

Fate and significance of phthalates and bisphenol A in liquid by-products generated during municipal solid waste mechanical-biological pre-treatment and disposal

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abstract

Samples of liquid by-products generated by municipal solid waste plants (MSWPs) were tested for the presence of phthalates (PAEs) and bisphenol A (BPA). The results indicated that the wastewater generated during mechanical (sorting unit - SU) and biological (composting unit - CU) pre-treatment (MBT) of residual (mixed) solid waste is a significant source of these compounds. The concentrations of PAEs (up to 32222 µg/L) and BPA (up to 1795 µg/L) in the SU and CU wastewaters were generally higher than those in landfill leachates tested in this and other studies. To date, MBT wastewaters have been poorly studied and are usually overlooked. However, in this study, despite their relatively small quantities, they constituted an important load of ammonia (up to 1610 mg/L) and organic matter (COD up to 52980 mg/L). Thus, to apply an effective treatment, it is critical to monitor the current quality and quantity of all liquid by-products generated at MSWPs and to prioritize the (micro)pollutants of concern.

Keywords: Municipal solid waste, Treatment technology, Landfill leachates and technological, wastewater, Emerging pollutants, Micropollutants, Endocrine disruptors

1. Introduction

Over the last few decades, household consumption patterns have changed significantly, especially in terms of the increased usage of plastic, paper and cardboard from the food and beverage industries. The products addressed herein usually contain phthalates (PAEs) and bisphenol A (BPA), which are suspected to have negative impacts on humans due to their endocrine-disrupting effects (Schug et al., 2011). BPA is an organic synthetic compound (4,4'-(propane-2,2-diyl)diphenol) that has been commercially used since 1957 as a monomer in the production of polycarbonate plastic and epoxy resins, which represented approximately 75% and 25% of the BPA production volume in 2008, respectively, with the remainder being used as a component in polysulfone and polyacrylate resins and flame retardants (Bakker et al., 2014). PAEs (alkyl/aryl esters of 1,2-benzenedicarboxylic acids) are another family of chemicals used in plastic manufacturing with increasing global production since the 1930s. PAEs, especially those of low molecular weight (LMW), are also used as solvents and/or vehicles for fragrance in cosmetics, personal care products, pharmaceuticals,

waxes, inks, detergents, insecticides and other products (Hubinger and Havery, 2006). The basic characteristics of BPA and selected PAEs, as well as their presence in natural and artificial niches, are listed in Table 1.

Because PAEs and BPA meet the needs of a wide variety of markets, they undergo different types of treatment after they enter the waste stream according to local legislation. In the European Union (EU), the solid waste management sector has been subjected to substantial changes driven by the Landfill Directive (EU, 1999), the Waste Framework Directive (EU, 2008), the Packaging and Packaging Waste Directive (EU, 2015b), the Waste Electrical and Electronic Equipment Directive (EU, 2012) and other policies. For example, the Waste Framework Directive has provided a new definition for sustainable management of municipal solid waste (MSW), which, besides the emphasis on the 'zero waste strategy', has also introduced the novel term 'waste hierarchy'. To fulfill the above and other legislative requirements, incineration is employed in several countries, e.g., Germany and Austria (Holm and Simon, 2017; Huber et al., 2016), whereas mechanical-biological pre-treatment (MBT) of residual (mixed) solid waste is used in other countries, including Poland, as the main process of biomass recovery prior to landfilling. The intensive application of high-temperature thermal treatment technique of solid waste and potential risk connected with the release of organic pollutants

Table 1

Basic parameters and presence of selected PAEs and BPA in natural and artificial niches (Jonsson et al., 2003; Zhang and Wang, 2009; Boonyaroj et al., 2012; He et al., 2015; Kalmykova et al., 2013; Liu et al., 2009; Urase and Miyashita, 2003; Morin et al., 2015; Schwarzbauer et al., 2002; Net et al., 2014; Sibali et al., 2013; Jin and Zhu, 2016; Tran et al., 2015; Guerra et al., 2015; Clara et al., 2010).

Abbreviation	DMP	DEP	DnBP	BBzP	DEHP	DnOP	BPA
Characteristics of phthalates (PAEs) and bisphenol A (BPA)							
Common name	Dimethyl phthalate	Diethyl phthalate	Di- <i>n</i> -butyl phthalate	Benzyl butyl phthalate	Bis (2-ethylhexyl) phthalate	Di- <i>n</i> -octyl phthalate	Bisphenol A
CAS No.	131-11-3	84-66-2	84-74-2	85-68-7	117-81-7	117-84-0	80-05-7
Molecular formula	C ₁₀ H ₁₀ O ₄	C ₁₂ H ₁₄ O ₄	C ₁₆ H ₂₂ O ₄	C ₁₉ H ₂₀ O ₄	C ₂₄ H ₃₈ O ₄	C ₂₄ H ₃₈ O ₄	C ₁₅ H ₁₆ O ₂
Sample origin ^a and references	Literature data [µg/L]						
LP	Jonsson et al. (2003)	ND	ND-33	ND-29	ND-7	ND-460	-
LP	Zhang and Wang (2009)	ND-2.8	ND-5.66	0.35-59.75	ND-21.80	1.61-232.5	ND-521.10
LP	Boonyaroj et al. (2012)	20.80	12.50	35.40	21.50	65.50	8.00
LP	He et al. (2015)	1.83	1.48	0.25	-	-	0.47
LP	Kalmykova et al. (2013)	<LOD	<0.10-22	<0.10-1.8	<0.48-0.76	<1.0-23	<0.10-0.28
LP	Liu et al. (2009)	ND-43.27	ND	ND-15.11	ND	ND-7.24	ND
LP	Urase and Miyashita (2003)	-	-	-	-	-	26-8400
LP	Morin et al. (2015)	-	-	-	-	-	0.7-200
LP	Schwarzbauer et al. (2002)	-	-	-	-	-	4200-25000
SW	Net et al. (2014)	0.02-0.25	0.26-6.98	0.22-3.86	ND	5.16-20.80	ND
SW	Sibali et al. (2013)	0.04-0.56	0.08-0.39	0.79-3.65	-	0.49-5.58	-
SW	Jin and Zhu (2016)	-	-	-	-	-	ND-0.141
WW	Tran et al. (2015)	1.5 ± 1.3	6.0 ± 7.7	4.1 ± 1.6	4.0 ± 3.0	33.3 ± 15.4	0.7 ± 0.5
WW	Guerra et al. (2015)	-	-	-	-	-	0.42-3.831
WW	Clara et al. (2010)	ND-2.4	0.77-9.2	ND-8.7	0.31-3.2	3.4-34	ND-1.1

^a Literature: samples originated from LP - landfill prism leachates; from SW - surface water; from WW - wastewater; ND - not detected; LOD - limit of detection.

has been already reflected in several studies (Huber et al., 2016; Liu et al., 2016a, 2016b; Zhao et al., 2017), while the by-products generated by MBT processes are neglected. MBT usually consists of mechanical and/or manual sorting followed by the aerobic stabilization (composting) of the 0-80 mm fraction. In many cases, the effectiveness of MBT is questionable, especially due to poor municipal waste separation (or its absence) at the source (household) level. As a result, non- or barely compostable materials in the waste stream, such as plastic, glass and metals, enter the MBT composting units, resulting in several usually negative or questionable consequences. Among others, a poor product called stabilate is obtained and is usually used only for the restoration of landfill prisms (LPs). This issue is of special concern in countries such as Poland, where the recycling and sorting of solid waste at the household level is a rather new concept. Additionally, in the municipal waste sector, the fact is often overlooked that MBT generates by-products. To date, the primary focus has been on the leachate emissions of LPs. However, currently, liquid by-products are also generated by MBT facilities in their sorting units (SUs) and composting units (CUs). The quality and quantity of these technological wastewaters are rarely reported (Fudala-Ksiazek et al., 2016; d'Antonio et al., 2015; Nayono et al., 2010) and are usually overlooked. Additionally, the former Central and Eastern Bloc countries usually have to handle two types of landfill leachates - those generated by modern prisms (MPs), which meet EU requirements (generally post - 2010 operation) and those by previous prisms (PPs), usually arranged in an ad-hoc manner without any liner or pollution-control systems and with unlimited disposal of organic wastes.

