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Phthalates accumulation inside an anaerobic membrane bioreactor for landfill leachate treatment

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ABSTRACT

In this study, the treatment of landfill leachate by an anaerobic membrane bioreactor (AnMBR) was investigated. The theoretical organic loading rate in the reactor was gradually increased from 4 to 10 g COD/L.d. During the optimal organic loading rate (7 g COD/L.d), the AnMBR achieved 94% of COD removal with 0.39 L biogas/g of COD removed. However, increasing the OLR to 8 g COD/L.d has led to the decrease of the reactor performance in terms of COD removal efficiency and biogas production. Organic pollutants in the raw leachate, during AnMBR operation, and in the permeate were analyzed through a gas chromatography coupled with mass spectrometry (GC–MS) system. GC–MS analysis revealed that organics were efficiently degraded inside the reactor. However, concentration of phthalates increased to reach 5.79 g/L inside the reactor at the end of the treatment process while their abundance in the permeate was lower. This study highlighted the correlation between the irreversible toxicity observed during the highest organic loading rates and phthalates accumulation inside the AnMBR.

Keywords: Anaerobic membrane bioreactor; GC–MS; Landfill leachate; Phthalates; Toxicity

1. Introduction

The sanitary landfill method for the ultimate disposal of solid waste material continues to be widely accepted and used due to its economic advantages [1]. However, solid waste landfills may cause severe environmental impacts if leachate and gas emissions are not controlled [2]. Leachate is a liquid formed primarily by the percolation of precipitated water through an open landfill or through the cap of a completed site [3]. At present, collection and treatment of landfill leachates is one of the most pressing issues surrounding the operation of landfill sites. One of the available options is biological leachate treatment by either aerobic or anaerobic processes. Anaerobic treatment methods are more suitable for concentrated leachate streams, offer lower operating costs, allow the production of a useable biogas product, and the production of a pathogen-free solids residue which can be used as cover material [4]. Long solid retention time is a crucial prerequisite for successful implementation of anaerobic treatment processes due to the slow growth rate of methanogenic bacteria. It is therefore

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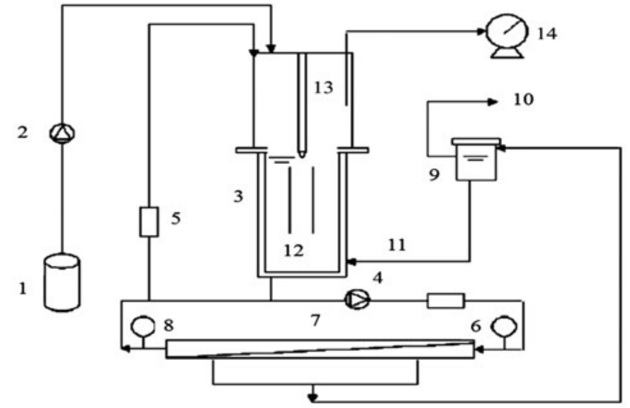

Fig. 1. Schematic diagram of the experimental set-up. Notes: 1: raw landfill leachate reservoir, 2: peristaltic feed pump, 3: jet flow anaerobic reactor, 4: circulation pump, 5: flow meter, 6: manometer, 7: ultrafiltration membrane, 8: manometer, 9: permeate tank, 10: permeate discharged, 11: permeate recycling, 12: inner tube, 13: nozzle, 14: gas flow meter.

unsurprising that anaerobic membrane bioreactors, (AnMBRs) a combination of anaerobic degradation and membrane filtration, are currently receiving increasing attention from both scientists and industrialists dealing with wastewaters treatment [5]. However, LFL often contains components which inhibit anaerobic digestion. In fact, LFL constitutes a very complex mixture, which may contain a large number of xenobiotic organic compounds encountered in the solid waste disposal site or formed as a result of chemical and biological processes within the landfill [6]. Among them, phthalic acid esters (PAE), commonly called phthalates, are usually detected at high concentrations in LFL ranging from 0.7 to 17,200 μg/L [7]. PAE are synthetic substances which constitute potentially hazardous pollutants due to their widespread occurrence in the environment. In fact, many consumer products contain specific members of this family of chemicals, including building materials, household furnishings, clothing, cosmetics, pharmaceuticals, nutritional supplements, medical devices, dentures, children's toys, glow sticks, modeling clay, food packaging, automobiles, lubricants, waxes, cleaning materials, and insecticides. Once PAEs enter in the environment, they partition between air, water, soil, and sediments. However, given the low solubility and high hydrophobic nature of these compounds, they will preferentially be sorbed to the organic fraction of soil or sediments, as well as to the organic matter suspended in water [8–10]. Widespread occurrence of PAE in the environment raised concern about their possible toxicity to humans and other organisms since some of them are considered as potential carcinogens, teratogens, and mutagens and their occurrence in leachate may pose a threat to receiving water quality [9]. In spite of their rapid aerobic metabolization, the biodegradation of PAEs under anaerobic conditions is still controversial. To date, many studies exist on the anaerobic biodegradation rates of PAE, especially of the easily biodegradable ones, whereas the higher molecular weight PAE have reported to be non-biodegradable under methanogenic conditions [8].

