradation of organic matters, and water percolation through the cover and inner layers of the landfill cells, so adding on dissolved or suspended material released by residual masses.

Treating landfill leachate has been a challenge itself, primarily due to the presence of high concentrations of refractory organic matter, ammonia, organic

*Corresponding author.

and inorganic pollutants, toxic compounds, high pH, and color values that may vary significantly depending on the maturity, age, and biochemical reactions occurring in the landfill [3], besides the high variation of the volume generated.

Although many techniques may be applied, selecting the most appropriate leaching treatment will depend on its properties, technical applicability, cost-effectiveness and other factors related to effluent quality requirements. Biological processes are very effective when they are used at young landfill leachates, but their efficiency decreases as the leachate ages increase [4,5]. In particular, conventional biological systems cannot effectively treat old landfill leachates containing recalcitrant contaminants, and if they are used alone, they will not have enough removal efficiency because of the inhibiting effects of some contaminants such as ammonium and heavy metal alloys. The physical and chemical treatment processes include coagulation and flocculation, chemical oxidation, chemical precipitation, activated carbon absorption, ozonation, and membrane processes. Usually, physico-chemical treatment units ensure the removal of metals, and partially ammonium, but that is not enough to remove organic compounds from leachate. The disadvantage of treating leachate by coagulation, precipitation and chemical oxidation processes is the excess of sludge left after the treatment, which is difficult to manage, besides its high operational cost. Therefore, it is necessary to search for effective alternative treatments that would meet the technical and economic sustainability standards.

Over the last years, many new landfill leachate treatment methods have been developed [6–8], among which nanofiltration (NF) technology has shown some advantages over the others regarding the removal of dissolved matters [9]. NF appears like one of the most promising and efficient landfill leachate treatment method, especially when it is associated with the membrane bioreactor (MBR). MBR technology is an advanced biological treatment process that replaces the traditional secondary clarifier of a biological process by using a membrane separation unit, and it has also shown to be particularly advantageous to treat old landfill leachates. The advantages of MBRs over conventional biological processes are well-known, and include better effluent condition, higher process stability, smaller footprint, increased biomass or mixed liquor suspended solids retention, and lower sludge production [10]. The advantages of using an NF membrane are many. It requires lower operating pressures, allows for higher fluxes than reverse osmosis (RO) membranes, provides higher retention than an ultrafiltration (UF) membrane [11], has lower operational

cost, high rejection of polyvalent ions and an organic compounds with a molar mass higher than 300 g/mol, requires lower investment, and has operational and maintenance costs. Furthermore, due to its unique properties among the membranes UF and RO, in leachate treatment the NF membrane has an important advantage of removing recalcitrant organic compounds and heavy metals [12].

NF membranes have their surfaces slightly charged. Most NF membranes are negatively charged at neutral pH. Such a surface charge plays a major role in the transportation mechanism, and as well in the separation properties of NF membranes [13]. In NF, the separation occurs based on two principles. Neutral species are separated according to their sizes (molecules larger than 200–300 g/mol are rejected), while inorganic ions are separated by electrostatic interaction between ions and membrane.

NF membrane fouling significantly impairs productivity, and increases operational costs. Organic matters such as humic acids have been identified as one of the commonest membrane foulants [14]. Humic acids comprise a group of heterogeneous polymeric organics with molecular weights ranging from few thousand to a few hundred thousand g/mol, depending on their source [15].

Organic matters contained in landfill leachates chiefly include volatile fatty acids and humic substances. The contents of the two organic fractions in leachate may vary considerably depending on the landfill age. At young landfills, most of organic carbons are present as volatile fatty acids [16]. However, in mature landfills, humic substances (humic acids, fulvic acids, and humins) dominate the organic fraction in methanogenic leachate by as much as 60% [17].

This study was primarily aimed to investigate the use of NF as the landfill leachate post treatment carried out by MBR.

2. Materials and methods

2.1. Sampling and leachate characterization

The leachate examined in this study was supplied by a landfill located in the state of Minas Gerais, Brazil. The characteristics of the corresponding leachate used in this study are shown in Table 1. Inert chemical oxygen demand (COD) indicated that the leachate used shows some typical properties of an old leachate because the substantial percentage of inert COD. The inert COD test was carried out using the method proposed by Germili et al. [18]. The BOD/COD ratio also corroborates the assumption that the leachate was an old one. The average ammonium concentration found

Table 1
Characteristics of leachate used in this study

Parameters	Average	Deviation
Real color, mg Pt-Co/L	1,087	65
Apparent color, mg Pt-Co/L	1,671	881
Total COD, mg/L	4,044	544
BOD ₅ , mg/L	116	89
Inert COD, %	48	4
BOD/COD	0.03	0.02
COT, mg/L	656	44
pH	8.1	0.2
Humic substances, mg/L	2,476	126
Total nitrogen, mg/L	2,207	80
Ammoniacal nitrogen, mg/L	1,716	386
Nitrite, mg/L	0.19	0.01
Nitrate, mg/L	5.29	1.20
Conductivity, mS/cm	24	3
Alkalinity, mg/L	7,690	2,333
Chloride, mg/L	2,762	137
Phosphorus, mg/L	34	8.8
TS, g/L	10.2	0.2
FTS, g/L	7.9	0.4
VTS, g/L	2.3	0.3
TSS, mg/L	22.2	7.9
FSS, mg/L	2.4	1.4
VSS, mg/L	19.4	7.4
Cadmium, mg/L	0.12	0.03
Lead, mg/L	0.34	0.35
Chrome, mg/L	2.9	0.40
Iron, mg/L	9.3	0.49
Zinc, mg/L	0.65	0.07

was 1,716 mg/L, which indicates that pretreatment requires ammonium removal. Nitrate in leachate may be considered as a partial nitrification resulting from the leachate flow in an uncovered outdoor disposal area. Low heavy metal removal has also been observed.