The aim of this work was to determine the physical and chemical characteristics of all liquid by-products generated at MSWPs, especially in the context 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). Although the presence of PAEs and BPA has already been confirmed in landfill leachate (Table 1), thus far, little is known about their occurrence in MBT units' (SU and CU) technological wastewaters or their presence in leachates generated by MPs. In this study, liquid by-products were collected from two MSWPs, one serving a municipal area and the other serving a rural area. These selections are advantageous for addressing the main objectives of this study because they incorporate the generation, source-separation and even utilization of waste. At least part of the biodegradable solid waste, especially in rural areas, is used by households for composting, animal feed and/or recovery. The samples were collected from the MBT's SUs and CUs and from PPs and MPs, with limited and unlimited deposition of biodegradable solid waste, respectively (Fig. 1). All liquid by-products were tested for the presence of BPA and the selected PAEs. To better understand the occurrences of these compounds, their physical and chemical characteristics were determined, along with a metagenomic analysis of the bacterial community. In addition, a correlation matrix analysis, principal component analysis (PCA) and hierarchical cluster analysis (HCA) were performed to determine potential relationships between the parameters in the datasets.

2. Methods

2.1. Sample collection

In this study, liquid by-products were collected from two MSWPs, one situated in an urban area (the L-MSWP in Lezyce, which is located near the Tri-City conurbation) and the other in a rural area (ND-MSWP Nowy Dwor, which is located near Chojnice) in northern Poland, specifically in the Pomerania region (Table 2). Both facilities had already undergone extensive modernization to

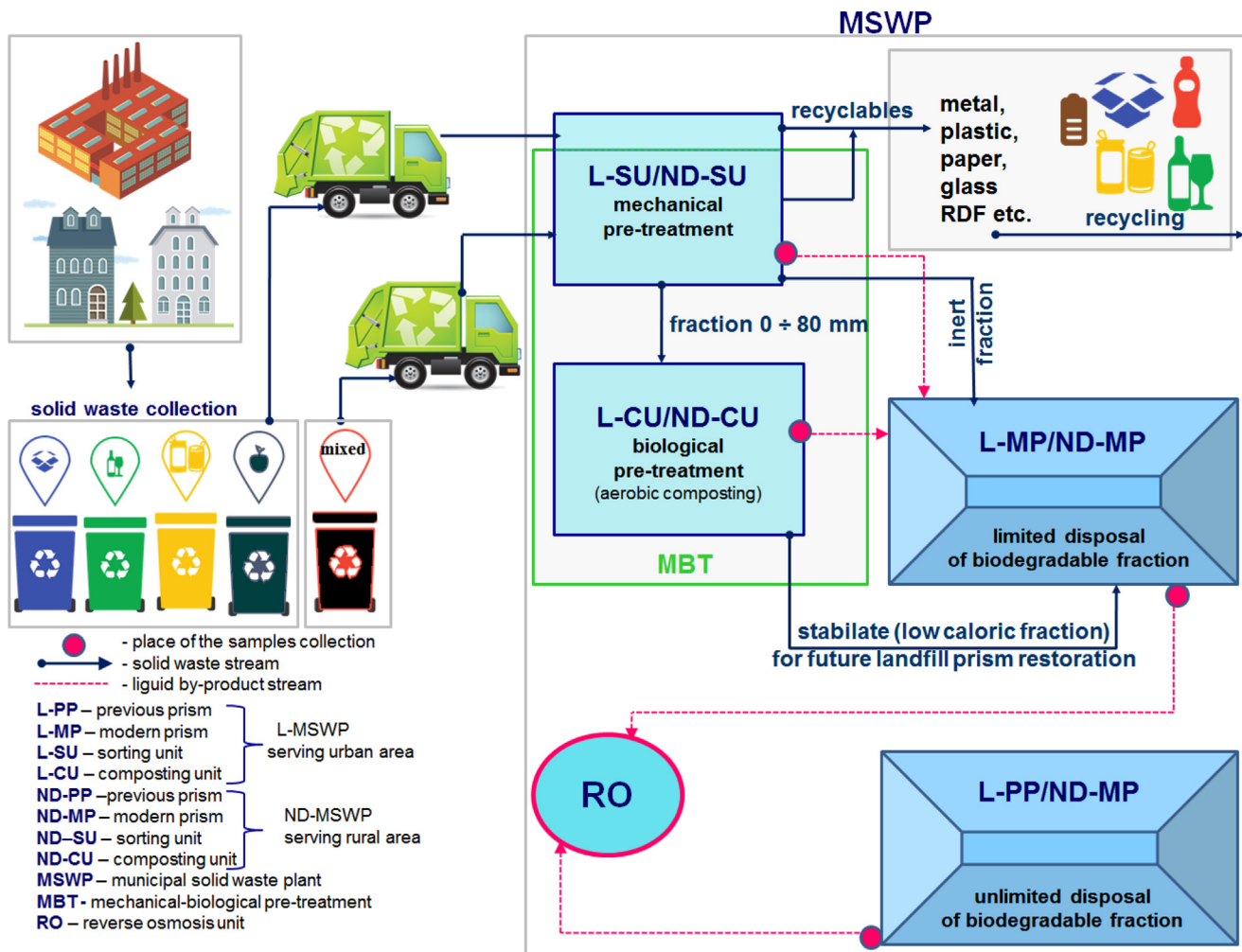


Fig. 1. Sample collection diagram.

improve waste pre-treatment prior to landfilling. Thus, the recycling of separately collected wastes is combined with the MBT of waste remaining in the residual (mixed) waste stream (see Fig. 1) via sorting (for recovery of secondary materials) and composting (aerobic bio-stabilization) of the 0–80 mm biodegradable fraction. The product obtained after composting (stabilate) at both MSWPs is used for prism restoration. The basic characteristics of the studied MSWPs are given in Table 2. In this study, the liquid by-products from the MPs, PPs, SUs and CUs were collected monthly as 24-h composite samples, although PAEs and BPA were tested every six months. Therefore, the analysis of some samples was performed monthly on an ongoing basis (mainly the basic physical and chemical analyses), whereas other samples were prepared/pre-treated and kept for analysis of PAEs, BPA and metagenomics toward the end of the study to minimize cost and ensure a consistent quality. The general characteristics of the municipal solid waste streams at the studied MSWPs during the sampling period are shown in Table S1.

2.2. Basic physical and chemical analyses

Among the basic physical and chemical characteristics, the following parameters were analysed according to APHA (American Public Health Association et al., 2005): pH and conductivity (Cond) using a portable multi-parameter meter (HL-HQ40d multi, HACH, Germany); total (TN) and inorganic N compounds (N-NH₄, N-NO₃,

and N-NO₂), total phosphorus (TP), orthophosphate (P-PO₄), chemical oxygen demand (COD), chloride (Cl⁻), and sulphate (SO₄²⁻) using a XION 500 spectrophotometer (Dr. Lange, GmbH, Germany); 5- and 20-day biochemical oxygen demand (BOD₅ and BOD₂₀) using the manometric respirometric BOD OxiTop® method; and total (TSS), mineral (MSS) and volatile suspended solids (VSSs) using the gravimetric method. Data from these physical and chemical analyses, although obtained monthly, were reported in this study only for samples that were simultaneously tested for PAEs and BPA.

2.3. Analysis of phthalates (PAEs) and bisphenol A (BPA)

The presence of PAEs and BPA was analysed every six months in the samples obtained from MSWPs, from December 2013 to June 2016 for ND-MSWP and from May 2012 to November 2015 for L-MSWP. In November 2011, an additional sample of the L-MP was taken because it was the first month of this prism operation.

The following components were added to a round-bottom test tube with a nut and gasket: 1 ml of THF/ACN 20/80 (v/v), 1 ml of 1% formic acid solution, 5 ml of sample, 3 g of sodium chloride, and 2 g of magnesium sulphate. The test tube and contents were shaken by hand for approximately 2 min. Then, the test tube was centrifuged (3000 rpm for 2 min). The upper organic layer was collected and analysed using gas chromatography–mass spectrometry (GC/MS).

Table 2
Basic characteristics of studied MSWPs.

