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A modern solid waste management strategy – the generation of new by-products

Sylwia Fudala-Ksiazek [a,*], Mattia Pierpaoli [b], Eliza Kulbat [a], Aneta Luczkiewicz [a]

[a] Faculty of Civil and Environmental Engineering, Gdansk University of Technology, Narutowicza 11/12, 80-233 Gdansk, Poland
[b] Department of Materials, Environmental Sciences and Urban Planning (SIMAU), Università Politecnica delle Marche, Ancona, Italy

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ABSTRACT

To benefit the environment and society, EU legislation has introduced a 'zero waste' strategy, in which waste material should be converted to resources. Such legislation is supported by the solid waste hierarchy concept, which is a set of priorities in waste management. Under this concept, municipal solid waste plants (MSWPs) should be equipped with sorting and recycling facilities, composting/incineration units and landfill prisms for residual bulk disposal. However, each of the aforementioned facilities generates by-products that must be treated. This project focuses on the leachates from landfill prisms, including modern prism (MP) that meet EU requirements and previous prism (PP) that provide for the storage of permitted biodegradable waste as well as technological wastewaters from sorting unit (SU) and composting unit (CU), which are usually overlooked. The physico-chemical parameters of the liquid by-products collected over 38 months were supported by quantitative real-time PCR (qPCR) amplifications of functional genes transcripts and a metagenomic approach that describes the archaeal and bacterial community in the MP. The obtained data show that SU and especially CU generate wastewater that is rich in nutrients, organic matter and heavy metals. Through their on-site pre-treatment and recirculation via landfill prisms, the landfill waste decomposition process may be accelerated because of the introduction of organic matter and greenhouse gas emissions may be increased. These results have been confirmed by the progressive abundance of both archaeal community and the methyl coenzyme M reductase (mcrA) gene. The resulting multivariate data set, supported by a principal component analysis, provides useful information for the design, operation and risk assessment of modern MSWPs.

1. Introduction

EU solid waste legislation has introduced a 'zero waste' strategy designed to promote the extended responsibility of producers and households and stimulate the development of new waste treatment technologies. Considerable effort has been undertaken to emphasize the importance of waste prevention, reuse, recycling and recovery (nutrients and energy), with waste disposal as an option of last resort. The above solid-waste hierarchy concept is supported by Directive 1999/31/EC, which requires European member states to progressively reduce the landfilling of biodegradable waste to 75% of the 1995 level by 2006, 50% of the 1995 level by 2009, and 35% of the 1995 level by 2016. However, Poland, which landfilled more than 80% of its municipal waste in 1995, was given a four-year extension (75% by 2010, 50% by 2013, 35% by 2020), as were several other member states (Bulgaria, Cyprus, Czech Republic, Estonia, Greece, Ireland, Latvia, Lithuania, Malta, Portugal, Romania, Slovakia and the UK).

As a result, significant increases in municipal waste recycling and composting have been observed among all EU members (from 18% in 1995 to 42% in 2012); however, the statistics indicate a widening gap between Central and Eastern EU members (including former Eastern Bloc nations) and Western EU members (Eurostat, 2014). For example, in 2012, 13% of the municipal waste was recycled, 12% was composted, <1% was incinerated and 75% was landfilled in Poland (Eurostat, 2014). Similar trends have been observed in other former Eastern Bloc countries, where the majority of municipal waste is still landfilled.

Because of EU legislation, each municipal solid waste plant (MSWP) should be equipped with sorting and recycling facilities, composting/incineration units and landfill prisms for the residual bulk disposal. However, the by-products generated by the sorting units (SUs) and composting units (CUs) as well as by the modern

* Corresponding author.
E-mail addresses: sksiazek@pg.gda.pl (S. Fudala-Ksiazek), m.pierpaoli@pm.univpm.it (M. Pierpaoli), ekul@pg.gda.pl (E. Kulbat), ansob@pg.gda.pl (A. Luczkiewicz).

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prisms (MPs) are not well understood for the post-2010 operations in Poland, which meet EU requirements and are biochemically influenced by low-organic waste deposition. Such reductions in the disposal of organic matter may limit methane generation by MPs and the potential use of methane in energy production. However, many former Eastern Bloc countries have experience managing MPs and previous prisms (PPs), which were usually ad-hoc arranged without liners or pollution-control systems and unlimited disposal of organic wastes. In terms of landfill leachates, their quality and quantity may depend on several factors, including the raw solid waste composition, the effectiveness of sorting, the composting technology, and the age of the landfill prism. However, landfill leachates generated by PPs are suspected to differ from that generated by MPs. The typical composition of the leachates generated by PPs are provided in Table 1.