2.2. Experimental setup

The pilot plant (Fig. 1) consists of an air stripping reactor for ammonia removal, a submerged aerobic MBR for organic matter and nutrient removal, and a NF system for polishment (organic and inorganic pollutant removal). The treatment capacity of the pilot plant is 3 m³/d.

2.2.1. Air stripping

The air stripping for ammonia removal was performed by using a 3 m³ (effective volume) reactor feed with raw landfill leachate. The hydraulic retention time of the reactor was 48 h and no pH adjustment

was made. The aeration with an air flow rate of 60 m³/h was provided by an air compressor connected to a thick bubble diffuser.

2.2.2. Membrane bioreactor

The pilot-scale MBR and the membrane module used to carry out the tests were supplied by “PAM Membranas Seletivas Ltda.” The MBR had a submerged hollow fiber microfiltration membrane module, with an average pore size of 0.5 μm, a membrane area of 14 m, and a packing density of 500 m²/m³. The MBR comprised three tanks, a biological tank that operated with an effective volume of 3,000 L, a 200-L membrane tank plus a 10,000-L storage tank for the permeate, a diaphragm pump used to perform both, microfiltration and backwash, three-way solenoid valves, level sensors, needle valves for flow adjustment, rotameters to indicate the permeate, backwash and air flows, a pressure gage, and a skid with an electric panel for automatic control of permeation and backwash operations.

The MBR was inoculated with the sludge from the activated sludge reactor of a sewage treatment plant, followed by an acclimation period aimed to acclimatize the microorganisms to the system and wastewater conditions. The hydraulic detention time and solid retention time of 48 h and 60 d respectively. The system operated continuously for 220 d. The average MLVSS in the MBR was 6,000 mg/L. A 15-s backwash was performed at every 15-min period which was applied to control the fouling. The membrane properties and MBR conditions are shown in Table 2.

The MF membranes were submitted to weekly maintenance cleaning procedures as following: the membrane was maintained in a 500 mg/L sodium hypochlorite solution for 4 h plus a citric acid solution with pH lower than 2 for 20 min, and as well a bimonthly recovery cleaning procedure with a 1,000 mg/L sodium hypochlorite solution for 24 h and a citric acid solution with pH lower than 2 for 30 min.

2.2.3. Nanofiltration

The effluent from the MBR was post-treated by a NF unit. The unit was operated with a feed and a permeating flow of 160 and 15 L/h, respectively, pressure of 10 bar and recovery rate of 60%. The spiral NF type membrane module (NF90–2540, commercial membrane supplied by Dow-Filmtec) (restraint 90% salts stabilized and an effective filtration area of 2.6 m²) was adopted for this study. The concentrate was returned to the landfill cells.

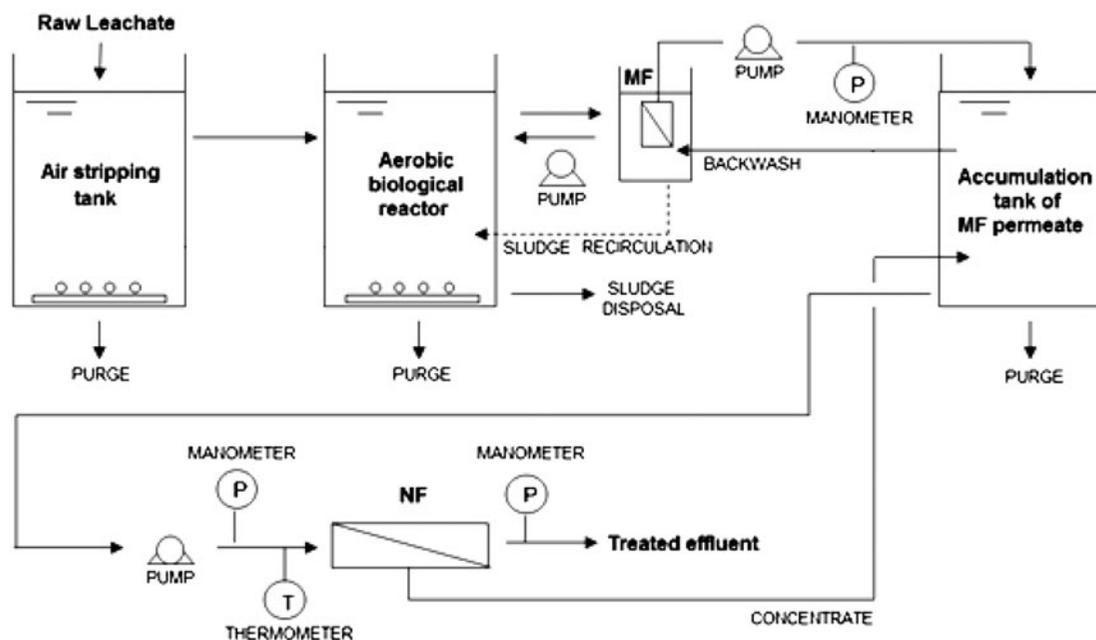


Fig. 1. Pilot unit flowchart.

Table 2
Membrane properties and MBR operational conditions

Parameters	Value
Effective biological tank volume, m ³	3
Permeate tank volume, m ³	0.2
Hydraulic retention time, h	48
Sludge retention time, D	60
Organic load rate, mg/L	5,242
Air flow rate in biological tank, m ³ /h	6
Air flow rate in membrane tank, m ³ /h	2.4
MLSSV, mg/L	6,000
Membrane configuration	Submerged hollow fiber
Membrane materials	Poly(ether)imide
Average membrane pore size, μm	0.5
Effective membrane area, m ²	14
Flux, L/mh	5
Backwash, s/min	15/15

The NF performance was assessed by monitoring the following process variables periodically: permeation flow, pressure, temperature, pH, and characteristics of the permeation and concentrate produced.