	Eko Dolina Lezyce (L-MSWP)	Nowy Dwor (ND-MSWP)
Location and service	<ul style="list-style-type: none"> - Total area 106.5 ha; serves metropolitan area, approximately 460000 people - Receives approximately 200000 Mg of waste annually: 130000 Mg are municipal, and 97000 Mg are biodegradable 	<ul style="list-style-type: none"> - Total area 70 ha; serves small towns and agricultural area, ca. 150000 people - Receives approximately 34000 Mg of waste annually: 28000 Mg are municipal, and 8514 Mg are biodegradable
Solid waste collection scheme ^a	Householders and institutions have a free choice for waste collection; since 2013, monetary incentives have been used to promote source sorting and recycling; the schedules for source waste collection include I) unsorted (mixed) waste; II) source sorted waste - glass, plastics, paper and cardboard collected with selective manner plus residual waste (including food waste); separately collected: construction, large-size, public green waste and electrical/electronic equipment, batteries and others	since 2013, monetary incentives have been used to promote source sorting and recycling; the schedules for source waste collection include I) unsorted (mixed) waste; II) source sorted waste - glass, plastics, paper and cardboard collected with selective manner plus residual waste (including food waste); separately collected: construction, large-size, public green waste and electrical/electronic equipment, batteries and others
MBT Sorting unit (SU)	<ul style="list-style-type: none"> - L-SU in operation since March 2010; capacity of 150000 tons per year - Wastewater generated in the sorting process – approximately 0.7 m³/d - Initially, technological wastewater generated by the L-SU was directed to the L-PP; since March 2012, it has been directed to the L-MP <p>Technological wastewater contains: liquids used for washing the sorting lines and the areas where incoming solid wastes and final products (plastic, paper, textiles and glass) are stored, rainwater and some liquids, which are squeezed out of plastic bottles and other containers during their compression and baling</p>	<ul style="list-style-type: none"> - ND-SU in operation since July 2013; capacity of 35000 tons per year - Wastewater generated in the sorting process – approximately 0.5 m³/d - Technological wastewater generated by the ND-SU was directed to the ND-MP
MBT Composting unit (CU)	<ul style="list-style-type: none"> - L-CU - indoor unit; in operation since March 2010 - Wastewater generated in the composting process – approximately 15 m³/d - Initially, technological wastewater generated by the L-CU was directed to the L-PP; since May 2012, it has been directed to the L-MP <p>Technological wastewater is generated during the aerobic biodegradation of the pre-sorted waste stream</p>	<ul style="list-style-type: none"> - ND-CU - indoor unit, in operation since July 2013 - Technological wastewater generated in the composting process – approximately 2.5 m³/d - Technological wastewater generated by the ND-CU was directed to the ND-MP
Modern prism (MP)	<ul style="list-style-type: none"> - L-MP in operation since November 2011; the age in the sampling span: 1–49 months old - Leachates generated by the L-MP – approximately 71 m³/d - L-MP leachates have been directed to the L-RO unit 	<ul style="list-style-type: none"> - ND-MP in operation since July 2013, the age in the sampling span: 6–36 months old - Leachates generated by the ND-MP – approximately 18 m³/d - Initially, the ND-MP leachate was directed to the local wastewater system; since December 2013, it has been directed to the ND-RO
Previous prism (PP)	<ul style="list-style-type: none"> - L-PP in operation from January 2003 to November 2011; the age of the L-PP during the sampling was from 112 to 154 months - The restoration of the prism took place from September 2014 to September 2015; performed using stabilized solid waste (stabilate) from the L-CU - Approximately 39 m³/d of leachates are generated by the L-PP - Initially, the L-PP leachates were directed to the local wastewater system; since April 2005, they have been directed to the L-RO unit 	<ul style="list-style-type: none"> - ND-PP was in operation from July 1994 to July 2013^b; the age of the ND-PP during the sampling span was from 234 to 264 months - The restoration of the prism took place from November 2013 to September 2014 and was performed using stabilized solid waste (stabilate) obtained from the ND-CU - Approximately 12 m³/d of leachates are generated by the ND-PP - Initially, the ND-PP leachates were directed to the local wastewater system; since December 2013, they have been directed to the ND-RO unit
Leachates	Landfill leachates are generated as a result of precipitation, filtration, compaction and waste degradation	Landfill leachates are generated as a result of precipitation, filtration, compaction and waste degradation
Reverse osmosis unit (RO)	<ul style="list-style-type: none"> - Landfill leachate on-site pre-treatment in operation since April 2005- L-RO concentrate (approximately 52 m³/d) up to May 2012 was directed to the L-PP; then, to the L-MP; L-RO permeate is discharged to the local wastewater system 	<ul style="list-style-type: none"> - Landfill leachate on-site pre-treatment in operation since December 2013 - ND-RO concentrate (approximately 15 m³/d) is directed to the ND-MP - ND-RO permeate directed to the local wastewater system
Biogas	<ul style="list-style-type: none"> - The L-MSWP is equipped in the segment for utilizing biogas and has a transformer station. - Biogas is generated mainly by the L-PP and contains 50–75% CH₄, 25–45% CO₂, 10–10000 ppm H₂S and small amounts of other gases. - In November 2014, the L-MP also started to generate biogas containing methane (>1%), which, in June 2015, it reached 0%; since April 2015, biogas has been extracted 	<ul style="list-style-type: none"> - The L-MSWP - not equipped in biogas utilizing segment - biogas is burned in flare - At the ND-MSWP, the degassing wells to collect the landfill biogas were built at the ND-PP during the prism restoration (from November 2013 to September 2014); in 2015, the biogas contained up to 72% of methane. - In January 2014, the ND-MP started to generate biogas containing methane (>1%), which, in 2015, reached 27%

^a Dz.U. 2013 nr 0 poz. 38.

^b Constructed without liner system; personal communication from the MSWP operator responsible for the landfill gas management.

The GC/MS analyses were performed on a Shimadzu QP2010 SE gas chromatograph-mass spectrometer. The chromatographic separations were conducted using a ZB-5MS capillary column (30 m length, 0.25 mm internal diameter, and 0.50 µm film thickness) from Phenomenex. The carrier gas was helium with a flow rate of 1 ml/min. The injection was performed in split mode (5:1) at a temperature of 325 °C. The oven temperature was programmed as follows: 100 °C for 2 min; 40 °C/min increase to 325 °C, where it was held for 4 min. The injection volume was 2 µl. The interface temperature was 325 °C, and the ion source temperature was

200 °C. The ion energy for electron impact (EI) was 70 eV. The solvent cut time was 4 min. Mass detection was performed in the single-ion monitoring (SIM) mode. The selected ions (*m/z*) used for qualitative and quantitative purposes, the recoveries, reproducibility and blank experiments are shown in Table S2.

2.4. Microbiological analyses

The total genomic DNA was extracted from leachate samples generated by the L-MP using the commercially available Sherlock

AX kit (A&A Biotechnology, Poland). The samples were first transferred to 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 suggestions. The DNA concentrations of samples were determined using an ND-1000 UV-Vis spectrophotometer. The extracted DNA was stored at 4 °C. The microbial community in the tested L-MP samples was analysed using high-speed multiplexed 16S microbial sequencing on a MiSeq platform (Illumina). The V4 region of the 16S rRNA gene was amplified using an F515/R806 primer combination (5'-GTGC CAGCMGCCGCGTAA-3; 5'-CGACTACHVGGGTWCTAAT-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 Illumina-curated GreenGenes v13.5 database, using 16S Metagenomics Workflow.

2.5. Data evaluation using multivariate statistical methods

Unless otherwise specified, the dataset was divided by source. Parameters with too many values below the limit of detection (LOD) or obtained from a combination of other data (e.g., typical ratios) were excluded, while observations between the LOD and the limit of quantification (LOQ) were estimated using the misSMDA R package and checked (Josse and Husson, 2016).

Two multivariate statistical methods, hierarchical cluster analysis (HCA) and principal components analysis (PCA), were applied to subgroups of the full dataset to allow grouping of the technological wastewaters/landfill leachate samples and to determine correlations between variables, respectively. In this study, both HCA and PCA were performed using R statistical software (R Foundation, 2016) and the FactoMineR package (Lê et al., 2008).