This study investigated the treatment of landfill leachate, sampled from the controlled discharge of "Jebel Chakir" in an AnMBR and the correlation between the irreversible toxicity observed during the highest organic loading rates and phthalates accumulation inside the AnMBR.

2. Methods

2.1. Landfill leachate

LFL was collected from the controlled discharge of Jebel Chakir. The site of Jebel Chakir is the first controlled discharge in Tunisia, located at 10 km from the western south of Tunis. It receives 2,000 tons of domestic and assimilated waste per day coming from great Tunis since 1999. Approximately, 68% of disposed wastes are of organic matter origin (food waste) [11]. Leachate was collected in 20-L tanks and transported to the laboratory to be stored at 4°C. The average composition of the sampled leachate is given in Table 1.

2.2. AnMBR and operational scheme

The AnMBR was previously described by Zayen et al. [11] and Mnif et al. [12]. The jet flow anaerobic bioreactor is constructed of Plexiglass having a working volume of 50 L and coupled with a cross-flow ultrafiltration membrane with 1 m^2 area and 100 kDa cut-off. The temperature was maintained constant at 37°C (Fig. 1). The seed sludge was obtained from a full-scale anaerobic wastewater treatment plant.

In a previous study, the same AnMBR was operated for more than five months. The OLR was increased, in a step wise manner, from 1 to 6.27 g COD/L.d. The AnMBR has showed a good performance [11]. After two months of rest, the same AnMBR was restarted. The HRT was kept constant (HRT = 7 d) during all the treatment process and the OLR was increased by the decrease of the dilution of the feed solution.

Table 1
Characteristics of raw LFL and permeate during the optimal OLR compared to the Tunisian Standard for disposal in the public hydraulic field

Parameter	Raw LFL	Permeate during the optimal OLR (7 g COL/L.d)	Tunisian standard for disposal in the public hydraulic field
pH (25˚C)	6.84	8.295 ± 0.24	6.5–8.5
CE (mS/cm)	24.9	–	–
COD (g/L)	84.29	3.33 ± 0.58	0.09
BOD_5 (g/L)	46.1	1.2 ± 0.35	0.03
BOD_5/COD	0.58	–	–
TSS (mg/L)	1970	ND	0.03
VSS (mg/L)	1,460	ND	–
TKN (mg/L)	3,177	–	–
$[NH_4^+]$ (mg/L)	2,800	–	1
VFA (g/L)	22.79	0.762 ± 0.03	–
Ca (mg/L)	975.5	–	500
Cu (mg/L)	533.3	–	0.5
Fe (mg/L)	67.75	–	1
K (mg/L)	8.02	–	50
Mg (mg/L)	14.37	–	200
Na (mg/L)	20	–	–

Note: ND - Not detected.

Table 2
Operating conditions and reactor performance

	Run 1	Run 2	Run 3	Run 4	Run 5
HRT (d)	7	7	7	7	7
Theoretical OLR (g COD /L.d)	4	5.6	7	8	10
COD feed	36.01 ± 4.33	48.39 ± 2.78	55.22 ± 1.71	64.66 ± 5.22	72.89 ± 2.49
COD removal (%)	90.05 ± 2.29	92.97 ± 1.29	93.97 ± 1.85	90.9 ± 2.044	86.3 ± 0.81
Gas production rate (L biogas/g $COD_{removed}$)	0.19	0.21	0.39	0.36	0.27
$[VFA]_{reactor}$ (g/L)	0.13 ± 0.01	0.101 ± 0.029	0.762 ± 0.03	0.937 ± 0.07	1.24 ± 0.18

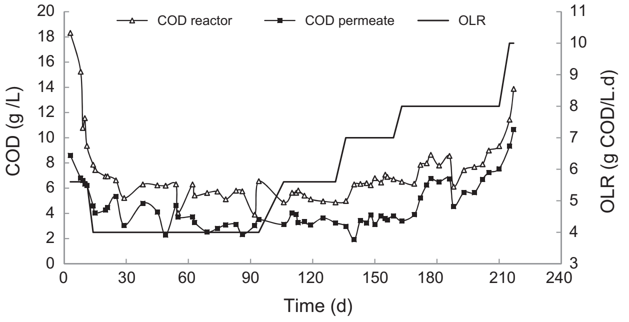

Fig. 2. COD removal efficiency in the AnMBR during the treatment process.

