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Efficiency of landfill leachate treatment in a MBR/UF system combined with NF, with a special focus on phthalates and bisphenol A removal

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ABSTRACT

In this study, a pilot-scale membrane bioreactor (MBR) was operated at a municipal solid waste plant (MSWP) to treat a mixture of landfill leachates (LLs) obtained from modern (MP-LLs) and previous (PP-LLs) waste cells. The MBR unit combined anoxic and aerobic zones with external ultra- and nanofiltration (MBR/UF and MBR/UF/NF, respectively). In addition to the removal of macropollutants, special attention was given to phthalates (PAEs) and bisphenol A (BPA). According to the obtained results, the MBR/UF system with acclimated biomass was effective for treating LLs, and the obtained effluent was generally similar in quality to raw municipal wastewater. The MBR biomass showed high potential for BPA and PAEs biodegradation/biotransformation as confirmed by a metagenomic approach. Only a high chloride concentration (1960 mg Cl^-/L), which was twice the value acceptable by Polish regulations for industrial wastewater entering the municipal wastewater system, justifies the additional usage of the NF unit. Notably, a decreasing amount of biodegradable organic matter in MBR influent is expected with time because of changes in the biochemistry of modern waste cells; therefore, an external carbon source would probably be needed to support denitrification. However, the cooccurrence of an aerobic and anaerobic ammonia-oxidizing community with denitrifying bacteria provides the opportunity for advanced removal of nitrogen and organic carbon.

1. Introduction

The 'waste hierarchy' concept has introduced solid waste prevention, followed by waste reuse and recycling, as well as biomass and/or energy recovery (EU, 1999). Nonetheless, in many countries, waste disposal is still an accepted option. The main disadvantage of this method is the generation of highly polluted landfill leachates (LLs), which are significant sources of macro- and micropollutants (Fudala-Ksiazek et al., 2017; Renou et al., 2008). The presence of micropollutants in the LL streams generated by municipal solid waste plants (MSWPs) is mainly because of unsatisfactory solid waste separation at the source level (Boer et al., 2010; Fudala-Ksiazek et al., 2017, 2016). Because residual household wastes are not strictly controlled, materials such as plastic, glass, metals, personal care products, and even medical waste, drugs, and pesticides may enter the residual (mixed) solid waste stream (Fudala-Ksiazek et al., 2017, 2016; Trulli et al., 2018). Thus, potentially hazardous substances can be disposed in landfill waste cells and then emitted via leachates. This is of special concern in countries where the 'waste hierarchy' philosophy focusing on recycling and recovery is relatively new, e.g., new EU members. In those countries, in addition to solid waste collection and management, LLs treatment is undergoing rapid development. Currently, LLs are simultaneously generated by modern cell (MP-LL), which meet EU requirements (generally post-2010 operations), and by previous cell (PP-LL), which are usually arranged ad hoc without any liner or pollution-control systems and with greater disposal of organic wastes.

In general, to meet the standards for direct discharge into surface water or sewage systems, LLs need to be at least pre-treated on site. However, a single-stage treatment is not sufficient for LLs because in addition to their complex composition, their quality and quantity vary with time (Fudala-Ksiazek et al., 2017, 2016; Kulikowska and Klimiuk, 2008; Renou et al., 2008; Wiszniowski et al., 2006). In the case of LLs classified as 'young' (usually generated by waste cells less than 5 years old), biological processes are usually considered; however, because easily biodegradable organic matter decreases with time, LLs obtained from mature landfill waste cells (greater than 5 years old) require combined physical and chemical processes (Fudala-Ksiazek et al., 2017, 2016; Kulikowska and Klimiuk, 2008; Renou et al., 2008).

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LLs are often treated along with municipal wastewater, although tightening the emission limits for wastewater treatment plants (WWTPs) significantly threatens the sustainability of such co-treatment. Thus, in many countries, including Poland, reverse osmosis is usually used to pre-treat LLs on site. However, *sensu stricto*, reverse osmosis is not a specific treatment process, because, although it separates pollutants and generates high-quality permeates, it also generates concentrates (condensed pollutants), which must be treated or discarded. Often, the concentrate is re-injected into the landfill waste cell, which may influence the chemical composition of LLs over time, e.g., through the re-concentration of monovalent ions in the LLs because such ions are hardly or not at all retained in the landfill waste cells (Fudala-Ksiazek et al., 2016). Consequently, treating LLs by reverse osmosis requires additional amounts of chemical reagents, such as sulphuric acid and/or antiscaling products. Thus, in recent years, other processes have been tested as promising methods to solve the overwhelming LL treatment problems (Boonnorat et al., 2016; Fudala-Ksiazek et al., 2018; Liu et al., 2017; Mandal et al., 2017; Wojciechowska, 2017). Notably, MSWP exploiters usually must treat two types of LLs simultaneously: those generated by modern (MP-LLs) and those generated by previous (PP-LLs) landfill waste cells. However, the feasibility of co-treating PP-LLs and MP-LLs remains uncertain.

Because increasing attention is being given to environmentally friendly technologies that combine biological methods with other treatment technologies, this study investigated the efficiency of the on-site co-treatment of MP-LL/PP-LL mixtures in a pilot-scale two-stage membrane bioreactor (MBR) combined with ultrafiltration (MBR/UF) followed by optional nanofiltration (MBR/UF/NF) (Fig. 1). The MBR system is considered more effective than the conventional wastewater treatment process based on activated sludge, especially for micropollutant removal (Grandclément et al., 2017), because of the combination of the activated sludge process with membrane separation. As a result, more efficient solid-liquid separation and longer solid retention time can be obtained. Such conditions highly influence the activated sludge microbial consortia, thereby enabling the development of microorganisms better adapted to the treated wastewater (here, the MP-LL/PP-LL mixture). The presence of easily biodegraded organic carbon in MP-LL may support denitrification in the MBR unit, which combines anoxic and aerobic zones. Additionally, this study verified the effectiveness of the ultrafiltration (MBR/UF) and/or nanofiltration (MBR/UF/NF) steps as an advanced method for removing potential micropollutants. Special attention was given to phthalates (PAEs) and bisphenol A (BPA), which are both suspected endocrine disruptors, because of their anticipated high concentration in the studied LLs (MP-LLs and PP-LLs), as reported in our previous study (Fudala-Ksiazek et al., 2017). To better elucidate the MBR biochemistry, the innovative metagenomic approach was used to analyse the changes in the taxonomic composition of the activated sludge because of the acclimation process. Special attention was given to bacterial (AOB) and archaeal (AOA) ammonia oxidizers and anammox organisms together with denitrifying bacteria because such cooccurrence can provide the opportunity for the efficient removal of nitrogen and organic carbon, including endocrine disruptors. Additionally, in terms of PAEs and BPA removal, sludge adsorption was also considered.

2. Methods

2.1. Experimental setup and reactor operation

A pilot-scale two-stage MBR was installed at a MSWP located in northern Poland (N 54°51′742′ S 18°36′854″). The MSWP serves a metropolitan area of approximately 460,000 people and receives approximately 200,000 Mg of waste annually. Two landfill waste cells are located at the MSWP. The previous waste cell (PP-LL) was in operation from January 2003 to November 2011, and the modern waste cell (MP-LL) has been in operation since November 2011. The waste cells generated approximately 71 m^3 of PP-LL and 39 m^3 of MP-LL daily. Previously, the PP-LL and MP-LL were treated using a reverse osmosis unit, and the permeate was discharged into the local wastewater system while the concentrate (approximately 50 m^3/d) was re-injected into the modern waste cell (for details, see Fudala-Ksiazek et al., 2016 and Table S1). To eliminate operational problems caused by reverse osmosis, pre-treatment of the LLs with the pilot-scale MBR system was tested; the system consisted of an anoxic reactor followed by an aerobic reactor and combined UF and NF units (Fig. 1).