Hierarchical cluster analysis refers to a collection of methods that seek to construct a hierarchically arranged sequence of parti-

tions for a given dataset. To understand the hierarchical dissimilarity relationship between data points, HCA was performed on the entire PCA-transformed dataset (BPA, PAEs and typical ratios excluded) using a combination of the Ward's linkage method as an agglomerative criterion (Ward, 1963) and the Euclidean distance as a measure of dissimilarity.

The central idea of PCA is to reduce the dimensionality of a dataset consisting of many interrelated variables while retaining as much information as possible about the variations present in the dataset. This process is achieved by transforming the data into a new set of uncorrelated variables ordered such that the first few retain most of the variation present in all of the original variables (Jolliffe, 2014).

PCA was conducted on different data subsets. Numerous BPA and PAE values for the ND-MSWP (in ND-PP, ND-MP, and ND-CU) were below the LOD, whereas a few were between the LOD and the LOQ. The L-PP, L-MP and L-CU PAE concentrations in the L-MSWP were also low. Because an eventual estimation of all of these uncertainties can strongly influence the PCA results, the correlation matrix and PCA for PAE was evaluated using only the SU data from both the L-MSPW and the ND-MSWP. Similarly, the PCA and correlation matrix for BPA concentrations were evaluated using only the L-PP and L-CU data and the ND-CU data.

3. Results and discussion

3.1. Basic physical and chemical analyses

The basic physical and chemical parameters of the samples analysed in this study are summarized in Table 3. As expected, the quality of the liquid by-product generated at each MSWP varied significantly.

In this study, special attention was given to the technological wastewater generated by the MBT processes, i.e., sorting and composting (aerobic stabilization), which fulfil the requirement of

Table 3
Physical and chemical data obtained for liquid by-products generated at the studied MSWPs.

Parameter	ND-MSWP				L-MSWP			
	min-max median	PP	MP	SU	CU	PP	MP	SU
pH	7.6–8.0 7.9	7.5–8.0 7.7	5.6–7.5 5.8	6.7–7.8.0 7.4	7.3–8.3 7.8	6.4–8.1 6.7	5.3–8.2 6.9	5.4–8.1 6.9
Cond [mS/cm]	4.1–15.3 10.7	4.4–18.9 8.8	5.2–10.7 6.2	6.9–22.1 17.8	23.6–31.1 28.9	5.4–23.2 12.9	1.9–8.3 4.5	4.2–19 15.7
COD [mgO ₂ /L]	336–2590 927	310–2125 1257	3000–21280 14695	8420–28500 13420	3260–4715 4270	1011–12620 1925	1370–20854 7180	6250–52980 27860
BOD ₅ [mgO ₂ /L]	54–293 168	42–244 248	1102–11440 8750	240–18080 7485	338–677 361	236–7490 439	749–11260 5390	2766–32200 15960
TN [mgN/L]	214–852 683	110–828 387	258–408 310	356–1418 1051	1725–2510 2325	145–810 705	1120–1750 307	1120–1750 1362
Norg [mgNorg/L]	11–192 78	9–145 33	64–119 97	14–146 96	6–303 69	7–132 37	6–308 74	77–226 126
N-NH ₄ [mgN-NH ₄ /L]	195–786 563	85–657 339	152–274 221	255–1392 916	1704–2295 2175	155–820 652	137–468 229	926–1610 1222
N-NO ₃ [mgN-NO ₃ /L]	3.4–7.9 5.9	1.6–18 10.8	2.5–10.2 5.6	4–15.9 10.4	14.7–24 21	3.3–15.9 5.4	1.1–28.2 3.5	13.1–48.7 21.1
N-NO ₂ [mgN-NO ₂ /L]	0.1–5.4 0.6	0.1–14.4 2.2	0.6–4.6 2.5	0.4–14.5 2	0.2–0.6 0.4	0.2–0.5 0.4	0.4–5.5 2.9	1.7–5.6 3.1
TP [mgP /L]	1–13.5 4.3	1.5–12.8 4.9	21.3–47 38.2	4.9–23.6 12.4	17.7–26.4 23.4	5.1–15.3 6.1	14.7–120 53.2	7.3–83.3 31.2
P-PO ₄ [mgP-PO ₄ /L]	0.4–11.8 3.7	1.2–8.6 4.2	2.6–33 28.8	2.3–18.1 5.4	14–43.6 20.0	2.1–12.7 4.1	12–95.5 39.8	3.2–40.2 16.3
TSS [mg/L]	10–244 70	35–730 284	235–4040 1240	75–990 379	13–80 31	13–596 230	95–3140 790	323–9667 1785
MSS [mg/L]	8–104 16	7–380 181	120–290 275	55–193 138	2–24 9	8–174 23	28–1055 87	90–1267 327
VSS [mg/L]	2–140 55	20–350 88	115–3755 913	20–825 230	4–60 22	1–574 97	50–2690 1420	220–8400 1420
SO ₄ ²⁻ [mgSO ₄ ²⁻ /L]	46–232 72	208–1318 660	154–1580 894	146–1080 585	1015–2430 1640	42–475 161	141–9460 545	860–2420 1250
Cl ⁻ [mgCl ⁻ /L]	355–2127 1312	511–3971 1419	603–1716 993	659–4254 1879	2836–4786 3545	851–2595 1489	177–1454 390	411–2039 1241
BOD ₅ /COD [-]	0.05–0.35 0.18	0.11–0.45 0.15	0.07–0.64 0.56	0.25–0.72 0.61	0.07–0.16 0.11	0.17–0.59 0.28	0.36–0.82 0.56	0.44–0.87 0.6
TN/BOD ₅ [-]	2.38–8.04 3.54	0.42–4.9 0.7	0.03–0.27 0.04	0.07–0.2 0.12	3.28–6.97 4.88	0.07–2.88 0.08	0.04–0.25 0.08	0.04–0.48 0.14
Norg/TN [-]	5.0–31.4 9.4	5.1–20.9 15	21.5–28 30.1	1.0–27.2 12.4	0.4–12.1 3.2	1.0–18.6 21.5	4.5–38.8 9.6	5.0–17.2 9.6
N-NH ₄ /TN [-]	65.8–94.5 88.3	77.6–91.3 80.4	58.8–77.5 66.8	71.6–98.2 86.4	87.0–98.8 95.7	80.5–98.2 84.5	57.8–94.5 76.0	80.5–92.7 89.1
N-NO ₃ /TN [-]	0.5–3.7 0.8	1.2–10.5 1.6	0.8–3.0 2.0	0.7–1.5 1.2	0.8–1.0 0.9	0.7–2.0 1.4	0.7–3.5 0.8	1.2–2.8 1.4
N-NO ₂ /TN [-]	0.01–1.25 0.14	0.05–1.96 0.78	0.2–1.13 0.79	0.11–1.5 0.18	0.0–0.02 0.01	0.04–0.1 0.05	0.19–1.19 0.55	0.15–0.43 0.18
P-PO ₄ /TP [-]	36.5–88.6 87.4	54.5–97.8 71.9	5.9–81.6 70.7	18.1–76.7 59.0	75.7–173 81.6	34.3–83.0 72.6	66.5–94.5 79.4	25.7–62.3 46.5
VSS/TSS [-]	19–80 45	8–62 34	7–49 27	17–47 29	14–92 75	8–96 54	53–93 77	68–97 80

waste stream pre-treatment prior to landfilling but generate liquid by-products (see Table 2).