The critical flux was determined by means of the adapted flux-step method [19]. Initially, the membranes were chemically cleaned, and then the critical flux test was initiated, and the pressure was checked

for constant flux levels. For each flow rate, the filtration time was 15 min, after which the flow rate was increased. The critical flux corresponded to the value at which the pressure applied increased over 15 min of constant flux permeation.

The NF membrane was submitted to maintenance cleaning procedures at every 14-d period by using the combination of 500 mg/L sodium percarbonate solution for 4 h and a citric acid solution with pH lower than 2 for 30 min, allowing a 99% recovery of hydraulic permeability of the membrane (according to previous research carried out by the same authors of this study). There was no need for recovery cleaning for the NF membrane during the pilot unit operation.

2.3. Analytical procedures

The feed and permeation of the MBR and NF were ascertained periodically as to color (2120-B using a Hach DR2800 Spectrophotometer), total nitrogen (4500-N), conductivity (2510-B), and COD (5220 B), BOD (5210 B), ammoniacal nitrogen (4500-NH₃ C), phosphorus (4500-P B), chlorides (4500-Cl C), nitrite (4500-NO₂), nitrate (4500-NO₃), alkalinity (2320 B), and total solids (2540 B) according to the recommendations of the Standard Methods for Water and Wastewater Examination [20] and humic substances according to the modified Lowry method for protein [21]. The analysis of humic substances consisted in implementing the Lowry method, with and without the addition of

CuSO₄, where the color interference in the test without the addition of CuSO₄ was primarily attributed to the presence of humic substances in the sample.

3. Results and discussions

3.1. Air stripping and MBR

Table 3 shows the physical–chemical characteristics of raw, post-stripping and post-MBR leachate.

It has been noticed that the stripping had high removal efficiency for ammonia (60%), whereas no pH adjustment, or temperature increase was required to favor the equilibrium displacement. Ammonia stripping requires the passage of large quantities of air over the exposed surface of leachate, thus causing the partial pressure of the ammonia gas within the water to change the ammonia from the liquid to the gas phase. Free ammonia begins to be formed when the pH is above 7, but it is favored when the pH is greater than 11 [21]. The increase in the COD leachate after stripping may be due to the concentration reached during the stripping because the long exposure time (48 h) of the leachate to aeration, which favors evaporation.

The MBR showed a small organic matter removal of COD and total organic compounds (44%) considering the removal of the total organic matter concentration in the raw leachate. Nevertheless, if one considers that 48% of the organic matter present in the leachate is of an inert nature, the MBR has then showed excellent performance in the removal of biodegradable organic matter. This result reinforces the demand for post treatment of the leachate. The feasibility of

utilizing MBRs to remove organic matter of old leachate remains one of the primal research objectives of most of these studies [22]. Sadri et al. [23] reported a COD removal efficiency of 54–78% for organic load rates of 0.90–2.74 g COD/Ld. The authors in this study used leachate with BOD/COD ratios as low as 0.1 (indicating a biodegradable fraction of only 10%) coupled with HRTs of only 1–2 d, and achieved remarkable COD removal.

Regarding ammonia removal, 45% of ammonia was removed before feeding the MBR, while the total ammonia removed (stripping + MBR) was up to 79%. This result exceeded the expectations estimated for aerobic MBR, but it may be explained by the fact that MBR has also a significant effect on nitrification as high sludge retention time promotes the growth of nitrifying bacteria, and as well it contributes to the air stripping pre-treatment. Extremely high NH₃-N concentrations may also have an adverse effect on N-removal by MBR. High NH₃-N removal efficiency level (>97%) was primarily obtained when the feed of NH₃-N concentrations were low [23–25]. WWs containing nitrogen concentrations higher than 1,000 mg/L (either in NH₃-N and organic N form) may cause inhibition of nitrifying bacteria growth [26]. Hence, applying ammonia stripping process as a pretreatment may improve the performance of MBRs. It was also noticed high color removal at the MBR, which may be associated with the removal of humic substances.

3.2. Nanofiltration

Dissolved nonbiodegradable organic matters and negatively charged nitrified compounds are effectively

Table 3
Physical–chemical characteristics of raw and treated leachate

Parameter	No. samples	Raw leachate	Post-stripping leachate	MBR permeate	MBR removal
COD, mg/L	86	3,520 ± 750	4,163 ± 828	2,392 ± 951	44 ± 18
TOC, mg/L	48	1,175 ± 281	–	678 ± 184	40 ± 16
Humic substances, mg/L	48	1,639 ± 472	–	622 ± 252	57 ± 16
Color, mg Pt-Co/L	88	1,069 ± 312	1,333 ± 550	726 ± 309	49 ± 21
Total nitrogen, mg/L	48	1,810 ± 364	–	1,133 ± 544	37 ± 25
Ammoniacal nitrogen, mg/L	48	1,310 ± 311	528 ± 310	280 ± 176	45 ± 25
Nitrite, mg/L	45	0.2 ± 0.03	–	540 ± 279	–
Nitrate, mg/L	45	3.3 ± 1.6	–	206 ± 103	–
Phosphorus, mg/L	47	26 ± 6	–	18 ± 5	31 ± 18
Alkalinity, mg/L	74	6,866 ± 1,500	–	642 ± 761	90 ± 11
Conductivity, mS/cm ²	75	19 ± 3	16 ± 3	13 ± 4	30 ± 16
Chloride, mg/L	26	2,463 ± 977	–	1,917 ± 634	21 ± 10
Total solids, g/L	47	8.5 ± 1.1	–	6.9 ± 1.4	20 ± 13
Fixed total solids, g/L	47	6.3 ± 0.8	–	5 ± 1.1	19 ± 14
Volatile total solids, g/L	47	2.3 ± 0.3	–	1.8 ± 0.4	21 ± 15