The MBR was inoculated with 1 m^3 of activated sludge (TSS equal to 17 kg/m^3) obtained from the MBR system working at MSWP Cronheim (Ennest Deponie Cronheim) in Germany. This sludge was delivered in a special container and transported at 4 °C for approximately 24 h. The activated sludge had a typical structure for MBR systems, with small, dispersed flocs and very limited numbers of filamentous bacteria. The experiment was divided into two phases: the biomass acclimation phase (1st phase) and the treatment phase (2nd phase). To achieve biomass acclimation in the activated sludge, the MBR was initially fed with a diluted mixture of MP-LL and PP-LL for 14 days, as shown in Fig. 1. Then, for the next 8 weeks, the MBR system was fed only with the MP-LL/PP-LL mixture (550–650 L per day) at a volumetric ratio of 1:1. In the anoxic reactor, 75% phosphoric acid was added to maintain the appropriate C:N:P ratio (100:10:1) because the concentration of phosphorus in the MP-LL/PP-LL mixture, which was lower than the chemical oxygen demand (COD) or TN, could limit biological processes. Within the aerobic reactor, aeration was continuously supplied to maintain the dissolved oxygen (DO) level at 4–5 mg/L. Additionally, if needed, NaOH was dosed to maintain the pH in the range from 7.0 to 7.5. The mixed liquor sludge from the aerobic tank was continuously recirculated (internal recirculation) into the anoxic tank at a rate equivalent to 200–560% of the feed flow rate. The food to microorganism ratio (F/M) was between 0.43 and 1.86 kg COD/kg MLVSS d. The hydraulic retention time in both tanks was maintained at 8–10 days. The sludge age was 25 days because prolonged sludge retention times have been suggested to result in higher microbial diversity and increase the micropollutant removal potential (Besha et al., 2017; Boonnorat et al., 2016).

In this study, the MBR compartment was combined with a single tubular polyethersulfone (PES) UF membrane in a Berghof HyperFlux module, which is used for separating solids (pore size of approximately 30 nm, with molecular weight cut-off of ca. 100 kD). The UF was operated in a sidestream (external to MBR) configuration at a maximal operating pressure of 7–8 bar. The intermittent suction and backwash were set to 9 and 1 min, respectively, (2 h on and off) to maintain an average of 0.35 m^3 of permeate per day. This operation yielded an average membrane permeate flux rate of 0.77 L/m^2/h, which was next directed to the DOW Filmtec™ NF90-400/34i NF unit. The NF module was equipped with a single polyamide thin-film composite membrane with a spiral-wound element and provided a molecular weight cut-off of ca. 200–400 D and a maximal operating pressure of 41 bar. An NF with a pore size of approximately 1 nm ensured the removal of all solids and even some dissolved compounds.

2.2. Sample collection

In the present study, flow-proportional wastewater samples (24 h) were collected twice a week from the MBR influent and effluent taken after the MBR/UF and MBR/UF/NF, as presented in

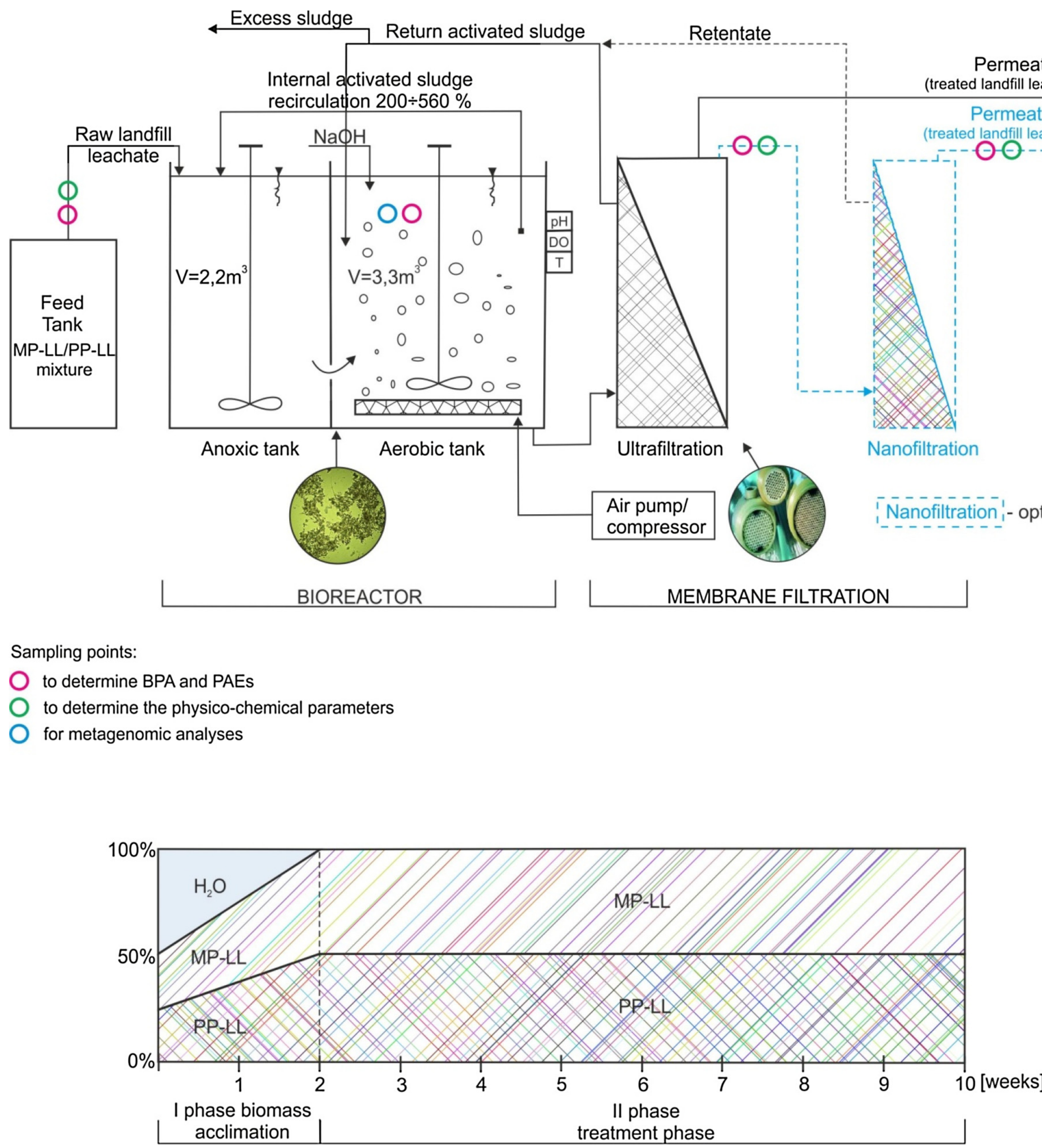

Fig. 1. Scheme of the MBR/UF/NF system indicating the sampling points.

Fig. 1. After collection, the samples were kept on ice and arrived at the laboratory within 3 h. Then, the samples were homogenized and divided into the appropriate number of aliquots as described below. To maintain consistent quality, the physical and chemical parameters of the MBR influent and effluent taken after the MBR/UF and MBR/UF/NF were monitored on an ongoing basis, whereas for the PAEs and BPA analyses, the collected samples were pretreated and kept at −20 °C towards the end of the experiment. In the case of PAEs and BPA, their sorption on activated sludge was also controlled. Therefore, biomass samples were collected from the aerobic tank every two weeks. Then, in each series, part of the sample was centrifuged at 3500 r.p.m. for 10 min (SIGMA 4-16K, SIGMA Labourzentrifugen GmbH, Osterode am Harz, German); next, the obtained supernatant and the whole sample were kept separately for future PAEs and BPA analyses. For metagenomic analyses, the samples of activated sludge used as MBR inoculum (INC-AS) and acclimated activated sludge (ACL-AS) obtained at the end of the experiment were collected and frozen at −80 °C for subsequent DNA isolation and next-generation sequencing.

2.3. Analytical methods

2.3.1. Analyses of physical and chemical parameters

Twice a week, the following physical and chemical characteristics were analysed according to APHA (American Public Health Association, 2005): pH, conductivity (by a portable multi-parameter metre HL-HQ40d multi, HACH, Germany); inorganic N compounds ($N\text{-}NH_4$, $N\text{-}NO_3$, $N\text{-}NO_2$), total phosphorus (TP), orthophosphate ($P\text{-}PO_4$), chemical oxygen demand (COD), chloride (Cl^-), and sulphate (SO_4^{2-}) using a XION 500 spectrophotometer (Dr. Lange, GmbH, Germany); 5- and 20-day biochemical oxygen demand (BOD_5 and BOD_{20}) using the manometric respirometric BOD OxiTop® method; and total suspended solids (TSS) and volatile suspended solids (VSSs) using the gravimetric method.