The technological wastewaters generated by both sorting units (L-SU and ND-SU) and both composting units (L-CU and ND-CU) contained high concentrations of suspended solids (up to 9667 mg/L) and acted as a rich source of ammonia nitrogen (up to 1610 mg N-NH₄/L) and organic matter (up to 52980 mg O₂/L), more than half of which was biodegradable (BOD₅/COD > 0.5) (Table 3). The results obtained in this study for the CU in the municipal area were similar to the data presented by d'Antonio et al. (2015) but lower than those given by Nayono et al. (2010). It should be noted that the technological wastewaters obtained from L-SU and L-CU were characterized by higher chemical parameter values than those obtained from ND-SU and ND-CU. In this case, the differences in solid waste compositions should be considered. In Poland, there is a striking difference in municipal waste composition between urban and rural areas (den Boer et al., 2010), and this difference is also evident in the MSWPs chosen for this study. However, in both MSWPs, the forms of the source waste are similar: sorted (glass, plastics, paper and cardboard plus residual (mixed) waste, which includes food waste) or unsorted (mixed). At L-MSWP, 74.6% of the municipal solid waste is regarded as biodegradable, while at ND-MSWP, only 30% (see Table 2). Apart from a general pattern of higher paper and plastic generation in urban areas (mainly connected with food consumption habits), the difference in wastes is also explained by the fact that, in the countryside, certain fractions are frequently used as a fuel for individual heating systems due to their energy value. This pattern is also confirmed by the higher share of the fine (ash) fraction in the ND-MSWP waste stream, especially during winter (personal communication). Nevertheless, the obtained physical and chemical data indicate that the SU and CU at both MBTs are new important sources of highly contaminated wastewater streams in the liquid by-product management of MSWPs.

Because it is difficult to develop effective and affordable technologies for treatment of such technological wastewater, the liquid by-products generated by CUs and SUs are currently directed to the landfill prisms according to the schematic given in Fig. 1 and explained in Table 2. The on-site pre-treatment of the highly loaded wastewater mentioned above and the solid waste compositions are thought to affect the processes occurring in the landfill prisms. For example, the closed and already restored prisms (L-PP and ND-PP) both generated leachates containing rather consistent amounts of organic matter (expressed as COD and BOD₅) and ammonia, but the average values were approximately 4 times higher in the L-PP samples than in the ND-PP samples (e.g., the median COD values were 4270 mg O₂/L and 927 mg O₂/L, respectively, and the median BOD₅ values were 361 mg O₂/L and 168 mg O₂/L, respectively) (Table 3). In this case, however, besides the composition of solid waste stream, the lower physicochemical parameter values in the ND-PP leachates could be also explained by the lack of a proper liner system. The existing natural insulating barrier is considered to be insufficient both in terms of protecting the prism from possible periodical scouring by local groundwater and protecting the groundwater and soil environment from contamination by ND-MSWP-sourced pollutants.

Nonetheless, the low biodegradability of the leachates (BOD₅/COD < 0.2) obtained from both PPs (L-PP and ND-PP), combined with the high ammonia concentration, constituted more than 83% of the total nitrogen (up to 2295 mg N-NH₄/L and 786 mg N-NH₄/L, respectively), and the generated biogas with a high proportion of methane (>40%) suggests methanogenic waste decomposition (see Tables 2 and 3). The obtained data agree with previous studies (Crawford and Smith, 1985; Spagni et al., 2008) that suggest that, after five years of operation, a landfill prism becomes stable, while methane production can be typically

observed for more than 20 years. This phenomenon was observed in the case of the ND-PP, in which the sampling was conducted over prism ages of 235–265 months. In contrast, the L-PP was sampled at ages of 112–154 months (see Table 2).

Currently, the progressively decreasing disposal of biodegradable organic matter is suspected to have a profound influence on the biological and chemical processes occurring in modern landfill prisms (L-MP and ND-MP). Thus, in this study, the emission potentials of MPs (L-MP and ND-MP, which, during the sampling span, had been operated from 1 to 49 months and from 6 to 36 months, respectively; see Table 2) are expected to significantly differ from the previous cases (L-PP and ND-PP). In general, during the first 6 and 12 months of L-MP and ND-MP operation, high values of suspended solids (up to 596 mg/L and up to 730 mg O₂/L, respectively), COD (up to 12,620 mg O₂/Land up to 2125 mg O₂/L, respectively) and BOD₅ (up to 7490 mg O₂/Land up to 344 mg O₂/L, respectively) were observed, with BOD₅/COD ratios exceeding 5. Then, these parameters tended to decrease, especially the easily biodegradable organic matter (expressed as BOD₅), which was assumed to be hydrolysed into fatty acids, amino acids, and monosaccharides. As confirmation of the aforementioned process, the L-MP began generating biogas in November 2014, and by June 2015, the biogas contained approximately 70% of methane (Table 2). These observations suggest that the L-MP entered the methane fermentation phase (confirmed by metagenomic assay; compare with Fudala-Ksiazek et al. (2016)). In the case of the ND-MP, biogas with 1% of methane has been observed since January 2014; however, up to the end of the study, it was still of very poor quality (only up to 30% of methane) (Table 2). The data obtained for the ND-MP were correlated with the slow increase in the *mcrA* copy gene abundance (as a ratio to the 16S rDNA copy genes; data not shown).

As mentioned above, the unexpected high biogas production, especially that observed in the L-MP, can be partly explained by the on-site pre-treatment discussed above for the technological wastewater generated during solid waste sorting and composting. In general, these wastewaters injected into the landfill prism serve as an additional stream of easily biodegradable organic matter (BOD₅/COD > 0.6 in both the L-CU and L-SU) (Table 3) and potentially also a source of undesirable pollutants, which can be released into the landfill leachates.

3.2. Phthalates (PAEs) and bisphenol A (BPA)

According to the obtained results, PAEs and BPA were detected in leachates generated by PPs and MPs and in technological wastewaters obtained from SUs and CUs (Table 4). Their incidence and concentrations were highly dependent on the sampling point (Table S3). In general, both types of compounds appeared with the highest frequency in technological wastewater generated by the SUs and CUs. Moreover, the presence of PAEs and BPA has not been previously reported and studied in wastewater generated during the MBT sorting and composting processes.

In the case of the SUs, it was assumed that the presence of PAEs and BPA was mainly connected with their release from the products that incorporated them and not from biodegradation. Additionally, it was suspected that LMW phthalates, due to their relatively hydrophilic nature, could be easily released from resin and more readily leached from the waste matrix. The obtained data confirmed this statement to some extent. In the technological wastewater generated by both SUs, LMW phthalates, such as DMP and DEP, were detected with the highest frequency (see Table S3 for details). In the case of the L-SU, the DMP and DEP concentrations ranged from above the LOD to 117 µg/L and from 20.4 µg/L to 260.4 µg/L, respectively, while for the ND-SU, the DMP and DEP values were higher, up to 1405.2 µg/L and

Table 4

The presence of selected PAEs and BPA obtained in this study.