2.3. Analytical methods

The pH and the conductivity were determined using a pH meter model Istek-NeoMet and a conductivimeter model CONSORT C 831, respectively. Chemical oxygen demand (COD) was determined according to Knechtel et al. [13]. Five-day biological oxygen demand (BOD_5) was determined by the manometric method with a respirometer (BSB-controller Model 620 T (WTW)). Total Kjeldahl Nitrogen (TKN) and ammonia nitrogen (NH_4^+) were determined as described by Kjeldahl et al. [14]. Total suspended solids (TSS) and volatile suspended solids (VSS) were determined according to the standard methods [15]. Volatile fatty acids (VFAs) were analyzed by a gas chromatograph (SHIMADZU GC-9A) equipped with a flame ionization detector (SHIMADZU CR 6A) as previously described by Saddoud et al. [16]. Heavy metals concentrations were determined by flame atomic absorption spectrometry as described previously by Ellouze et al. [17]. Biogas was quantified daily by the use of a biogas flow meter (Gallus 2000, Germany).

2.4. Extraction with Ethylacetate and GC–MS analysis

40 mL samples of LFL were extracted two times with Ethylacetate (EtAc) (1:2 volumes). The extract was concentrated in vacuum to dryness at 40˚C and the residue obtained was redissolved in 1 mL of EtAc. After EtAc extraction, a derivation step was carried out by silylation: 200 μL volumes of extract were reconstituted with 100 μL of pyridine and 150 μL of BSTFA (Bis trimethylsilyl trifluoro acetamide. $C_8H_{18}F_3NOSI_2$). The reaction tube was introduced into a water bath of 80˚C for 2 h. After derivation, the resultant solution was injected into the chromatograph for GC–MS analysis.

An aliquot of 1 μL of the derivated extract was injected splitless into the GC/MS (5975B inert MSD Agilent). The data were obtained on a DB-5MS column, 30 m length, 0.25 mm i.d., and 0.25 mm thickness (Agilent Technologies, J&W Scientific Products, USA). Carrier gas was helium. GC oven temperature started at 100˚C and was held for 1 min at 260˚C and then for 10 min with program rate 4˚C/min. The injector and detector temperatures were set at 250˚C and 230˚C, respectively. The mass range was scanned from 50 to 550 amu. The control of the GC/MS system and the data peak processing were carried out by means of the MSDCHEM Software.

The external standard quantification method was employed to quantify the different components in LFL samples. The compounds are quantified using the relation between the analyte response and that of the external standard (peak area). Bis-(2-ethylhexyl) phthalate (DEHP) has been chosen as an external standard since it was abundant in all analyzed LFL samples. Sample concentrations were calculated on the basis of peak areas compared to that of the external standard.

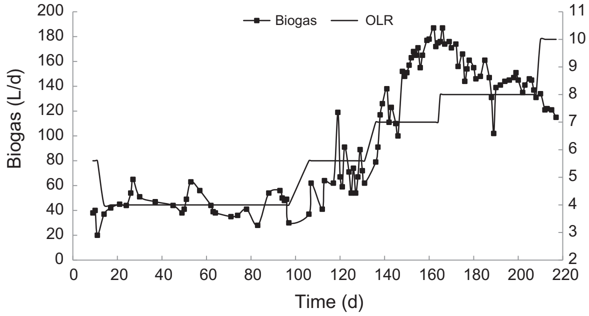

Fig. 3. Biogas production in the AnMBR during the treatment process.

3. Results

3.1. LFL characterization

The characteristics and average composition of the raw leachate are given in Table 1.

The concentrations of COD were measured as soluble COD in this study. The raw landfill leachate had a high BOD_5/COD ratio of 0.58 with high concentration of VFA exceeding 22 g/L, which suggested that this LFL was in its acidification stage and thus appropriate to the anaerobic digestion.

3.2. AnMBR performance

The AnMBR performance is evaluated regarding COD removal efficiency (Fig. 2) and biogas production (Fig. 3). Table 2 summarizes the operating conditions and the AnMBR performance during the treatment process.