2.3.2. PAEs and BPA analysis

Once a week, the presence of BPA and selected PAEs (DMP, dimethyl phthalate; DEP, diethyl phthalate; DnBP, di-n-butyl phthalate; BBzP, benzyl butyl phthalate; DEHP, bis(2-ethylhexyl) phthalate; and DnOP, di-n-octyl phthalate) was tested in the MP-LL and PP-LL mixture entering the MBR/UF unit in the effluent of both the MBR/UF and MBR/UF/NF units. Additionally, the sorption of BPA and PAEs on the activated sludge flocs was also monitored. Therefore, samples taken from the aerobic MBR reactor were divided into two parts, one of which was centrifuged to collect the supernatant (as described in Section 2.2). Then, the concentrations of PAEs and BPA were tested in both the entire liquor and in the supernatant. In each sample, the BPA and PAEs contents were determined by the GC/MS technique after prior liquid-liquid extraction with acetonitrile in the presence of inorganic salts. The GC/MS analyses were performed on a semipolar ZB-5MS column in split mode. The ion energy for electron impact (EI) was 70 eV, and mass detection was performed in the single-ion monitoring (SIM) mode. The analytical procedure is described in detail by Fudala-Ksiazek et al. (2017). The selected ions (m/z) used for qualitative and quantitative purposes are shown in Table 1.

2.3.3. Metagenomic analyses

The total genomic DNA was extracted from two activated sludge samples; the first was used as an inoculum for the MBR system and was generated by the L-MP using the commercially available Sherlock AX kit (A&A Biotechnology, Poland). The samples of 15 ml were first transferred into the microcentrifuge tubes containing 0.5 g of 0.5 mm zirconia beads and supplemented with 300 µl of sterile water, 300 µl of L 1.4 buffer and 20 µl of proteinase K. Next, the samples were placed in a Beadbeater for 60 s. The isolation protocol was then followed according to the manufacturer's instructions. The DNA concentrations of the samples were determined by an ND-1000 UV–Vis spectrophotometer. The extracted DNA was stored at 4 °C. The microbial community in the tested INC-AS and ACL-AS samples were analysed using high-speed multiplexed 16S microbial sequencing on a MiSeq platform (Illumina). The V4 region of the 16S rRNA gene was amplified using a F515/R806 primer combination (5′-GTGCCAGCMGCCGCGGTAA-3; 5′-GGACTACHVGGGTWTCTAAT-3′) with Illumina adapter overhang. Libraries were dual-indexed and sequenced on MiSeq in paired-end mode 2 × 250 bp. Taxonomic analysis was performed with MiSeq Reporter v2.3 based on the Illumina-curated GreenGenes v13_5 database using the 16S Metagenomics Workflow.

2.4. Data evaluation

In this study, the statistical data and plots were obtained using R statistical software (R Development Core Team, 2016) and the ggplot2 package (Wickham, 2009), respectively.

3. Results and discussion

The LL treatment is challenging because of the variability and complex characteristics of LLs, especially in terms of organic matter, ammonia and possible micropollutant load (Fudala-Ksiazek et al., 2016; Kalmykova et al., 2014; Renou et al., 2008). Although commonly used in Poland, reverse osmosis generates many technological problems, whereas the co-treatment of LLs with municipal wastewater is of special concern because the Urban Waste Water Treatment Regulations (EU, 1991) and Water Framework Directive (EU, 2008) have posed tighter regulations on WWTP effluents. In this study, the efficiencies of biological degradation in a MBR system was combined with UF unit, which separated the treated wastewater from the mixed liquor (MBR/UF), and in an optional NF unit advanced ion removal were assessed (MBR/UF/NF). The treatment efficacies of MBR/UF and MBR/UF/NF were then compared with the requirements imposed on wastewater discharged into municipal wastewater systems and surface water receivers in Poland (Table 2).

3.1. Characteristics of the MBR system influent

During the 1st phase of the experiment, to obtain the activated sludge acclimation, the mixture of MP-LL and PP-LL was initially diluted with water (50% v:v) (Fig. 1). Then, for 14 days, a stepwise increasing acclimatization process was performed (up to 100%) for the volumetric amount of the MP-LL/PP-LL mixture in the MBR system influent. However, even the half-diluted mixture of MP-LL and PP-LL was characterized by physical and chemical parameters that were much higher than what is typically found in municipal wastewater (Table 2). Additionally, the high variability of the influent introduced into the MBR system was observed during the 2nd phase (treatment phase) of the experiment. The COD varied significantly from 3948 mg O_2/L to 6509 mg O_2/L. The average COD-to-BOD_5 ratio, 0.52 ± 0.11, indirectly indicated the susceptibility of organic pollutants to biological decomposition. In addition, the average TN-to-BOD_5 ratio was 0.56 ± 0.19, suggesting that a certain proportion of readily biodegradable organic matter was present for efficient denitrification. However, the TN-to-BOD_5 ratio variability (from 0.35 to 0.97) indicated that the denitrification process can occasionally be impaired, resulting in higher nitrate values in the outflow. Maintaining the high efficiency of the denitrification process was essential in this study because of the high concentration of nitrogen (from 1151 mg N/L to 1458 mg N/L) in the inflow of the MBR system (Table 2). Note that TN consisted mainly of ammonia nitrogen (>85%), which is typically reported for LLs. Additionally, the raw mixtures of MP-LL and PP-LL were found to contain relatively high chloride concentrations (ranging from 1780 to 2180 mg Cl^-/L). Although chloride is commonly found in LL, such a high concentration of chloride at the studied landfill site is likely a result of the municipal landfill plant operation system (after

Table 1
Details of the GC/MS analyses.

Analyte[a]	Quantifier [m/z]	Qualifier [m/z]	Retention time [min]	Limit of quantification (LOQ) [µg/L]	Limit of detection (LOD) [µg/L]
DMP	163	77	5.636	7.7	2.3
DEP	149	177	6.101	20.3	6.1
DnBP	149	76	7.161	53.6	16.1
BBzP	149	91	8.290	1.0	0.3
DEHP	149	167	8.616	149.5	44.8
DnOP	149	279	9.258	4.0	1.2
BPA	213	228	7.797	124.9	37.3

[a] DMP – dimethyl phthalate, DEP – diethyl phthalate, DnBP – di-n-butyl phthalate, BBzP – benzyl butyl phthalate, DEHP – bis(2-ethylhexyl) phthalate, DnOP – di-n-octyl phthalate; BPA – bisphenol A.

Table 2
Efficiency of landfill leachate treatment by the MBR/UF and MBR/UF/NF systems.