Abbreviation	DMP	DEP	DnBP	BBzP	DEHP	DnOP	BPA	
Characteristics of phthalates (PAEs) and bisphenol A (BPA)								
common name	Dimethyl phthalate	Diethyl phthalate	Di-n-butyl phthalate	Benzyl butyl phthalate	Bis (2-ethylhexyl) phthalate	Di-n-octyl phthalate	Bisphenol a	
CAS No	131-11-3	84-66-2	84-74-2	85-68-7	117-81-7	117-84-0	80-05-7	
Molecular Formula	C ₁₀ H ₁₀ O ₄	C ₁₂ H ₁₄ O ₄	C ₁₆ H ₂₂ O ₄	C ₁₉ H ₂₀ O ₄	C ₂₄ H ₃₈ O ₄	C ₂₄ H ₃₈ O ₄	C ₁₅ H ₁₆ O ₂	
Sample origin ^a	Data obtained in this study [µg/L] given as			$\frac{\text{min-max}}{<LOD/<LOQ/>LOQ}$ ^b				
ND-MSWP	PP	$\frac{<LOD}{6/0/0}$	$\frac{<LOD,<LOQ}{4/2/0}$	$\frac{<LOD,<LOQ}{5/1/0}$	$\frac{<LOD}{6/0/0}$	$\frac{<LOD,<LOQ}{3/3/0}$	$\frac{<LOD-6.3}{5/0/1}$	$\frac{<LOD-183}{5/0/1}$
	MP	$\frac{<LOD}{6/0/0}$	$\frac{<LOD,<LOQ}{5/1/0}$	$\frac{<LOD}{6/0/0}$	$\frac{<LOD}{6/0/0}$	$\frac{<LOD,<LOQ}{4/2/0}$	$\frac{<LOD,<LOQ}{5/1/0}$	$\frac{<LOD-151}{3/2/1}$
	SU	$\frac{28.3-1405}{0/0/6}$	$\frac{21.5-229.9}{0/0/6}$	$\frac{<LOD-241.9}{2/1/3}$	$\frac{<LOD-124.6}{3/0/3}$	$\frac{<LOD-1683}{2/1/3}$	$\frac{<LOD-114}{3/1/2}$	$\frac{<LOD-292}{0/2/4}$
	CU	$\frac{<LOD-993.5}{3/2/1}$	$\frac{<LOD}{6/0/0}$	$\frac{<LOD-144}{5/0/1}$	$\frac{<LOD-4.2}{4/0/2}$	$\frac{<LOD-561.2}{4/1/1}$	$\frac{<LOD-87.1}{5/0/1}$	$\frac{<LOD-418.1}{0/1/5}$
L-MSWP	PP	$\frac{<LOD-27.9}{6/1/1}$	$\frac{<LOD,<LOQ}{7/1/0}$	$\frac{<LOD}{8/0/0}$	$\frac{<LOD}{8/0/0}$	$\frac{<LOD-256.7}{4/1/3}$	$\frac{<LOD}{8/0/0}$	$\frac{856-8052}{0/0/8}$
	MP	$\frac{<LOD-23.1}{4/4/1}$	$\frac{<LOD-46.2}{5/2/2}$	$\frac{<LOD}{8/0/0}$	$\frac{<LOD-1.7}{8/0/1}$	$\frac{<LOD-536.4}{6/2/1}$	$\frac{<LOD}{8/0/0}$	$\frac{<LOD-150.1}{0/6/3}$
	SU	$\frac{<LOD-117.7}{4/0/4}$	$\frac{20.4-260.4}{0/0/8}$	$\frac{<LOD-2321}{4/1/3}$	$\frac{<LOD-828.9}{3/0/5}$	$\frac{<LOD-8201}{0/4/4}$	$\frac{<LOD-47.7}{4/0/4}$	$\frac{<LOD-257.5}{0/7/1}$
	CU	$\frac{<LOD-61.6}{6/0/2}$	$\frac{<LOQ-65.8}{0/5/3}$	$\frac{<LOD-139.6}{6/1/1}$	$\frac{<LOD}{8/0/0}$	$\frac{<LOD-612.5}{0/6/2}$	$\frac{<LOD-7.3}{7/0/1}$	$\frac{<LOD-1795}{0/0/8}$

^a In this study - samples originated from ND-MSWP and L-MSWP – municipal solid waste plant serving rural and urban area, respectively, from landfill leachate generated by PP- previous prism and MP – modern prism, and from technological wastewater obtained from SU – sorting unit and CU – composting unit;

^b <LOD/<LOQ/>LOQ – number of samples with the results below LOD (<LOD), with results between LOD and LOQ, with results over LOQ (>LOQ).

229.9 µg/L, respectively. Among the higher-molecular-weight and more hydrophobic phthalates, DEHP was most frequently detected. In the L-SU, the highest concentrations of total PAEs were noted in May 2012 (5201.3 µg/L) and November 2015 (8966.8 µg/L) and were caused by the presence of all tested phthalates, but mainly by elevated concentrations of DEHP (1626.1 µg/L and 8205.8 µg/L, respectively) and DnBP (2320.5 µg/L and 656.6 µg/L, respectively). These two phthalates, together with BPA, for which an elevated concentration was noted in May 2012 (828.9 µg/L), are of special concern in the EU because these compounds are restricted in toys and some large and small household appliances according to Annex XVII to REACH Regulation (EC) No. 1907/2006 (EU, 2006) and Directive (EU) 2015/863 (EU, 2015a). Additionally, DEHP is considered to be carcinogenic to humans (Group 2B) (International Agency for Research on Cancer, 1987) and is identified by EU legislation as hazardous and one of 45 priority substances in the field of water policy (EU, 2013). Despite the prohibition or restriction of some PAEs, they remain present in packaging materials and appear in food residues and personal care products (Fierens et al., 2012; Hubinger and Havery, 2006; Koniecki et al., 2011).

Thus, these products may serve as a source of PAEs if they enter the waste stream. In a manner similar to that of PAEs, the waste stream can serve as a source of BPA. Interestingly, in this study, the technological wastewater generated by the ND-SU contained higher concentrations of BPA (from 160.9 µg/L to 292.0 µg/L) than that of the L-SU, where BPA was detected in each sample but quantified only once, namely, in May 2012 (257.0 µg/L). However, in addition to the waste stream, the products used for facility washing could not be excluded as a source of PAEs and BPA in both SUs. The sorting lines and the waste-receiving and staging areas are washed at least twice per month and even more often when necessary during the summertime. According to the material safety data, the washing products used contain, among other compounds, hydrotropes (e.g., sodium cumenesulfonate; CAS No. 28348-53-0), which increase the solubility of hydrophobic compounds in aqueous solutions, including compounds that may contain PAEs and BPA, e.g., synthetic resins, adhesives, and printing inks. According to the obtained results, wastewater generated by both SUs serves as a rich source of PAEs and BPA, with concentrations several times higher than the PAEs/BPA concentrations noted

in rivers, wastewaters and even landfill leachates reported in this and other studies (see Tables 1 and 4).

Similar to the SUs, the technological wastewaters generated by the sampled CUs also served as a rich source of PAEs and BPA (Tables 4 and S3). Specifically, BPA was detected in high concentrations ranging from 159.8 µg/L to 1795.0 µg/L in the L-CU and from 141.0 µg/L to 418.1 µg/L in the ND-CU. In the case of PAEs, DEP and DEHP were detected in each sample obtained from the L-CU but quantified in only three (average of 58.9±6.4 µg/L) and two (293.0 µg/L and 612.5 µg/L) samples, respectively. Other PAEs were occasionally detected (Table S3). PAEs were also occasionally detected in technological wastewater obtained from the ND-CU. Especially high concentrations of selected PAEs (DMP = 993.5 µg/L, DnBP = 144 µg/L, DEHP = 561.6 µg/L and DnOP = 87.1 µg/L) were noted in December 2015. In the case of the composting process, the presence/absence of PAEs and BPA in technological wastewaters must be analysed in terms of the quality of the solid waste subjected to composting and the leaching/biodegradation susceptibility of the tested compounds. Currently, despite the recommendations for recycling of different plastic and packaging materials, their debris (scattered fragments) can often be found in the household residual (mixed) waste stream. This is of special concern in countries such as Poland, where the tradition of household recycling and sorting of solid waste is rather new (see Table 2). In addition to kitchen waste and green waste, the residual (mixed) waste is often contaminated by foil, thin plastic carry bags, other plastic elements, paper and multi-layered or multi-material packaging, used batteries, textiles, mineral wastes, ash originating from individual heating systems, and crushed glass (den Boer et al., 2010). In the residual (mixed) waste stream, these products are directed to the sorting facility. Because they are often not recovered (e.g., soggy paper or plastic debris), they end up in the biological stabilization and/or as ballast for landfilling (Fig. 1). Several sources of BPA emission were recognized (Table 1) (Liao and Kannan, 2011). It is suspected that the amount of BPA derived from the paper waste stream may be underestimated because Biedermann et al. (2010) revealed that thermal printing papers contained up to 17 mg BPA/g paper (with an average of 13.3 mg/g). This is up to 1000 times higher than the amount of BPA found in canned food or leached from a BPA-based plastic baby bottle (Lunder et al., 2010). In addition to BPA (and its analogues), paper products may also contain

many other chemical substances (Bradley et al., 2008), including PAEs, which are used in printing inks, lacquers, dispersion glues, and softeners in tissue paper (Abildgaard et al., 2003).