removed by subsequent NF membranes. The feed COD concentration in the (MBR permeate) and NF permeate was monitored to investigate COD removal performance in the NF process (Fig. 2). As shown in Fig. 2, the feed COD (MBR Permeate) was between 2,675 and 4,864 mg/L. These concentration levels of COD decreased to approximately 114–716 mg/L in the NF permeate, which shows a COD removal ranging from 80 to 96%. The NF process proved to be effective in COD removal, and was able to frame the treated effluent within this parameter according to current Brazilian laws (180 mg/L of COD or removal >55%). The literature reports the possibility of applying a subsequent NF unit to MBRs treating leachate [27,28]. Jakopović et al. [29] used NF membrane on the MBR effluent, and observed an improvement in COD removal efficiency from 23% (MBR alone) to 92% (MBR associated with NF).

According to the results shown in Fig. 3, the form in which the overall process was operated, i.e. without pH correction in the air stripping step, the system was able to remove up to 90% of the ammonia. This efficiency is mainly due to the stage of the air stripping (75%) supplemented by NF that accounted for removing 64% of the ammonia from the MBR permeate, which is equivalent to 11% of the total ammonia present in the effluent. This result can be explained by the fact that as the farther as they were from their isoelectric point, which is approximately between 3.0 and 4.3, the higher the charge was, thus helping the ammonium ion retention (NH_4^+) in the areas of the negatively charged active layer. Coupling ammonium ions to the membrane active layer results in a positively charged superficial layer, and those charged functional groups attract ions of the opposite charge [30], which manages the repulsion of other ammonium ions. This explains

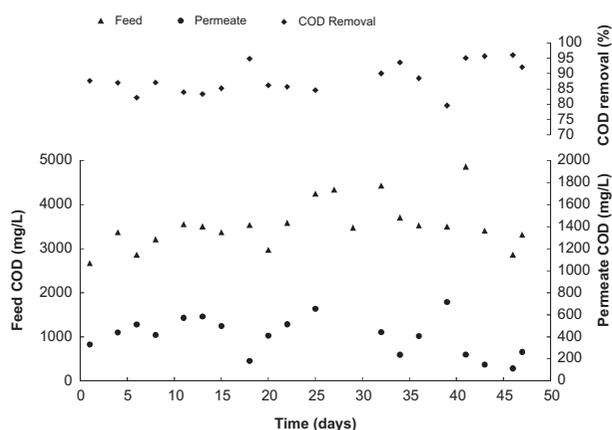


Fig. 2. COD concentrations of MBR permeate, NF permeate, and COD removal efficiency of NF.

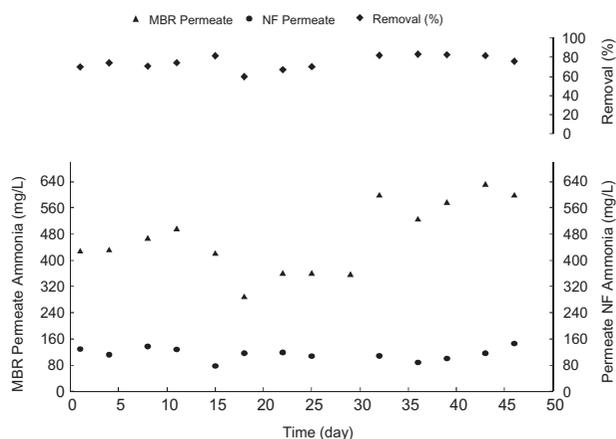


Fig. 3. Ammonia concentrations of MBR permeate, NF permeate, and NF Ammonia removal efficiency.

the maintenance of the ammonia concentration in the NF permeate at relative low value even with an increased ammonia concentration in the feed (MBR permeate).

The contribution of the MBR in ammonia removal is related to the nitrification process (conversion of N-NH_3 to nitrite and nitrate), and corresponds to 45%. Fig. 4 shows that all ammonia removed was converted into nitrite and nitrate, which are unhealthy, and therefore must be removed.

The NF membrane shows nitrite and nitrate removal efficiency ratios of 56 and 31%, respectively. However, the literature has reported that NF90 may remove nitrate within the range between 85 and 95% [31–33]. Although nitrite and nitrate will not be able to speciate, and have poorly complexes with others species, a variety numbers of co-ions existing in raw effluents and the complicated interactions between nitrite and nitrate, and as well the membrane make it difficult to predict the efficiency of a NF membrane to remove nitrite and nitrate. In the case of landfill

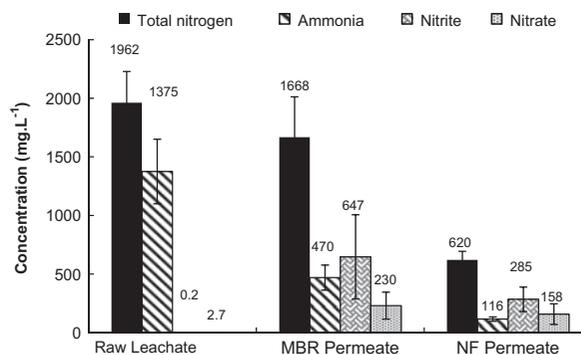


Fig. 4. Profile of nitrogen forms in the treatment system.