Influent	MBR influent	MBR/UF effluent		MBR/UF/NF effluent		MBR influent	MBR/UF effluent		MBR/UF/NF effluent		Allowable limits for the wastewater discharge into	
	$\frac{mean/\sigma}{min \div max}$	$\frac{mean/\sigma}{min \div max}$	Treatment effectiveness mean [%]/σ	$\frac{mean/\sigma}{min \div max}$	Treatment effectiveness mean [%]/σ	$\frac{mean/\sigma}{min \div max}$	$\frac{mean/\sigma}{min \div max}$	Treatment effectiveness mean [%]/σ	$\frac{mean/\sigma}{min \div max}$	Treatment effectiveness mean [%]/σ		
	1st phase – biomass acclimation					2nd phase – treatment phase					Wastewater system	Surface water[a]
pH	7.17/0.08 7.12 ÷ 7.29	7.47/0.23 7.20 ÷ 7.69	–	7.22/0.16 7.12 ÷ 7.45	–	7.08/0.25 6.43 ÷ 7.45	6.88/0.48 5.86 ÷ 7.54	–	7.05/0.24 6.51 ÷ 7.45	–	6.5 ÷ 12.5	6.5 ÷ 9
Conductivity [mS/cm]	13.98/4.19 8.74 ÷ 18.11	10.06/2.15 7.24 ÷ 12.02	26/7	1.07/0.14 0.87 ÷ 1.20	92/3	18.80/2.37 11.13 ÷ 21.70	11.88/2.32 8.05 ÷ 14.84	36/14	1.12/0.25 0.79 ÷ 1.60	94/2.00	[b]	–
TSS [mg/L]	154/81 70 ÷ 250	13/6.62 6 ÷ 22	90/6.37	1.02/0.21 0.75 ÷ 1.22	99/0.56	287/233 64 ÷ 890	10/7.44 2 ÷ 27	95/5.25	1.11/0.25 0.74 ÷ 1.60	99/0.57	[b]	35
Cl^- [mg/L]	1213/470 712 ÷ 1794	1034/370 619 ÷ 1443	14/34	184/44 143 ÷ 237	82/10.79	1946/127 1780 ÷ 2180	1565/245 1140 ÷ 1960	19/12	63/26 37 ÷ 136	97/1.27	1000	1000
TN [mg/L]	906/305 529 ÷ 1210	384/75 287 ÷ 457	55/78	66/8 57 ÷ 75	92/3.72	1320/79 1151 ÷ 1458	157/63 80 ÷ 257	88/5	13/8 6 ÷ 32	99/0.69	[b]	10
$N\text{-}NH_4$ [mg/L]	790/247 471 ÷ 1011	99/32 55 ÷ 125	85/9	2.48/1.89 0.98 ÷ 5.12	99.58/0.46	1131/161 631 ÷ 1371	5/10 1 ÷ 34	99.59/0.83	0.26/0.13 0.20 ÷ 0.54	99.98/0.01	100 ÷ 200	–
TP [mg/L]	30.83/8.02 21.80 ÷ 41.30	16.35/3.54 12.40 ÷ 20.80	46/12	1.67/0.46 0.98 ÷ 1.98	96.08/1.61	47.65/22.78 15.00 ÷ 108.00	6.64/5.68 1.01 ÷ 22.10	87/9	1.21/0.25 0.59 ÷ 1.59	99.64/0.40	[b]	1
$P\text{-}PO_4$ [mg/L]	27.08/8.02 19.50 ÷ 38.40	13.96/88/83.81 8.63 ÷ 17.20	47/17	1.16/0.44 0.84 ÷ 1.80	93.47/2.70	41.74/20.20 11.20 ÷ 94.44	5.29/4.95 0.90 ÷ 19.80	88/8	0.15/0.18 0.03 ÷ 0.59	96.51/1.48	[b]	–
BOD_5 [mg/L]	1500/452 1023 ÷ 2014	109/43 56 ÷ 158	91/6	4.75/1.71 3.00 ÷ 7.00	99.64/0.20	2585/736 1350 ÷ 3540	13/14 3 ÷ 45	99.53/0.47	2.21/0.77 1.00 ÷ 3.10	99.91/0.03	[b]	15
COD [mg/L]	2791/997 1745 ÷ 3947	772/97 677 ÷ 896	68/15	33.50/3.70 30.00 ÷ 38.00	98.64/0.60	4903/742 3948 ÷ 6509	740/98 580 ÷ 954	84/4	22.67/3.39 18.00 ÷ 28.00	99.53/0.07	[b]	125
BPA [μg/L]	282//241 64 ÷ 796	110.2/37.7 66.6 ÷ 150.6	74/18	<LOD	100/0	606/702 125 ÷ 2416	<LOD	100/0	<LOD	100/0	–	–
PAE [μg/L]	<LOD ÷ 125	<LOD ÷ 21	–	<LOD	–	<LOD ÷ 298	<LOD	–	<LOD	–	–	–
DMP [μg/L]	<LOD ÷ 27.9	<LOD	–	<LOD	–	<LOD ÷ 23.1	<LOD	–	<LOD	–	–	–
DEP [μg/L]	<LOD	<LOD	–	<LOD	–	<LOD ÷ 50.3	<LOD	–	<LOD	–	–	–
DnBP [μg/L]	<LOD	<LOD	–	<LOD	–	<LOD	<LOD	–	<LOD	–	–	–
BBzP [μg/L]	<LOD ÷ 1.7	<LOD	–	<LOD	–	<LOD ÷ 1.2	<LOD	–	<LOD	–	–	–
DEHP [μg/L]	<LOD ÷ 125	<LOD ÷ 21	–	<LOD	–	<LOD ÷ 298	<LOD	–	<LOD	–	–	–
DnOP [μg/L]	<LOD	<LOD	–	<LOD	–	<LOD	<LOD	–	<LOD	–	–	–

[a] Requirements of Urban Waste Water Treatment Directive for discharge from WWTP with more than 100,000 PE.
[b] Value determined by the wastewater system owner.

reverse osmosis, the concentrate is returned to the landfill waste cell).

In the case of PAEs and BPA, their concentrations in the MP-LL/PP-LL mixture entering the MBR system varied (Table 2). BPA was in a range from 125 to 2416 μg/L, whereas the total amount of PAEs ranged from <LOD to 298 μg/L. The environmental fate of PAEs and BPA has received increasing attention because of their potential adverse health effects on animals and humans. In the case of tested LLs, the high values of total PAEs in the MP-LL and PP-LL mixtures entering the MBR system were caused mainly by elevated concentrations of DEHP (up to 298 μg/L), which were identified by EU legislation as hazardous among 45 priority substances in the field of water policy (EU, 2013) and are considered to be carcinogenic to humans (Group 2B) (IARC, 1987).

3.2. Effectiveness of treatment in the MBR/UF

The removal rate of tested pollutants varied significantly during the activated sludge acclimation phase (1st phase of the experiment) but was generally increasing to >90% for TSS, BOD_5, and N-NH_4 (Fig. 2). A lower removal rate of approximately 60% was obtained for phosphorus and nitrogen. In the case of TN, the lower rate was a result of limited nitrification and denitrification efficiency as well as the presence of N-NO_3, N-NH_4, and to a lesser extent, N-NO_2 in the MBR/UF effluent. At the end of biomass acclimation (1st phase), the total nitrogen concentration in the MBR/UF effluent was equal to 367 mg N/L, and its major mineral forms ammonia and nitrate reached 284 mg N-NO_3/L and 55 mg N-NH_4/L, respectively. A higher removal rate was obtained for COD, and at the end of the 1st phase of the experiment, it equalled 82.8% (Fig. 2). Interestingly, COD removal did not greatly increase in the 2nd phase (treatment phase) of the experiment and varied from 77.2% to 90% (Fig. 2). In the MBR/UF effluent the average COD value was equal to 740 ± 98 mg O_2/L, exceeding the value permissible for discharge into the surface water receiver (Table 2) although acceptable in the municipal wastewater system. In the case of nitrogen, however, in the 2nd phase, its removal rate increased to 94.2% (Fig. 2). The TN concentration was still high in the MBR/UF effluent, equalling 80 mg N/L at the end of the experiment, and 81% consisted of N-NO_3. Interestingly, a similar TN removal rate (>90%) was obtained for TP and TSS, which resulted in reduction of phosphorus below 1.5 mg P/L and TSS below 15 mg/L. These parameters indicated that MBR/UF effectively treated MP-LL and PP-LL mixtures. The quality of the MBR/UF effluent was generally similar to that of raw municipal wastewater; therefore, the effluent could be discharged into the municipal wastewater system. The objections, however, are connected with the suspected low removal of chloride (19%): its concentration in MBR/UF effluent reached 1960 mg Cl^-/L, which was two times higher than the concentration acceptable by Polish regulation for industrial wastewater entering the municipal wastewater system (Table 2). The above regulations for Cl^- concentrations in wastewater are provided to limit chloride-related corrosion in wastewater collecting and treating systems. The efficiency of micropollutant removal in the MBR system is also of concern. In this study, PAEs and BPA were tested because of their