Since the reactor was previously operated with the same effluent, the OLR was increased starting from 5.6 g COD/L.d. However, the AnMBR showed poor performance regarding COD removal efficiency (< 65%) and biogas production (0.14 L biogas/g COD removed). To allow a better biodegradation inside the bioreactor, the OLR was decreased to 4 g COD/L.d. At the optimal OLR (7 g COD/L.d), the AnMBR achieved the highest COD removal and biogas production (94% and 0.39 L biogas/g COD removed, respectively). Nevertheless, during this OLR, the permeate having more than 3 g/L of COD, exceeded the Tunisian standard for disposal in the public hydraulic field (COD < 90 mg/L) and need to be further treated to meet these standards.

However, increasing the OLR to 8 g COD/L.d has led to the decrease of the AnMBR performance resulting in a lower COD removal and biogas productivity (Figs. 2 and 3).

In a previous study, the composition, diversity, and changes in microbial communities of the same AnMBR were analyzed using the polymerase chain reaction single-strand conformation polymorphism (PCR-SSCP) fingerprint method during the increase of

Table 3
Concentrations of organic pollutants detected in the raw leachate, during treatment process and in the permeate

Abundance (% area)								
			Organic compounds detected in LFL during the treatment process					
Analyte	Ret. time (min)	Raw LFL	0	117	157	182	217	Permeate D 217
Nicotine	3.580	0.2	ND	ND	ND	ND	ND	ND
Benzyl alcohol, TMS	5.208	0.63	ND	ND	ND	ND	ND	ND
Benzoic acid, TMS	5.397	0.24	ND	ND	ND	ND	ND	ND
Cyclohexanecarboxylic acid, TMS	6.078	1.49	ND	ND	2	0.45	0.36	0.35
Heptanoic acid, TMS	6.810	9.82	ND	ND	ND	ND	ND	ND
Octanoic acid, TMS	7.057	1.96	ND	ND	ND	ND	ND	ND
Diethyl phthalate	7.169	ND	ND	ND	ND	ND	0.1	ND
Benzenepropionic acid, TMS	7.394	1.46	ND	ND	ND	ND	ND	ND
1,3-Dihydroxybenzene, 2 TMS	10.467	0.11	ND	ND	ND	ND	ND	ND
Galoxolide	11.297	0.1	ND	ND	ND	ND	0.14	0.11
4-Hydroxyphenylethanol, di-TMS	15.611	0.28	ND	ND	ND	ND	ND	ND
Paracetamol di-TMS	17.356	0.27	ND	ND	ND	ND	ND	ND
Benzene acetic acid, TMS	17.851	1.46	ND	ND	ND	ND	ND	ND
M-trimethylsiloxyphenyl, TMS propionate	19.794	0.15	ND	ND	ND	ND	ND	ND
Butyl, 8-methylnonyl phthalate	35.825	14.54	3.89	25.8	13.42	15.53	28.42	25.3
Bis(2-ethylhexyl) phthalate	40.47	8.06	4.04	6.18	22.07	6.26	8.34	7.84
Di(isodecyl) phthalate	43.634	ND	5.06	5.3	10.28	29.91	48.12	30.5
Di(n-decyl) phthalate	43.664	3.18	2.99	4.78	6.36	8.62	10.04	8.84

Note: TMS - trimethylsilyl ester; ND - not detected.

OLR. This study revealed that, during the highest OLR applied, the bacterial diversity drastically decreased in parallel with the decrease of the AnMBR performance, whereas the archaeal community remained stable [12]. The decline in biogas production observed in the digester during the highest OLR is evidence of a process imbalance which is commonly indicated by a rise in the level of VFAs. However, VFAs concentration during these operating periods was far from its inhibitory level (Table 2). This could be attributed to the accumulation of non- or slowly-biodegradable organic substances inside the bioreactor revealed by the increase of the residual COD inside the reactor.

3.3. *Removal efficiency of pollutants in landfill leachate monitored by GC–MS*

In order to gain insight into the organics in the landfill leachate before, during, and after treatment process leachate contents were analyzed using GC–MS. The minimum percentage of similarity accepted, between the spectra of the sample and from the

Table 4
Decription of phthalates found in LFL during AnMBR operation

Phthalic acid ester	Acronym	Mol. weight (g/mol)	Mol. formula
Diethyl phthalate	DEP	222.24	$C_{12}H_{14}O_4$
Butyl, 8-methylnonyl phthalate	BMNP	362.25	$C_{22}H_{34}O_4$
Bis (2-ethylhexyl) phthalate	DEHP	390.56	$C_{24}H_{38}O_4$
Di(isodecyl) phthalate	DIDP	446.7	$C_{28}H_{46}O_4$
Di(n-decyl) phthalate	DnDP	446.7	$C_{28}H_{46}O_4$

library, was 90%. For values inferior to 90%, the peak was considered as non-identified (Table 3).