Table 4
Nitrite and nitrate properties [34,35]

Parameter	Nitrite	Nitrate
Ionic radius (R_{ion} , 10^{-10} m)	3	3
Hydrated radius (R_{hyd} , 10^{-10} m)	5	5.1
Hydration free energy (ΔG_{hyd} , kcal/mol)	-81	-73
Bulk diffusion coefficient, 25°C (D_w , 10^{-9} m ² /s)	1.91	1.9

leachates, this argument is strengthened by its diversified composition. The higher removal of nitrite compared to nitrate removal may be associated with its higher hydration energy since their sizes are similar (Table 4). A more recently proposed process for ions removal in NF is the partial or full dehydration of hydrated solutes during transport. According to Richards et al. [34], the ions fully or partially dehydrate from bound water molecules in their hydration shell in order to pass through the membrane. Dehydration could occur as a result of a limited space availability as the pore entrance is smaller than the fully hydrated ion, and it is due to forces on the ion that pressure it, so squeezing it into the membrane, and also the concentration gradient and/or the temperature, which results in losing bound water molecules. If dehydration would take place during the passage, the retention trends and the magnitude of energy barriers would be correlated with the hydration free energy of the ions and the amount of dehydration required for each. Higher hydration energy of nitrite implies that the energetic expense of stripping water molecules from nitrite's hydration shell is higher than nitrate, making transport more difficult.

Fig. 5 shows the color removal by NF membrane. It may be seen that NF shows 99% of color removal

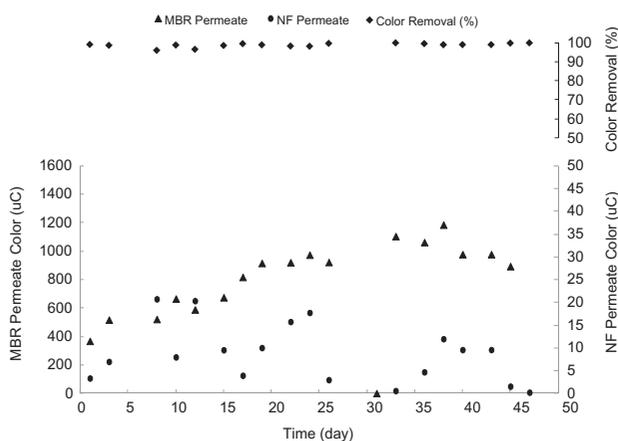


Fig. 5. Colors of MBR permeate and NF permeate, and color efficiency removal by NF.

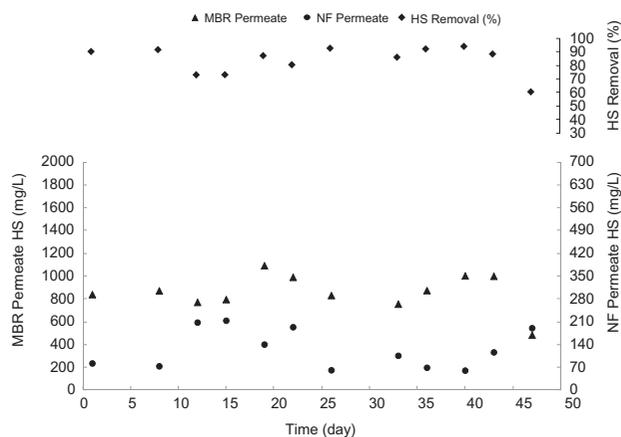


Fig. 6. Humic substances in MBR permeate, and NF permeate, and its removal efficiency by NF.

efficiency. The color removal may be related to humic substance removal as the color range, from dark brown to black color, of landfill leachate is due to the presence of humic substances [36,37]. Humic substances are natural organic matters made of complex structures of polymerized organic acids, carboxylic acids, and carbohydrates [38]. Fig. 6 shows humic substances removal by NF.

The NF membrane provides an average removal efficiency of humic substances of 83%. Content of humic substances in leachate may vary considerably depending on the landfill age. In mature landfills, humic substances (humic acids, fulvic acids, and humins) prevail in the organic fraction of methanogenic leachate by as much as 60% [39]. Concentrations of humic acids in the range of units up to tens of mg/L in landfill leachates were reported in previous studies [40,41]. The content of humic acids also depends substantially on the type of waste deposited, and as well on the type of landfill, and that is why in this study concentration has been presented in hundreds of mg/L like in other studies [42]. Humic acids have been identified as some of the most prevailing foulants found in NF membranes [42], and have caused significant losses to productivity, besides increasing operational costs. The increase of humic substances contents during the 36th and 47th d may be related to the increase in fouling in NF membrane (Fig. 7). The fouling may be caused by adsorption of humic acid on the membrane surface, and also by pore blockage.

Due to the aromatic and aliphatic components of humic acid, the organic fouling layer was expected to be quite hydrophobic. This hydrophobic fouling layer has the potential to reduce the membrane permeability and has an influence upon the separation of humic substances by the NF filtration processes. Xu et al. [43]

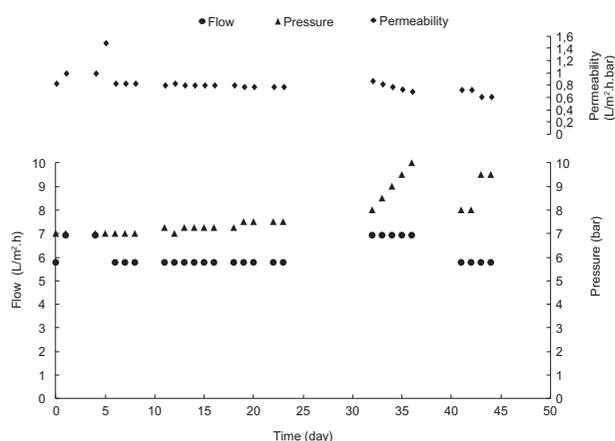


Fig. 7. Profiles of flow, pressure and permeability over the NF monitoring period.

also reported an increase in rejection of hydrophobic trace organics by humic acid fouled membranes. The formation of a hydrophobic membrane fouling layer may isolate and hinder the interactions between the membrane and solutes. This could result in less solute partitioning and diffusion across the membrane, which has been reported as a major transport mechanism of hydrophobic solutes in NF and RO filtration processes [44].