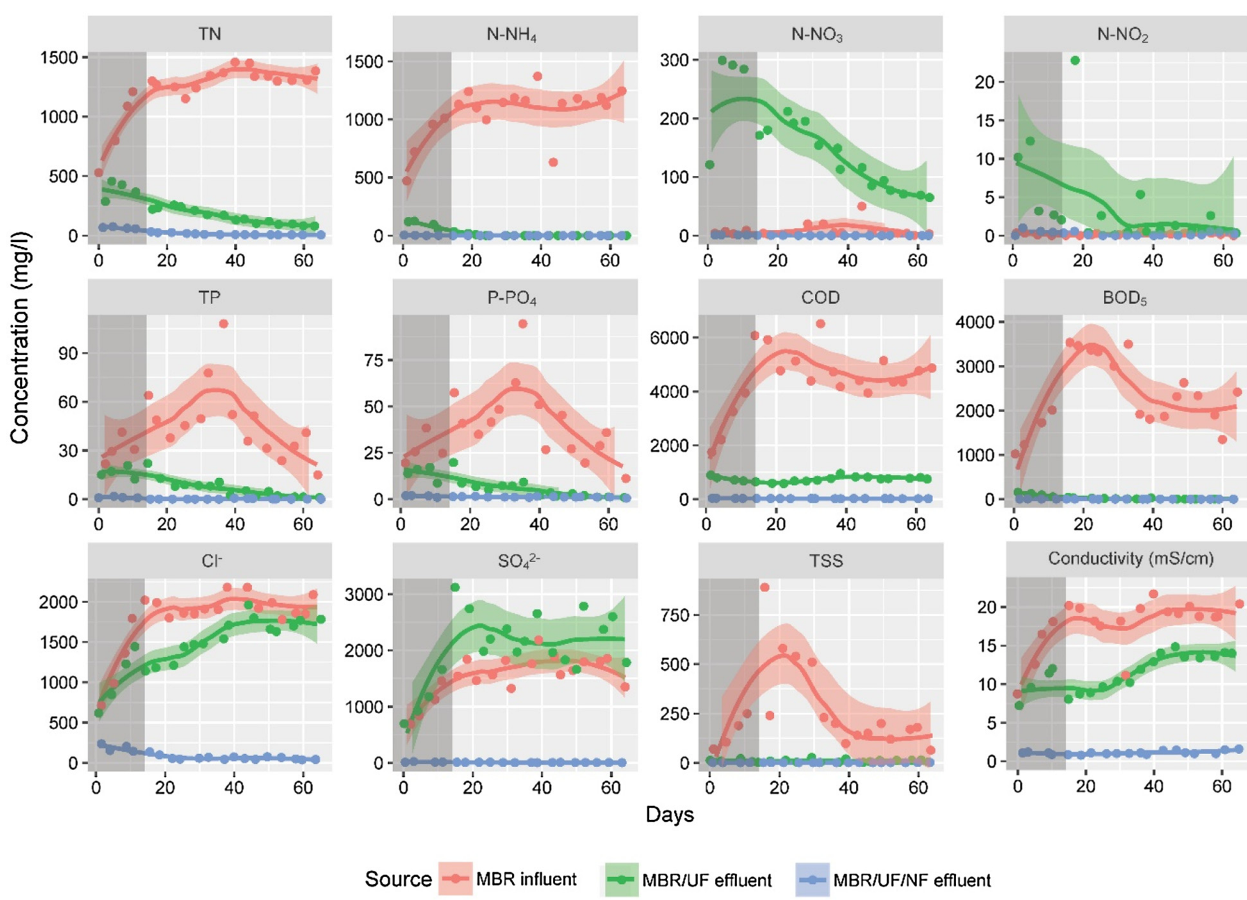

Fig. 2. Temporal evolution of several chemical/physical parameters measured in MBR influent, MBR/UF effluent and MBR/UF/NF effluent. The scores are of the real samples, the line is the LOESS fit, and the coloured shaded area outlines the 95% confidence interval. The grey shaded area represents the activated sludge acclimation phase (1st phase of the experiment).

suspected (Fudala-Ksiazek et al., 2017) high concentration in the MP-LL/PP-LL mixture entering the MBR system, and the results were confirmed by data obtained for the MBR influent (see Table 2).

The MBR/UF effectively removed PAEs, and throughout the experiment, none of the tested PAEs were detected in the MBR/UF effluent. In the case of BPA, its concentration continuously decreased during the 1st phase of the experiment and generally remained below the quantification limit in the 2nd phase. The high effectiveness of the MBR system in removing BPA (over 94%) was also confirmed by Chen et al. (2008).

Because PAEs and BPA are generally hydrophobic organic compounds, both adsorption on activated sludge flocs and biodegradation are believed to be the principal means for their removal, whereas volatilization and abiotic hydrolysis should be negligible, as suggested by other researchers. Thus, the contribution of adsorption was tested in this study using the activated sludge liquor taken from the aeration tank of the MBR system and by estimating the concentrations of BPA and PAEs in the entire sample and in the aqueous phase of that sample (supernatant after centrifugation and filtration). According to the obtained results, none of the tested PAEs were detected in the activated sludge liquor or its supernatant. BPA was detected only during the 1st phase of the experiment (acclimation of activated sludge) on the first day when the BPA concentrations in the entire sample and in the aqueous phase of that sample (supernatant) were 2057.2 μg/L and 147.7 μg/L, respectively, and on day 14 of the experiment when similar values of 2057.2 μg/ml and 102.1 μg/L, respectively, were obtained. Then, during the 2nd phase of the experiment, the BPA concentration decreased with time in both the entire sample of the activated sludge and in its supernatant. The BPA was noted only at day 21 at concentrations of 150.6 μg/L and 74.0 μg/L respectively, and it was subsequently detected under the limit of quantification (<50 μg/L). The obtained data indicated that PAEs were degraded with high effectiveness, whereas in the case of BPA, especially during activated sludge acclimation (1st phase of the experiment), sorption exceeded biodegradation. Thus, the gradual decrease in the BPA concentration in the activated sludge liquor suggests that flock adsorption might play an important role in BPA removal during activated sludge acclimation and that activated sludge microorganisms might require some time to adapt and effectively biodegrade BPA. The results of this study support the data obtained by Chen et al. (2008), which also show the limited contributions of sludge adsorption to BPA removal, with biodegradation as a dominating factor. In addition, Clara et al. (2005) achieved high BPA removal rates of more than 90% for both conventional activated sludge WWTP and MBR/UF, suggesting that the operated ultrafiltration membrane does not lead to any further retention of BPA because of size exclusion.

3.3. Biomass acclimation and its potential capability of PAEs and BPA biodegradation/biotransformation

The presence of PAEs and BPA has already been confirmed in LLs by several studies (Jonsson et al., 2003; Kalmykova et al., 2014; Fudala-Ksiazek et al., 2017) because these compounds meet the needs of a wide variety of markets and therefore end up in the waste stream. In this study, the capability of biomass to degrade/biotransform PAEs and BPA was verified using a metagenomic approach. Activated sludge is composed of a highly complex microbial community that is shaped by both wastewater quality and technological processes. Activated sludges have been suggested to exhibit unique bacterial communities (Ibarbalz et al., 2013). On the other hand, their metabolism is a key factor determining the effectiveness of wastewater treatment in terms of nutrient and organic matter removal. Thus, recognizing the bacterial structure that developed as a result of the activated sludge acclimation processes was crucial in this study.

For this reason, the metagenomic approach was employed to identify the taxonomic diversity of INC-AS activated sludge, which was used to seed the MBR system and ACL-AS and taken at the end of experiment. As is usually observed, the ratio of the unclassified sequence increased from the domain to the genus level. For the INC-AS samples, these levels were equal to 1.27% and 12.20%, and for the ACL-AS samples, they were equal to 5.97% and 20.88% of the reads (results are given as a portion of total effective bacterial sequences, see Methods in Section 2). These data also indicated that unclassified sequence portions in the total community increased during the acclimation of activated sludge to the co-treated mixture of MP-LL and PP-LL. The comparative analysis of metagenomic results showed, however, that the core microbiota in both INC-AS and ACL-AS consisted of eight main phyla (>1% of reads) accounting for 94.47% and 86.98% of the reads, respectively (Fig. 3). Among them, *Proteobacteria* was the most abundant phylum, accounting for 55.73% of the reads in INC-AS and 65.04% of the reads in ACL-AS. These observations are in agreement with the results obtained by Zhang et al. (2012) for WWTPs based on activated sludge processes and for other environments affected by municipal wastewater and as-treated wastewater receivers (McLellan et al., 2010). Other domain phyla detected in the INC-AS sample were *Bacteroidetes* (23.51%), followed by *Firmicutes* (5.53%), *Actinobacteria* (3.29%), *Thermi* (2.09%), *Chlorobi* (1.69%), *Chloroflexi* (3.4%), and *Crenarchaeota* (1.02%). A similar pattern was also obtained for acclimated sludge; however, the percentage share differed markedly because in the ACL-AS sample (Fig. 3). Bacterial phyla such as *Proteobacteria*, *Bacteroidetes*, *Firmicutes*, and *Actinobacteria* have also been noted in municipal and industrial activated sludge, although the proportions vary (Ibarbalz et al., 2013) and have been confirmed to be present in LLs generated by landfill waste cells (Fudala-Ksiazek et al., 2016). The structure of activated sludge consortia depends on the incoming substrate, technological parameters and activated sludge age (Shchegolkova et al., 2016). In this study, the physical, chemical and microbiological quality of the treated mixtures of MP-LL and PP-LL caused the significantly lower abundance of the phylum *Bacteroidetes* in the ACL-AS sample than in the INC-AS sample and the shift within the *Proteobacteria* phylum, which was dominated by *Betaproteobactera* (38.98%) in INC-AS. In the ACL-AS sample, the phyla were dominated by *Alphaproteobacteria* (42.37%). The *Alphaproteobacteria* in the ACL-AS sample, order *Rhizobiales,* were primarily represented by the *Hyphomicrobiaceae* family (34.80%) and the following four genera: *Rhodoplanes* (25.21%), *Hyphomicrobium* (5.95%), *Methylosinus* (2.50%) and *Pedomicrobium* (1.13%).