As observed in Table 3, the composition of the raw landfill leachate was fairly complex with the presence, among others, of alcohols (benzyl alcohol, 4-hydroxyphenylethanol, and paracetamol), linear carboxylic acids like heptanoic and octanoic acids, aromatic carboxylic acids such as benzoic acid and Benzenepropionic acid, and phthalates. Three phthalates were identified in the raw LFL (butyl, 8-methylnonyl phthalate, bis (2-ethylhexyl) phthalate, and di (n-decyl) phthalate).

GC–MS analysis of LFL during the treatment process shows that most of the identified organic compounds were effectively removed during anaerobic digestion. However, PAEs were not degraded and their abundance increased inside the AnMBR. During the treatment process, five phthalates were detected. Excepting DEP, all the detected PAE were with long alkyl-chains (Table 4). DEP and DIDP were not detected in the raw LFL and were present in LFL during treatment process. In fact, phthalates with longer side chains than DEP are occasionally converted to those with shorter chains by β-oxidation, which removes one ethyl group each time [18].

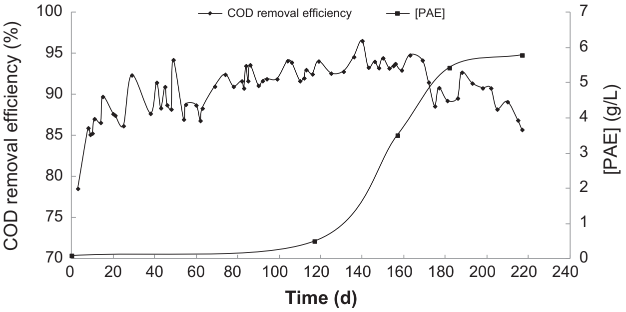

Fig. 4. COD removal efficiency and PAEs concentration inside the AnMBR.

Noticing that PAEs abundance increased inside the reactor during the treatment process, these PAEs were quantified using the previously described method. Fig. 4 shows that PAE concentration increased inside the reactor, especially during the highest OLR applied. It reached 5.79 g/L at the end of the treatment process. 2-D structures of the identified phthalates are represented in (Fig. 5).