The NF membrane was operated with a flux of 5.8 L/mh, except during 32nd–36th d, on which the flux was increased up to 6.9 L/mh. Although the permeability value is similar to value in the literature, 0.9 L/mh, this flux value is lower than the expected flux for NF membranes, 18–80 L/mh [1], due to the fouling potential of the leachate after MBR. Rautenbach and Mellis [45] evaluated the use of NF to treat MBR effluents, and also operated it with low flux (7–12 L/mh). During the first 23 d of monitoring, the pressure was relatively stabilized, which indicated low fouling potential and required only a maintenance cleaning procedure at every 14-d period in order to keep the permeability values steady. The increase in the flux up to 6.9 L/mh in the 32nd day resulted in a significant fouling rate increase. This increase may be imputed to the operation of the NF membrane in supercritical conditions. During this period, the critical flux value was determined by using the flux-step method proposed by Bacchin et al. [19], and a critical flux value of 6.7 L/mh was observed. These results show the importance of maintaining the flux below the critical flux for a more sustainable membrane operation. The permeate flux was again reduced to 5.8 L/mh, and it was observed an increase of pressure in order to maintain a constant flux due to membrane fouling that may be related to the increased concentra-

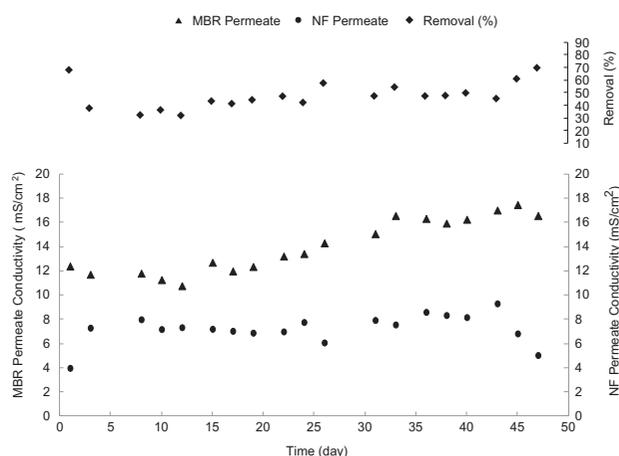


Fig. 8. Values of conductivity for MBR permeate and NF permeate, and removal efficiency.

tion of humic substances as discussed above. The membrane fouling over that period cannot be associated with inorganic fouling, since no significant variation was observed in the concentration of salts in the NF feed during such monitoring period (Fig. 8).

The increased conductivity removal from 45.4 to 69.7% after day 43rd may be associated with salt concentration increase in the feed during that period, which contributes to concentration polarization. The concentration polarization layer contributes to a greater removal of salts due to electrostatic interactions.

3.3. NF membrane for more landfill leachate treatment sustainability

Landfill leachate management has been a major environmental, economic, and social concern, and its treatment brings forth as a challenge. The stripping, MBR, and NF route evaluated in this study shows the importance of NF to improve the effluent quality and generate effluents treated with no chemical products and chemical sludge generation. The landfill leachate treated along such a route may be reused in the landfill as water for dust containment and in earthworks at construction site works. Table 5 summarizes the treated landfill leachate characteristics and shows the contribution of NF to such a high efficiency.

In Brazil, there is no specific law to set the standards and parameters for reusing water. Among the few initiatives in such a context, the most noteworthy is the standard of Brazilian Association of Technical Standards—ABNT NBR (13.969/97) that sets the standards for direct reuse of strictly treated domestic sewage. Sautchuk et al. [46] suggested as standards for

Table 5
Physical–chemical characteristics of the raw and treated leachate

Parameter	Raw leachate	MBR permeate	NF permeate	NF removal	Overall removal (%)
COD, mg/L	3,961 ± 682	3,540 ± 545	404 ± 173	88 ± 5	91 ± 5
BOD, mg/L	271.5 ± 142	179 ± 106	73 ± 42	53 ± 21	71 ± 10
Color, uC	1,043 ± 246	985 ± 379	3 ± 4	99 ± 1	100 ± 1
pH	8.6 ±	7.4 ± 1.7	7.8 ± 1.9	–	–
Ammonia, mg/L	1,375 ± 275	470 ± 107	116 ± 19	75 ± 7	91 ± 2
Phosphorus, mg/L	25 ± 5	16 ± 3	2 ± 1	90 ± 10	93 ± 7
Alkalinity, mg/L	7,758 ± 1,068	1,795 ± 1,255	236 ± 200	87 ± 7	97 ± 4
Conductivity, mS/cm ²	17.2 ± 1	4.6 ± 1	73.5 ± 4	78.1 ± 4	
Chloride, mg/L	2,945 ± 257	2,478 ± 261	367 ± 126	85 ± 5	97 ± 1
TS, g/L	9.3 ± 0.2	7.8 ± 0.7	2.1 ± 0.9	73 ± 13	79 ± 12
VTS, g/L	2.4 ± 0.2	2.1 ± 0.3	0.3 ± 0.1	86 ± 9	97 ± 2
TSS, mg/L	6.9 ± 1.2	6 ± 1	2 ± 1	68 ± 16	82 ± 11

reuse of water for dust arrestment at landfills and earth works the control of coliforms to smaller than 1,000/mL, pH between 6.0 and 9.0, no unpleasant smell and appearance, less than 1.0 mg/L of oils and greases, BOD lower than 30 mg/L, VOC absent and suspended solids less than 30 mg/L. The fraction of oil and grease and fecal coliform bacteria was not measured in this study. However there is no record of significant presence of oil and grease in leachate, and presence of coliforms is not expected in the NF permeate. For other parameters, the attendance to standards suggested is observed.