The predominance of *Alphaproteobacteria* has been suggested to be attributed to the salinity of wastewater (Zhang et al., 2012)), what was confirmed in this study (average conductivity and Cl^- concentration in MBR effluent were equal to 18.80 mS/cm and to 1946 mg CL^-/L, respectively), whereas long SRT additionally promotes *Rhizobiales*-affiliated OTUs (Ju and Zhang, 2014).

Because treated mixtures of MP-LL and PP-LL contained high ammonia concentrations (from 631 to 1371 mg N-NH_4/L, see Table 1) and the MBR performed with high removal efficiency, significant representation of ammonia and nitrite oxidizers was expected. However, as previously reported by Ju and Zhang (2014), Zhang et al. (2012), and Saunders et al. (2015) and as reported in this study, these organisms were poorly represented.

The results indicated that among ammonia oxidizers, ammonia-oxidizing archaea (AOA) were represented by candidatus *Nitrososphaera* (1.014% and 3.233%, respectively) and *Nitrosopumilus* (<0.001% and 0.0013%, respectively) in the tested INC-AS and ACL-AS samples, and ammonia-oxidizing bacteria (AOB) were represented by *Nitrosovibrio* (0.012% and 0.024%, respectively) and

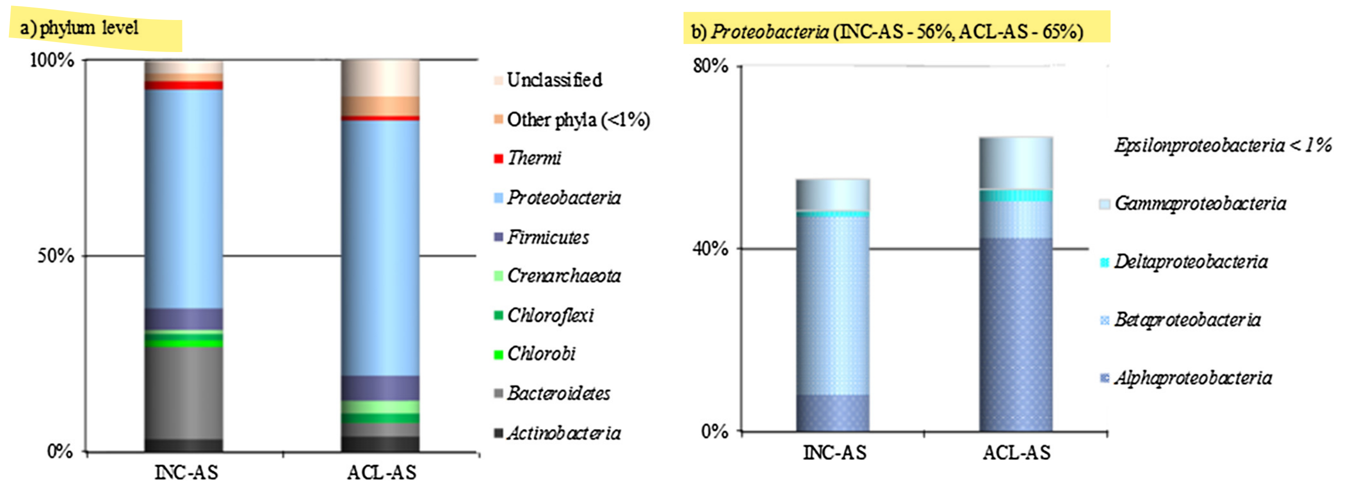

Fig. 3. Bacterial community compositions in activated sludge at the phylum level (a) and class diversity among *Proteobacteria* (b); INC-AS – activated sludge used for MBR inoculation; ACL-AS – acclimated activated sludge.

Nitosospira (0.008 and 0.001%, respectively) from the *Betaproteobacteria* family as well as by *Nitrosocossus* (0.138% and 0.324%, respectively) from the *Gammaproteobacteria* family. According to the obtained data, the AOA selected in this study are generally regarded as less abundant in WWTPs than AOB (Jin et al., 2010). The parameters that influence the occurrence and distribution of AOA are not fully recognized, although Park et al. (2006) reported AOA in WWTPs operating with low dissolved oxygen (DO) levels and long retention times, whereas Tourna et al. (2008) indicated that these species show tolerance to relatively high ammonia concentrations.

Nitrite oxidizing bacteria (NOB) were even less abundant in both INC-AS and ACL-AS samples and were represented only by *Nitrobacter* (0.003% and 0.007%, respectively) and *Nitrospira* (0.218% and 0.001%, respectively), which can convert ammonia directly to nitrate (comammox process), as shown by Daims et al. (2015). However, unidentified and/or unassigned AOB/NOB were possibly present in the INC-AS and ACL-AS samples. Interestingly, in this study anaerobic ammonium oxidation (anammox) process can also be involved in the nitrogen cycle because bacteria belonging to the *Candidatus Scalindua* genus, were noted in both INC-AS and ACL-AS samples. *Candidatus Scalindua* was represented mainly by *Candidatus Scalindua brodae*, which was detected at a comparable level during the experiment (0.15% in INC-AS and 0.13% in ACL-AS), whereas *Candidatus Scalindua wagneri* was found only in the ACL-SA samples and did not exceed 0.01%. Moreover, both species were originally detected by Schmid et al. (2003) in a wastewater treatment plant treating LL in Pitsea, United Kingdom.

In case of denitrification, such capability has been identified among a variety of organisms (Kumar and Lin, 2010). In this study, INC-AS denitrifers were predominantly represented by *Thauera* (24.36%) affiliated to *Betaproteobacteria*, whereas during activated sludge acclimation, *Alfaproteobacteria* affiliated *Rhodobacter* (25.21%) and *Hyphomicrobium* (5.95%) were selected and may greatly contribute to the nitrogen removal in the tested MBR system. Other potentially denitrifying bacteria were also present in the INC-AS and ACL-AS samples (as listed in Table S2), although they were represented by less than 1% of the total effective bacterial sequences.

In addition to nitrate (nitrate-based denitrification), sulphate can also be used as an electron acceptor (sulphate-based denitrification), especially in sulphate-rich media, such as LLs (see Table 2). Sulphate-reducing bacterial communities contain obligate anaerobes and members of a heterogeneous group of eubacteria and archaebacteria, which are able to carry out dissimilatory sulphate reduction and have been reported to play an important role in the biodegradation of drivers of micropollutants. In this study, these bacteria represented 0.46% and 0.85% of the total effective bacterial sequences in the INC-AS and ACL-AS samples, respectively and were affiliated mainly to *Desulfovibrio* and *Desulfuromusa* (*Deltaproteobacteria*) as well as *Desulfurispora* (*Peptococcaceae* family of *Clostridiales* order). Interestingly, sulphur-oxidizing chemolithotrophs were also detected in both the INC-AS and ACL-AS samples and belonged mainly to the genera: *Thiocapsa* (0.34% and 0.55%, respectively) and *Ectothiorhodospira* (0.23% and 0.17%, respectively), which belong to *Gammaroteobacteria* and were followed by genus *Thiomonas* (0.32% and 0.07%, respectively), which belong to *Betaproteobacteria*.