Additionally, PAEs (especially DBP and DEHP) and BPA may accumulate via recycling because higher concentrations of these compounds were noted in the recycled paper than in virgin fibre-based products (Pivnenko et al., 2015). Thus, the recycling loop may cause unintended spreading of these chemicals (Pivnenko et al., 2015). Furthermore, the residual ash from municipal solid waste plant incinerators was suggested to be another poorly recognized source of BPA by Kurata et al. (2008). In this study, high concentrations of BPA was noted in ash from local heating systems, especially in the ND-MSWP, which serves a rural area. Incineration, even under controlled conditions, is not able to completely destroy BPA (Morin et al., 2015), and ash originating from household heating systems that are fed with combustible waste may act as a source of BPA. However, Morin et al. (2015) noted that incineration conducted under controlled conditions significantly lowers the total amount of BPA compared to that in the source waste. Nonetheless, the presence of the products mentioned above in the stream of solid waste subjected to a 40-day composting process may serve as a source of PAEs and BPA. However, during composting, both PAEs and BPA are suspected to degrade especially under aerobic conditions (Juneson et al., 2002; Wang et al., 2004). To date, several pathways for the degradation of BPA and PAEs have been suggested, although they vary among bacterial genera (Zhou et al., 2015). For PAEs, their degradation into phthalic acid and corresponding alcohols may be direct or via monoesters (Jonsson et al., 2003); thus, elevated concentrations of these monoesters and/or phthalic acid in landfill leachate have been used to confirm the degradation of PAEs. Nonetheless, the presence of PAEs and BPA in technological wastewater generated by the sampled CUs indicates that these substances are typically not completely degraded and/or the leaching exceeds the biodegradation rate.

Biodegradation has been shown to be crucial in terms of the presence of PAEs and BPA in landfill leachates (Jonsson et al., 2003; Peng et al., 2015), and it is important to recognize the source of tested compounds in landfill prisms. PAE- and BPA-rich products could have been deposited without restriction during the activity of the PPs (L-PP and ND-PP). Although recycling and solid waste separation was not a common procedure, the usage of plastic in food production was rather rare in Poland before entering the EU in 2004. In the case of MPs (L-MP and ND-MP), BPA and PAEs are suspected to be mainly derived from unreacted monomers and primarily disposed with ballast waste. However, in light of the results reported above, the on-site pre-treatment of technological wastewater generated by SUs and CUs may also be important.

In general, in the landfill leachates tested in this study, DMP, DEP and DEHP were detected with the highest frequency and were quantified in some samples (Tables 4 and S3). As with the SUs and CUs, the results obtained for landfill leachates also indirectly confirmed that DMP and DEP could be more easily leached from the polymer chains than higher-molecular-weight phthalates. For example, DMP and DEP were present in L-MP leachate during the first month through the 18th month of L-MP operation (Table S3), while DEHP appeared after 19 months of L-MP operation. In contrast, in the ND-MP leachate, PAEs (only DEP, DEHP and DnOP) were detected only after 30 months of prism operation. Moreover, their concentrations were too low to be quantified.

In the case of the ND-PP, PAEs (mainly DMP, DEP, and DEHP) were detected with similar frequency as in the L-PP, although usually in concentrations below the LOQ (only DnOP was quantified in June 2015 at 6.3 µg/L). In general, in landfill leachates, DEHP was the compound most commonly detected among the tested PAEs and had the highest concentrations (536.4 µg/L for L-MP and 256.7 µg/L for L-PP).

Other researchers have confirmed that phthalates in landfill leachate might range from undetectable to concentrations above 450 µg/L, with DEHP reaching the highest concentrations (see Table 1).

In comparison to PAEs, BPA was more often detected and quantified in landfill leachates. In particular, the L-PP leachates were identified as an important source of BPA. Most of the results varied from 987.2 µg/L to 1599.3 µg/L. In November 2014 and May 2015, elevated concentrations of BPA were noted (8052.2 µg/L and 2547.0 µg/L, respectively). Interestingly, elevated concentrations of organic matter (COD and BOD₅), nitrogen, and phosphorus were simultaneously noted in the L-PP leachate (cf. Fudala-Ksiazek et al., 2016). During that time (from September 2014 to September 2015), L-PP restoration was conducted using stabilate obtained from the L-CU (see Table 2 for explanation). According to the accepted restoration procedure, stabilate was applied to the top of the L-PP and was exposed to environmental conditions for four months before covering. The stable residue obtained from the L-CU may serve as a source of BPA because the L-CU technological wastewater was rich in this pollutant, as was discussed above. Such a correlation was not so clearly seen during the restoration of the ND-PP conducted from November 2013 to September 2014. This can be partly explained by the restoration procedure. The prisms were partially covered by sections so that the stabilate was exposed to environmental conditions for a shorter period of time than that of the L-PP, usually for 30 days. However, in the ND-PP leachates, PAEs occurred only during and after the restoration of the prisms (see Table S3). In the case of the MPs, BPA was detected in each sample from the L-MP and was quantified in three samples in similar concentrations of 127.1–150.1 µg/L. In contrast, BPA was detected in the ND-MP after June 2015, although it was only quantified in June 2016. Because the technological wastewater generated by the SUs and CUs at both MSWPs was reported as being rich in BPA, their injection into the MPs likely acted as a source of this compound.

Both PAEs and BPA might undergo biodegradation in the prism body. Jonsson et al. (2003) observed a strong correlation between PAE hydrolysis and methanogenic flora. The data obtained in this study indirectly confirm this proposition. Because both PPs were in the methanogenic phase, no PAEs were detected in the PP leachate except at the time of prism restoration and when the wastewaters generated by the L-SU and L-CU were directed to the L-PP (May 2012). Moreover, in the case of the L-MP, PAEs were not detected in the L-MP leachate after two years of operation, when a significant increase in archaea in the complex microbial community was noted by a metagenomic approach, as discussed in detail by Fudala-Ksiazek et al. (2016).

To date, bacteria capable of degrading PAEs and BPA have been studied and isolated from several natural environments, such as river water, river sediments, seawater, coastal sediments, soil, plant rhizospheres, wastewater, and other engineered environments (see Table S4). In this study, the landfill leachates generated by L-MP were analysed using next-generation sequencing to determine the presence of bacteria with the specific ability to degrade PAEs and BPA. According to the obtained results, summarized in Table S4, bacteria, which are able to degrade PAEs and BPA as pure cultures, in studied MP leachates represented four phyla: *Proteobacteria*, *Firmicutes*, *Actinobacteria* and *Bacteroidetes* (Table S4). However, complete mineralization of PAEs and/or BPA requires diverse metabolic genes and enzymes that are rarely found in single bacteria cells. Thus, it is expected that in landfill prisms, the complete degradation of the studied compounds is performed by a complex community, in which different specialized microorganisms can be involved in certain degradation steps. It is especially important under anaerobic conditions because phthalate degradation is energetically unfavourable by bacteria and only degradation