4. Conclusions

In this study, the suggested landfill treatment configuration consists in an association of air stripping, MBR, and NF membrane. By this configuration, leachate has shown good enough quality standards for reuse applications such as water for dust arrestment and in earthworks in construction site works. The NF was extremely important to ensure high efficiency of the system. The permeability reduction in NF membrane was relatively low and acceptable for the effluent MBR, and was attributed to humic substance fouling. The low potential fouling also was due to an operation performed under subcritical conditions, which has been important to ensure low demand for chemical cleaning procedures, which is also important to ensure longer membrane lifetime. As future aspects worth highlighting the importance of research sustainable techniques for treatment and final disposal of the NF concentrate and strategies to improve the permeability of the membrane.

References

- [1] S. Renou, J.G. Givaudan, S. Poulain, F. Dirassouyan, P. Moulin, Landfill leachate treatment: Review and opportunity, *J. Hazard. Mater.* 150(3) (2008) 468–493.
- [2] E. Durmusoglu, C. Yilmaz, Evaluation and temporal variation of raw and pre-treated leachate quality from an active solid waste landfill, *Water, Air Soil Poll.* 171 (2006) 359–382.
- [3] W.Y. Ahn, M.S. Kang, S.K. Yim, K.H. Choi, Advanced landfill leachate treatment using an integrated membrane process, *Desalination* 149 (2002) 109–114.
- [4] A. Amokrane, C. Comel, J. Veron, Landfill leachates pretreatment by coagulation-flocculation, *Water Res.* 31 (1997) 2775–2782.
- [5] F. Kargi, M.Y. Pamukoglu, Adsorbent supplemented biological treatment of pretreated landfill leachate by fed-batch operation, *Bioresour. Technol.* 94 (2004) 285–291.
- [6] M. Van Praagh, J. Heerenklage, E. Smidt, H. Modin, R. Stegmann, K.M. Persson, Potential emissions from two mechanically–biologically pretreated (MBT) wastes, *Waste Manage.* 29 (2009) 859–868.
- [7] J. Bohdziewicz, A. Kwarcia, The application of hybrid system UASB reactor-RO in landfill leachate treatment, *Desalination* 222 (2008) 128–134.
- [8] S.K. Gupta, G. Singh, Assessment of the efficiency and economic viability of various methods of treatment of sanitary landfill Leachate, *Environ. Monit. Assess.* 135 (2007) 107–117.
- [9] F. Feki, F. Aloui, M. Feki, S. Sayadi, Electrochemical oxidation post-treatment of landfill leachates treated with membrane bioreactor, *Chemosphere* 75 (2009) 256–260.
- [10] L. Vandijk, G.C.G. Roncken, Membrane bioreactors for wastewater treatment: The state of the art and new developments, *Water Sci. Technol.* 35 (1997) 35–41.
- [11] O. Kwon, Y. Lee, S. Noh, Performance of the NF-RDM (Rotary Disk Membrane) module for the treatment of landfill leachate, *Desalination* 234 (2008) 378–385.
- [12] K. Linde, A.S. Jönsson, Nanofiltration of salt solutions and landfill leachate, *Desalination* 103 (1995) 223–232.