The possibility of PAEs and BPA removal in MBR systems was studied, and the metagenomics results were analysed in terms of the presence of bacteria capable of degrading the tested endocrine disruptors. To obtain complete degradation of those compounds, usually a complex bacterial community is involved, although this study reports bacteria that can degrade PAEs and BPA in pure culture. *Clostridium* (1.00%) was detected with the highest abundance, followed by *Mycobacterium* (0.41%) and *Rhodococcus* (0.31%) from *Actinobacteria* and *Alphaproteobacteria*-affiliated *Sphingomonas* sp (0.27%) and other bacteria from *Sphingomonadaceae*. A potential for PAEs and BPA metabolism has been already confirmed for several *Rhodococcus* species (Nalli et al., 2002; Roslev et al., 2007).

3.4. Effectiveness of treatment in the MBR/UF/NF

The size of the nanomembrane pores (typically 1–10 nm) created an effective barrier against contaminants with molecular weights of 200–400 Da; however, for inorganic ions and the rejection of lower-molecular-weight organic compounds, diffusion mechanisms and the charge effect of the membrane are considered. Nonetheless, the quality of the effluent obtained from the MBR/UF/NF module is believed to be more stable and influenced less by activated sludge flocculation characteristics, hydraulic shear in the reactor, and activated sludge acclimation or possible process breakdown. To mitigate NF membrane fouling, the inflow should be pre-treated with a previous UF, such as in this study.

As expected, the MBR/UF/NF removal rates for most of the tested compounds were >99%, with the occasional exception of TP and TN, especially during the 1st phase of biomass acclimation. However, the TP and TN concentrations in the MBR/UF/NF effluent in the 2nd phase were below 1 mg P/l and 10 mg N/l, respectively, and did not exceed the limits even for treated wastewater discharge into the receiver (Table 2, Fig. 2). The MBR/UF/NF system was, however, less effective for removing chloride (average 82% and 97% removal rates in the 1st and 2nd phases, respectively) but provided an average concentration of Cl^- in the effluent not exceeding 250 mg Cl^-/l.

Note that chlorides are commonly present in LLs, although their elevated concentration (from 1780 to 2180 mg Cl^-/L) in the MBR influent (the mixture of MP-LL and PP-LL) obtained in this study (see Section 3.2 for details) influenced the final treatment effectiveness.

In general, the efficiency of the MBR/UF/NF system was stable, and the obtained effluent satisfied several criteria. However, because such advanced technology is always associated with investment and operating costs, using the MBR system with a combined UF and NF module must be justified by health-related, environmental or other benefits. According to the obtained data, the NF module can be used during the biomass acclimation phase when MBR/UF effectiveness is limited, especially when the nitrogen concentration in the effluent does not meet the regulations for industrial wastewater entering the sewage system. Another MBR/UF/NF advantage is high efficiency in chloride and micropollutant removal. Cl^- should be limited because of its corrosive activity, whereas micropollutants should be limited to avoid their further adverse impacts.

The overall operating cost of LL treatment was calculated using the tested MBR/UF/NF unit as 22.5 PLN/m^3 (5.4 €/m^3), without staff costs. This cost is less expensive than the reverse osmosis system (approximately 29 PLN/m^3; 6.9 €/m^3), which is most commonly used at other MSWPs in Poland. The reverse osmosis system, despite having high operating expenses, also generates many operational problems, such as condensate management (as previously described). Additionally, the need to simultaneously treat leachates originating from waste cells of different ages is challenging, especially because the leachate generated by young waste cells may accelerate membrane fouling. In the case of the simultaneous treatment of leachates generated by modern and previous waste cells, the MBR system combined with a UF unit is the most reasonable configuration from technological and economical points of view (estimated operating costs were calculated to be 14 PLN/m^3; 3.3 €/m^3). Note that with time, a decreasing amount of biodegradable organic matter in MBR influent is expected because of changes in the biochemistry of modern waste cells. Thus, an external carbon source would probably be required to support denitrification. To avoid additional costs connected with supplementing the MBR system with a conventional carbon source (e.g., methanol or ethanol), other technological solutions are being tested by the authors, such as using wastewater generated by other MSWP units (sorting and/or composting unit) because of its high biodegradability (Fudala-Ksiazek et al., 2017, 2016).

4. Conclusions

In Poland, reverse osmosis is the most commonly applied technology to treat LLs; however, it represents a separation method rather than a treatment method, is expensive and generates several operational problems. Thus, more suitable technologies are required that consider the physical and chemical parameters of LLs, their time variation and legal discharge requirements, and their overall costs. In this study, a biological treatment combined with a membrane-based technology was tested to simultaneously treat LLs generated by modern and previous waste cells. This mixture was characterized by a high initial COD concentration (from 3948 to 6509 mg O_2/L) and high ammonia concentration (from 631 to 1371 mg N-NH_4/L) as the main component of TN (from 1152 to 1458 mg N/L). According to the obtained results, the effluent obtained from the MBR/UF/NF unit met not only the discharge requirements for wastewater entering the sewage system but also the requirements for treated wastewater discharging into the receiver. Due to its high costs, however, such an advanced treatment cannot always be justified, especially since less expensive MBR/UF units, after biomass acclimation, provide effluent with a quality acceptable for discharge into wastewater systems, except with respect to chloride levels, which concentrations occasionally exceed 1000 mg Cl^-/L. This research also provided a perspective on the application of a membrane bioreactor combined with UF to remove micropollutants, such as the commonly present PAEs and BPA, from LLs. The co-existence of aerobic and anaerobic (anammox) ammonia-oxidizing organisms together with denitrifying bacteria during LL treatment provides the opportunity for the simultaneous removal of nitrogen and organic carbon. However, further research is needed on the optimization of those biochemical routes.

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References

American Public Health Association, American Water Works Association, Water Environment Federation, 2005. Standard Methods for the Examination of Water and Wastewater. APHA-AWWA-WEF, Washington, D.C..

Besha, A.T., Gebreyohannes, A.Y., Tufa, R.A., Bekele, D.N., Curcio, E., Giorno, L., 2017. Removal of emerging micropollutants by activated sludge process and membrane bioreactors and the effects of micropollutants on membrane fouling: a review. J. Environ. Chem. Eng. 5, 2395–2414. https://doi.org/10.1016/j.jece.2017.04.027.

den Boer, E., Jędrczak, A., Kowalski, Z., Kulczycka, J., Szpadt, R., 2010. A review of municipal solid waste composition and quantities in Poland. Waste Manage. 30, 369–377. https://doi.org/10.1016/j.wasman.2009.09.018.

Boonnorat, J., Techkarnjanaruk, S., Honda, R., Prachanurak, P., 2016. Effects of hydraulic retention time and carbon to nitrogen ratio on micro-pollutant biodegradation in membrane bioreactor for leachate treatment. Bioresour. Technol. 219, 53–63. https://doi.org/10.1016/j.biortech.2016.07.094.

Chen, J., Huang, X., Lee, D., 2008. Bisphenol A removal by a membrane bioreactor. Process Biochem. 43, 451–456. https://doi.org/10.1016/j.procbio.2008.01.001.

Clara, M., Strenn, B., Gans, O., Martinez, E., Kreuzinger, N., Kroiss, H., 2005. Removal of selected pharmaceuticals, fragrances and endocrine disrupting compounds in a membrane bioreactor and conventional wastewater treatment plants. Water Res. 39, 4797–4807. https://doi.org/10.1016/j.watres.2005.09.015.

Daims, H., Lebedeva, E.V., Pjevac, P., Han, P., Herbold, C., Albertsen, M., Jehmlich, N., Palatinszky, M., Vierheilig, J., Bulaev, A., Kirkegaard, R.H., von Bergen, M., Rattei, T., Bendinger, B., Nielsen, P.H., Wagner, M., 2015. Complete nitrification by *Nitrospira* bacteria. Nature 528, 504. https://doi.org/10.1038/nature16461.

EU, 1991, n.d. Council Directive 91/271/EEC of 21 May 1991 concerning urban waste-water treatment.

EU, 1999., n.d. Council Directive 19999/31/EC of 26 April 1999 on the landfill of waste.

EU, 2008., n.d. Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives.

EU, 2013, n.d. Directive 2013/39/EU of the European Parliament and of the Council of 12 Agust 2013 amending Directives 200/60/EC and 2008/105/EC as a priority substances in the field of water policy.

Fudala-Ksiazek, S., Pierpaoli, M., Kulbat, E., Luczkiewicz, A., 2016. A modern solid waste management strategy – the generation of new by-products. Waste Manage. 49, 516–529. https://doi.org/10.1016/j.wasman.2016.01.022.