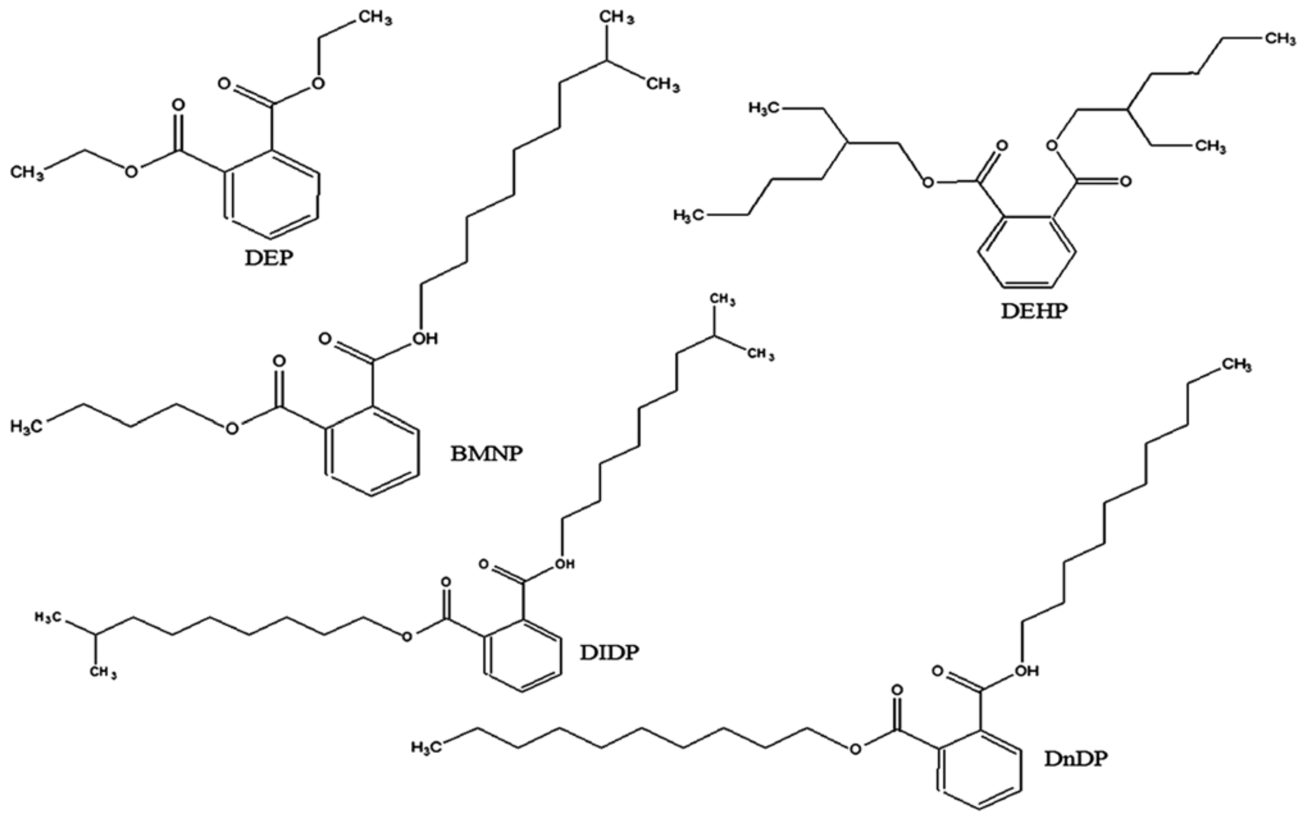

Fig. 5. 2D structures of the identified phthalates.

Numerous studies have demonstrated the biodegradation of several PAEs under aerobic conditions in soil, natural waters, and wastewaters [19]. However, under anaerobic conditions, their degradation is much slower [10]. In addition, PAEs with long ester chains are less susceptible to degradation and some of them are considered recalcitrant [20]. Studies have demonstrated that phthalates with shorter ester chains can be readily biodegraded and mineralized. On the other hand, phthalates with longer ester chains are less susceptible to biodegradation. The biodegradability difference of phthalates is likely due to the steric effect of phthalates side ester chains, which hinders the hydrolytic enzymes from binding to the phthalates and thereby inhibits their hydrolysis [21]. Although DEP is an ester with short alkyl chain, it was detected at the end of the treatment process (Table 3). This could be explained by the inhibition of the microbial community inside the reactor by the phthalate mixture. High levels of long alkyl-chain esters are likely to compromise the anaerobic digestion and the removal of biodegradable PAEs in landfill leachate [20]. Many studies revealed that high concentrations of DEHP in digested sludge are likely to have a negative effect on degradation of other phthalates, such as dibutyl phthalate, diethyl phthalate, and dimethyl phthalate, as well as the biogas production in sludge digesters [8,20].

In this study, the ultrafiltration membrane was a key player regarding the process efficiency. Indeed, all the micro-organisms were retained inside the reactor resulting in an effective consortia and high removal rates. However, the membrane retained also phthalates which resulted in their accumulation inside the reactor to a toxic level. This is revealed by the relative lower abundance of PAEs in the permeate at the end of the treatment process (Table 3). Due to their high hydrophobicity and low solubility, a part of PAEs was sorbed on the sludge and thus retained by the ultrafiltration membrane. Bodzek et al. investigated the removal of phthalates using pressure-driven membrane processes. This study revealed that ultrafiltration appeared to be of great effectiveness in phthalate separation from water [22].

The concentration of PAEs inside the AnMBR increased to reach high concentrations levels in parallel with the decrease of the reactor performance (Fig. 4), especially regarding biogas production efficiency. This resulted in the decrease of the bacterial diversity observed during the highest OLR applied and described in a previous study [12]. Thus, the decrease of the AnMBR performance can be correlated with the accumulation of PAE during the anaerobic digestion of LFL.

4. Conclusion

During the optimal OLR applied, the AnMBR achieved a high performance regarding COD removal efficiency (94%) and biogas production efficiency (0.39 L biogas/g COD removed). However, the increasing of the OLR to 8 g COD/L.d led to the decrease of the AnMBR performance.

The monitoring of the organic composition inside the reactor using GC–MS showed the positive correlation between the decrease of the AnMBR performance and the accumulation of phthalates inside the reactor. The concentration of phthalates reached 5.79 g/L at the end of the treatment process.

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