- [13] H.Y. Holds, J. Yacubowicz, Nanofiltration: Properties and uses, *Filtr. Separat.* (2005) 16–21.
- [14] C.Y. Tang, Y.N. Kwon, J.O. Leckie, Fouling of reverse osmosis and nanofiltration membranes by humic acid—Effects of solution composition and hydrodynamic condition, *J. Membr. Sci.* 290 (2007) 86–94.
- [15] J.D. Ritchie, E.M. Perdue, Proton-binding study of standard and reference fulvic acids, humic acids and natural organic matter, *Geochim. Cosmochim. Acta* 67 (2003) 85–96.
- [16] J. Harmsen, Identification of organic compounds in leachate from a waste tip, *Water Res.* 17 (1983) 699–705.
- [17] J. Artiola-Fortuny, W.H. Fuller, Humic substances in landfill leachates: I. Humic acid extraction and identification, *J. Environ. Qual.* 11 (1982) 663–668.
- [18] E. Germili, D. Orhon, N. Artan, Assessment of the initial inert soluble COD in industrial wastewaters, *Water Sci. Technol.* 23 (1991) 1077–1086.
- [19] P. Bacchin, P. Aimar, R.W. Field, Critical and sustainable fluxes: Theory, experiments and applications, *J. Membr. Sci.* 281(1–2) (2006) 42–69.
- [20] APHA, AWWA, WEF, Standard Methods for the Examination of Water and Wastewater, twenty-first ed., American Public Health Association, Washington, DC, 2005.
- [21] B. Frølund, T. Griebe, P.H. Nielsen, Enzymatic activity in the activated-sludge floc matrix, *Appl. Microbiol. Biotechnol.* 43(4) (1995) 755–761.
- [22] B. Wichitsathian, S. Sindhuja, C. Visvanathan, K.H. Ahn, Landfill leachate treatment by yeast and bacteria based membrane bioreactors, *J. Environ. Sci. Heal. A* 39 (2004) 2391–2404.
- [23] S. Sadri, N. Cicek, J. Van Gulck, Aerobic treatment of landfill leachate using a submerged membrane bioreactor—Prospects for on-site use, *Environ. Technol.* 29 (2008) 899–907.
- [24] N. Laitinen, A. Luonsi, J. Vilen, Landfill leachate treatment with sequencing batch reactor and membrane bioreactor, *Desalination* 191 (2006) 86–91.
- [25] J. Tsilogeorgis, A. Zouboulis, P. Samaras, D. Zamboulis, Application of a membrane sequencing batch reactor for landfill leachate treatment, *Desalination* 221 (2008) 483–493.
- [26] W. Ahn, M. Kang, S. Yim, K. Choi, Advanced landfill leachate treatment using an integrated membrane process, *Desalination* 149 (2002) 109–114.
- [27] G. Li, W. Wang, Q. Du, Applicability of nanofiltration for the advanced treatment of landfill leachate, *J. Appl. Polym. Sci.* 116 (2010) 2343–2347.
- [28] M. Ince, E. Senturk, G. Onkal Engin, B. Keskinler, Further treatment of landfill leachate by nanofiltration and microfiltration—PAC hybrid process, *Desalination* 255 (2010) 52–60.
- [29] H.K. Jakopović, M. Matošić, M. Muftić, M. Čurlin, I. Mijatović, Treatment of landfill leachate by ozonation, ultrafiltration, nanofiltration and membrane bioreactor, *Fresen. Environ. Bull.* 17 (2008) 687–695.
- [30] S. Chellam, J.S. Taylor, Simplified analysis of contaminant rejection during ground and surface water nanofiltration under the information collection rule, *Water Res.* 35(10) (2001) 2460–2474.
- [31] B. Van der Bruggen, M. Mänttari, M. Nyström, Drawbacks of applying nanofiltration and how to avoid them: A review, *Sep. Purif. Technol.* 63 (2008) 251–263.
- [32] F. Garcia, D. Ciceron, A. Saboni, S. Alexandrova, Nitrate ions elimination from drinking water by nanofiltration: Membrane choice, *Sep. Purif. Technol.* 52 (2006) 196–200.
- [33] A. Santafé-Moros, J.M. Gozálviz-Zafrilla, J. Lora-García, Nitrate removal from ternary ionic solutions by a tight nanofiltration membrane, *Desalination* 204 (2007) 63–71.
- [34] L.A. Richards, A.I. Schäfer, B.S. Richards, B. Corry, The importance of dehydration in determining ion transport in narrow pores, *Small* 8 (2012) 1701–1709.
- [35] A. Pinotti, N. Graiver, A. Califano, N. Zaritzky, Diffusion of nitrite and nitrate salts in pork tissue in the presence of sodium chloride, *Food Eng. Phys. Prop.* 67 (2006) 2165–2171.
- [36] H.A. Aziz, S. Alias, M.N. Adlan, F.A.H. Faridah, A. Asaari, Colour removal from landfill leachate by coagulation and flocculation processes, *Bioresour. Technol.* 98 (2007) 218–220.
- [37] B. Langlais, D.A. Reckhow, D.R. Brink (Eds.), *Ozone in Water Treatment: Application and Engineering*, Lewis Publishers, Inc, Chelsea, MI, 1991.
- [38] J. Artiola-Fortuny, W.H. Fuller, Humic substances in landfill leachates: I. Humic acid extraction and identification, *J. Environ. Qual.* 11 (1982) 663–668.
- [39] J.B. Christensen, D.L. Jensen, C. Grøn, Z. Filip, T.H. Christensen, Characterization of the dissolved organic carbon in landfill leachate-polluted groundwater, *Water Res.* 32 (1998) 125–135.
- [40] Y. Wu, S. Zhou, F. Qin, H. Peng, Y. Lai, Y. Lin, Removal of humic substances from landfill leachate by Fenton oxidation and coagulation, *Process Saf. Environ.* 88 (2010) 276–284.
- [41] C. Berthe, E. Redon, G. Feuillade, Fractionation of the organic matter contained in leachate resulting from two modes of landfilling: An indicator of waste degradation, *J. Hazard. Mater.* 154 (2008) 262–271.
- [42] C.Y. Tang, Y.N. Kwon, J.O. Leckie, Fouling of reverse osmosis and nanofiltration membranes by humic acid—Effects of solution composition and hydrodynamic conditions, *J. Membr. Sci.* 290 (2007) 86–94.
- [43] P. Xu, J.E. Drewes, T.U. Kim, C. Bellona, G. Amy, Effect of membrane fouling on transport of organic contaminants in NF/RO membrane applications, *J. Membr. Sci.* 279(1–2) (2006) 165–175.
- [44] C. Bellona, J.E. Drewes, P. Xu, G. Amy, Factors affecting the rejection of organic solutes during NF/RO treatment—A literature review, *Water Res.* 38(12) (2004) 2795–2809.
- [45] R. Rautenbach, R. Mellis, Waste water treatment by a combination of bioreactor and nanofiltration, *Desalination* 95 (1994) 171–188.
- [46] C. Sautchuk, H. Farina, I. Hespanhol, L.H. Oliveira, L.O. Costi, M.S.O. Ilha, O.M. Gonçalves, S. May, S.S.N. Boni, W. Schmidt, *Conservação e Reuso da Água em Edificações (Water reuse and conservation in buildings)*, Manual—Sindicato das Construções (SINDUSCON), São Paulo, 2005.