Fudala-Ksiazek, S., Pierpaoli, M., Luczkiewicz, A., 2017. Fate and significance of phthalates and bisphenol A in liquid by-products generated during municipal solid waste mechanical-biological pre-treatment and disposal. Waste Manage. 64, 28–38. https://doi.org/10.1016/j.wasman.2017.03.040.

Fudala-Ksiazek, S., Sobaszek, M., Luczkiewicz, A., Pieczynska, A., Ofiarska, A., Fiszka-Borzyszkowska, A., Sawczak, M., Ficek, M., Bogdanowicz, R., Siedlecka, E.M., 2018. Influence of the boron doping level on the electrochemical oxidation of

raw landfill leachates: advanced pre-treatment prior to the biological nitrogen removal. Chem. Eng. J. 334, 1074–1084. https://doi.org/10.1016/j.cej.2017.09.196.
Grandclément, C., Seyssiecq, I., Piram, A., Wong-Wah-Chung, P., Vanot, G., Tiliacos, N., Roche, N., Doumenq, P., 2017. From the conventional biological wastewater treatment to hybrid processes, the evaluation of organic micropollutant removal: a review. Water Res. 111, 297–317. https://doi.org/10.1016/j.watres.2017.01.005.
IARC, 1987, n.d. Overall evaluations of carcinogenenicity: an updating of IARC monographs (No. 1 to 42). IARC Monographs on the Evaluation of the Carcinogenic Risksto Humans. World Health Organization, Lyon, France.
Ibarbalz, F.M., Figuerola, E.L.M., Erijman, L., 2013. Industrial activated sludge exhibit unique bacterial community composition at high taxonomic ranks. Water Res. 47, 3854–3864. https://doi.org/10.1016/j.watres.2013.04.010.
Jin, T., Zhang, T., Yan, Q., 2010. Characterization and quantification of ammonia-oxidizing archaea (AOA) and bacteria (AOB) in a nitrogen-removing reactor using T-RFLP and qPCR. Appl. Microbiol. Biotechnol. 87, 1167–1176. https://doi.org/10.1007/s00253-010-2595-2.
Jonsson, S., Ejlertsson, J., Svensson, B.H., 2003. Transformation of phthalates in young landfill cells. Waste Manage. 23, 641–651. https://doi.org/10.1016/S0956-053X(03)00099-0.
Ju, F., Zhang, T., 2014. Bacterial assembly and temporal dynamics in activated sludge of a full-scale municipal wastewater treatment plant. ISME J. 9, ismej2014162. http://doi.org/10.1038/ismej.2014.162.
Kalmykova, Y., Moona, N., Strömvall, A.-M., Björklund, K., 2014. Sorption and degradation of petroleum hydrocarbons, polycyclic aromatic hydrocarbons, alkylphenols, bisphenol A and phthalates in landfill leachate using sand, activated carbon and peat filters. Water Res. 56, 246–257. https://doi.org/10.1016/j.watres.2014.03.011.
Kulikowska, D., Klimiuk, E., 2008. The effect of landfill age on municipal leachate composition. Bioresour. Technol. 99, 5981–5985. https://doi.org/10.1016/j.biortech.2007.10.015.
Kumar, M., Lin, J.-G., 2010. Co-existence of anammox and denitrification for simultaneous nitrogen and carbon removal—strategies and issues. J. Hazard. Mater. 178, 1–9. https://doi.org/10.1016/j.jhazmat.2010.01.077.
Liu, J., Zhang, H., Zhang, P., Wu, Y., Gou, X., Song, Y., Tian, Z., Zeng, G., 2017. Two-stage anoxic/oxic combined membrane bioreactor system for landfill leachate treatment: pollutant removal performances and microbial community. Bioresour. Technol. 243, 738–746. https://doi.org/10.1016/j.biortech.2017.07.002.
Mandal, P., Dubey, B.K., Gupta, A.K., 2017. Review on landfill leachate treatment by electrochemical oxidation: drawbacks, challenges and future scope. Waste Manage. 69, 250–273. https://doi.org/10.1016/j.wasman.2017.08.034.
McLellan, S.L., Huse, S.M., Mueller-Spitz, S.R., Andreishcheva, E.N., Sogin, M.L., 2010. Diversity and population structure of sewage derived microorganisms in wastewater treatment plant influent. Environ. Microbiol. 12, 378–392. https://doi.org/10.1111/j.1462-2920.2009.02075.x.
Nalli, S., Cooper, D.G., Nicell, J.A., 2002. Biodegradation of plasticizers by *Rhodococcus rhodochrous*. Biodegradation 13, 343–352. https://doi.org/10.1023/A:1022313810852.
Park, H.-D., Wells, G.F., Bae, H., Criddle, C.S., Francis, C.A., 2006. Occurrence of ammonia-oxidizing archaea in wastewater treatment plant bioreactors. Appl. Environ. Microbiol. 72, 5643–5647. https://doi.org/10.1128/AEM.00402-06.
Development Core Team, R., 2016. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria.
Renou, S., Givaudan, J.G., Poulain, S., Dirassouyan, F., Moulin, P., 2008. Landfill leachate treatment: review and opportunity. J. Hazard. Mater. 150, 468–493. https://doi.org/10.1016/j.jhazmat.2007.09.077.
Roslev, P., Vorkamp, K., Aarup, J., Frederiksen, K., Nielsen, P.H., 2007. Degradation of phthalate esters in an activated sludge wastewater treatment plant. Water Res. 41, 969–976. https://doi.org/10.1016/j.watres.2006.11.049.
Saunders, A.M., Albertsen, M., Vollertsen, J., Nielsen, P.H., 2015. The activated sludge ecosystem contains a core community of abundant organisms. ISME J. 10, ismej2015117. http://doi.org/10.1038/ismej.2015.117.
Schmid, M., Walsh, K., Webb, R., Rijpstra, W.I., van de Pas-Schoonen, K., Verbruggen, M.J., Hill, T., Moffett, B., Fuerst, J., Schouten, S., Sinninghe Damsté, J.S., Harris, J., Shaw, P., Jetten, M., Strous, M., 2003. Candidatus "*Scalindua brodae*", sp. nov., Candidatus "*Scalindua wagneri*", sp. nov., two new species of anaerobic ammonium oxidizing bacteria. Syst. Appl. Microbiol. 26, 529–538. https://doi.org/10.1078/072320203770865837.
Shchegolkova, N.M., Krasnov, G.S., Belova, A.A., Dmitriev, A.A., Kharitonov, S.L., Klimina, K.M., Melnikova, N.V., Kudryavtseva, A.V., 2016. Microbial community structure of activated sludge in treatment plants with different wastewater compositions. Front. Microbiol. 7. https://doi.org/10.3389/fmicb.2016.00090.
Tourna, M., Freitag, T.E., Nicol, G.W., Prosser, J.I., 2008. Growth, activity and temperature responses of ammonia-oxidizing archaea and bacteria in soil microcosms. Environ. Microbiol. 10, 1357–1364. https://doi.org/10.1111/j.1462-2920.2007.01563.x.
Trulli, E., Ferronato, N., Torretta, V., Piscitelli, M., Masi, S., Mancini, I., 2018. Sustainable mechanical biological treatment of solid waste in urbanized areas with low recycling rates. Waste Manage 71, 556–564. https://doi.org/10.1016/j.wasman.2017.10.018.
Wickham, H., 2009. ggplot2 – Elegant Graphics for Data Analysis. Springer.
Wiszniowski, J., Robert, D., Surmacz-Gorska, J., Miksch, K., Weber, J.V., 2006. Landfill leachate treatment methods: a review. Environ. Chem. Lett. 4, 51–61. https://doi.org/10.1007/s10311-005-0016-z.
Wojciechowska, E., 2017. Potential and limits of landfill leachate treatment in a multi-stage subsurface flow constructed wetland – evaluation of organics and nitrogen removal. Bioresour. Technol. 236, 146–154. https://doi.org/10.1016/j.biortech.2017.03.185.
Zhang, T., Shao, M.-F., Ye, L., 2012. 454 pyrosequencing reveals bacterial diversity of activated sludge from 14 sewage treatment plants. ISME J. 6, 1137–1147. https://doi.org/10.1038/ismej.2011.188.