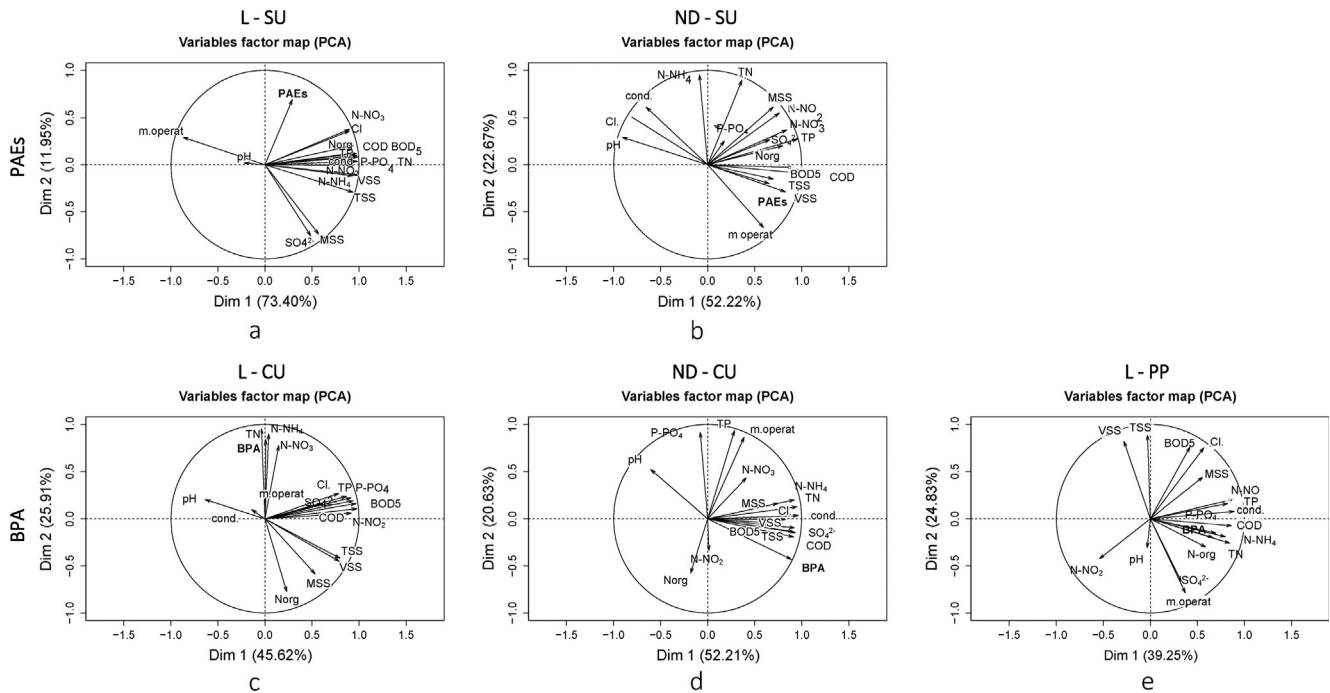


Fig. 2. PCA Variables factor map for selected datasets.

by syntrophic bacterial cultures make the entire reaction energetically feasible (Kleerebezem et al., 1999; Qiu et al., 2006).

3.3. Data analysis (HCA, PCA, and correlation plot)

In this study, two multivariate statistical methods, hierarchical cluster analysis and principal component analysis, were applied to analyse the dataset on liquid by-products generated at the SUs, CUs, PPs and MPs of the studied MSWPs.

According to the hierarchical clustering dendrogram presented in the supplementary information (Fig. S1) and the first principal component, the information can be sorted into three different clusters. The first comprised four samples originating from the L-MSWP, which serves an urban area. They were taken from the L-SU in May and November 2012 and from the L-CU in May 2014 and May 2015. These samples differed from the other samples because they were characterized by relatively high COD, BOD₅, Cl⁻, TSS, VSS and SO₄²⁻ concentrations.

The second cluster accounts for all of the L-PP samples. In the third cluster, the remaining samples are similar in composition but originate from different sampling points (L-SU and ND-SU, L-CU and ND-CU, and ND-PP jointly with the ND-MP leachates).

Principal component analysis was used in this study to determine if any pattern existed among the PAE and BPA concentrations obtained in the tested liquid by-products and other chemical-physical parameters. However, because many BPA and PAEs concentration values were found to be below the LOD or between the LOD and the LOQ, the PCA was conducted on a limited dataset so that these uncertainties would not influence the PCA results. Thus, for PAEs, the correlation matrix and PCA were applied only to technological wastewater samples taken from the SUs (L-SU and ND-SU); for BPA, the statistical analysis was applied only to samples from the L-PP, L-CU and ND-CU.

According to the obtained results, the first two components in the L-SU PCA explained 85.36% of the total variance, with the first component accounting for 73.4%, as shown in Fig. 2a and in Table 5. The first component accounted for almost all the physical and

chemical parameters. Similarly, for the ND-SU (Fig. 2b), the first two components accounted for 74.9% of the total variance and exhibited a similar trend, accounting for all variables except for the Cl⁻ concentration, pH and conductivity. However, for these parameters, the correlations were not significant (p -value < 0.05). Interestingly, the PCA of the L-SU datasets did not show any significant correlation with PAEs; for the ND-SU dataset, PAEs were positively correlated with TP, N-NO₃ and BOD₅.

In the case of the L-PP, a PCA plot of PC1 vs. PC2 (Fig. 2e) indicated that these components accounted for 64.1% of the total variation and showed that all individuals were well represented by only the first dimension (with the exception of the first sample taken in May 2012, which showed higher BOD₅ and Cl⁻ concentrations than the others). All significant correlations had a positive contribution in this component. However, the BPA concentration is present in the first component of this dataset, although it is not significant (p value = 0.071).

For technological wastewater originating from both CUs (L-CU and ND-CU), the BPA correlation was found to be significant. In the L-CU (Fig. 2c), the first two components explained 71.53% of the cumulative variance, with the first component accounting for 45.62%. BPA was grouped into the second component, together with N-NH₄, TN, and N-NO₃. These parameters were inversely correlated with N-org. This relationship aspect can be explained by biological processes, which typically occur during composting, and by the oxidation of nitrogen-containing organic compounds, which contribute to the formation of ammonia and nitrite/nitrate (Williams, 2005).

In the ND-CU, a PCA plot for PC1 vs. PC2 (Fig. 2d) indicated that these components accounted for 72.84% of the total variation and showed that BPA is part of the first component, along with many other parameters. Nevertheless, wastewater samples taken specifically from this source contained relatively high concentrations of PAEs and BPA. In addition, based on the first component in the ND-CU analysis, BPA was positively correlated with N-NH₄ and suspended solids. This observation requires future study because, in landfill leachates and storm water runoff, the organic pollutants,

Table 5

Significant correlations (correlation > 0.7 and p-value < 0.05) between variables in the first two PCs (ordered by decreasing correlation). Some non-significant correlations (correlation > 0.7 and p-value < 0.07) are reported in italics.

	L - SU				ND - SU			
	PC1 (73.4%)		PC 2 (11.95%)		PC 1 (52.22%)		PC 2 (22.67%)	
PAEs	COD	0.99	PAEs	0.69	TP	0.96	N-NH ₄	0.95
	TN	0.98	MSS	-0.74	BOD ₅	0.91	TN	0.89
	P-PO ₄	0.98	SO ₄ ²⁻	-0.76	N-NO ₃	0.84		
	VSS	0.97			PAEs	0.83		
	cond.	0.97			Norg	0.8		
	BOD ₅	0.97			N-NO ₂	0.76		
	TP	0.95			Cl ⁻	-0.81		
	N-NO ₂	0.94						
	Norg	0.94						
	TSS	0.94						
	N-NH ₄	0.93						
	N-NO ₃	0.9						
	Cl ⁻	0.9						
	m.op.	-0.87						

	L - CU				ND - CU		L - PP					
	PC 1 (45.62%)		PC 2 (25.91%)		PC 1 (52.21%)		PC 2 (20.63%)		PC 1 (39.25%)		PC 2 (24.83%)	
BPA	COD	0.98	N-NH ₄	0.95	cond.	0.96	TP	0.93	cond.	0.89	TSS	0.89
	BOD ₅	0.97	TN	0.9	TN	0.95	P-PO ₄	0.91	N-NO ₃	0.89	VSS	0.82
	SO ₄ ²⁻	0.94	BPA	0.84	SO ₄ ²⁻	0.93	m.op.	0.86	COD	0.86	BOD ₅	0.76
	N-NO ₂	0.92	N-NO ₃	0.78	TSS	0.92			TN	0.84	Cl ⁻	0.75
	P-PO ₄	0.91	Norg	-0.77	N-NH ₄	0.92			TP	0.82	SO ₄ ²⁻	-0.66
	TP	0.86			COD	0.91			N-NH ₄	0.8	m.oper	-0.79
	VSS	0.8			VSS	0.91			P-PO ₄	0.7		
	TSS	0.79			BPA	0.89			BPA	0.67		
	Cl ⁻	0.78			Cl ⁻	0.82						

such as PAEs and BPA, are commonly reported as being truly dissolved and/or attached to colloids (particles < 0.7 mm) (Kalmykova et al., 2014).

4. Conclusions

Currently, at MSWPs, the MBT of residual (mixed) solid waste generates additional liquid by-products streams that are poorly studied. The data obtained in this study show that wastewater generated by MBT units (SUs and CUs), if untreated, potentially release significant amounts of PAEs and BPA, in addition to ammonia and organic matter, to the aquatic environment. These wastewaters contain PAEs and BPA in concentrations usually higher than those in the leachates from MPs and PPs and several times higher than those in municipal wastewaters reported elsewhere. Thus, monitoring data are extremely valuable for understanding the release of PAEs and BPA during MBT processes. Such data will assist in prioritizing chemicals of concern in liquid by-product treatment technology.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.wasman.2017.03.040>.

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