Moreover, the introduction of sorting and composting processes at MSWPs generates additional technological wastewaters that are usually neglected. In SUs, the recyclable materials are separated through a combination of manual and mechanical sorting, and the wastewater is generated during polyethylene terephthalate (PET) waste crushing as well as during the washing and cleaning of waste disposal sites, equipment and trucks. In the case of CUs, the wastewater is mainly generated during solid waste biodegradation, which is occasionally supported by irrigation.

Determining the proper (qualitative and quantitative) characteristics of all liquid by-products generated by modern MSWPs is of special concern, particularly when identifying the appropriate on-site or off-site treatment operations and predicting the potential harmful effects of these by-products on the environment.

The aim of this work was to determine the physical and chemical variables of all liquid by-products generated at MSWPs over the course of a long-term (38 months) study. Additionally, a principal component analysis (PCA) was employed to determine the relationships in the obtained data set for the MPs, and a metagenomic approach was used to analyse the bacterial and archaeal succession in selected leachate samples.

2. Methods

2.1. Site description and sampling

In this study, PP and MP landfill leachates as well as SU and CU wastewaters were collected from an MSWP situated in the Pomerania region of northern Poland (Fig. 1). The MSWP serves a metropolitan area of approximately 460,000 people and receives ca. 190,000 Mg of waste per year, of which 130,000 Mg is municipal and 97,000 Mg is biodegradable. In recent years, the studied MSWP has undergone extensive modernization to achieve the mandated reduction in the landfilling of biodegradable waste, reaching 25% and 50% of the 1995 level (63,493 Mg) in 2010 and 2013, respectively. In spring 2010, the sorting and recycling of solid wastes and the composting of their organic fraction were introduced (Fig. 1). However, it should be noted that solid waste segregation (into paper, glass, plastic and mixed) was made a household obligation in the tested area in January 2013.

The studied MSWP is equipped with a municipal solid waste SU with a capacity of 150,000 Mg per year in a three-shift operation. The solid wastes delivered to the MSWP are initially pre-sorted to remove large elements (e.g., furniture, electronics, foil and cardboard) and then directed to the sorting line, where the waste stream is divided into four fractions: the >160 mm fraction is sent directly into the cabin for manual waste sorting together with the 90–160 mm fraction after preliminary air and magnetic separation; the 20–90 mm fraction, which primarily consists of biodegradable components, is sent to an indoor CU; and the 0–20 mm fraction is landfilled. All of the wastewater generated in the sorting process (ca 0.7 m^3/d) was initially directed to the PP; however, since March 2012, it has been directed to the MP (Fig. 1).

The organic wastes are directed to the indoor CU, which was designed for the biological treatment of 30,000 Mg of biodegradable waste per year (75,000 m^3). The main part of the composting hall is a bioreactor, which consists of compost heaps located at nine stations. Each station is equipped with aeration and irrigation systems as well as a temperature prism controlling system. After 28 days, the composted material is transported outside and directed to the yard, where further maturation is conducted for 40 days. The stabilized biodegradable wastes are subjected to sieving to remove non-biodegradable contaminants (diameter > 20 mm), which are intended for landfilling. This stabilized waste is usually sold to external customers or used for the recultivation of old prisms (see below). The wastewater generated during composting (ca. 15 m^3/d) is collected and directed to either the PP (before March 2012) or the MP (after March 2012) (Fig. 1).

Two types of leachate are currently collected from disposed solid waste: one from the MP and one from the PP. The PP was in operation from January 2003 to October 2011 (Fig. 1).

Table 1
Comparative summary of the selected parameters of leachates generated by landfills implemented before the introduction of Council Directive 1999/31/EC (Council of the European Union, 1999).

Type (age) of leachate		Young (<1 year)	Intermediate (1–5 years)	Stabilized (>5 years)
Parameter[a]		Min–max		
pH	–	5.8–6.5	5.6–7.5	7.5–8.1
Conductivity	mS/cm	2.5–35		
BOD_5	mg/dm^3	12,000–24,000	1600	150–260
COD		23,800–6200	4000–10,000	100–4000
BOD_5/COD		0.39–0.5	0.1–0.3	0.04–0.2
N–NH_4		790–1400	1500	230÷841
TP		1–80	1–17	0.5–4.2
Cl^-		150–4500		
SO_4^{2-}		8–7750		
TSS		200–1200	400–2390	100–2980
Zn		0.53–170	0.18–38.0	0.19–0.37
Cr		0.13–8.40	0.12–0.20	0.04–0.23
Cu		0.08–0.30	0.02–0.78	0.03–0.12
Cd		0.02–0.45	0.02–0.08	0.01
Ni		0.42–6.11	0.01–0.02	0.09–0.47
Pb		0.05–1.6	0.02–0.08	0.01–0.14

[a] Ranges are based on Fudala-Ksiazek et al. (2014), Kulikowska and Klimiuk (2008), Renou et al. (2008), Wiszniowski et al. (2007), Alvarez-Vazquez et al. (2004), Christensen et al. (2001), and Lema et al. (1988).

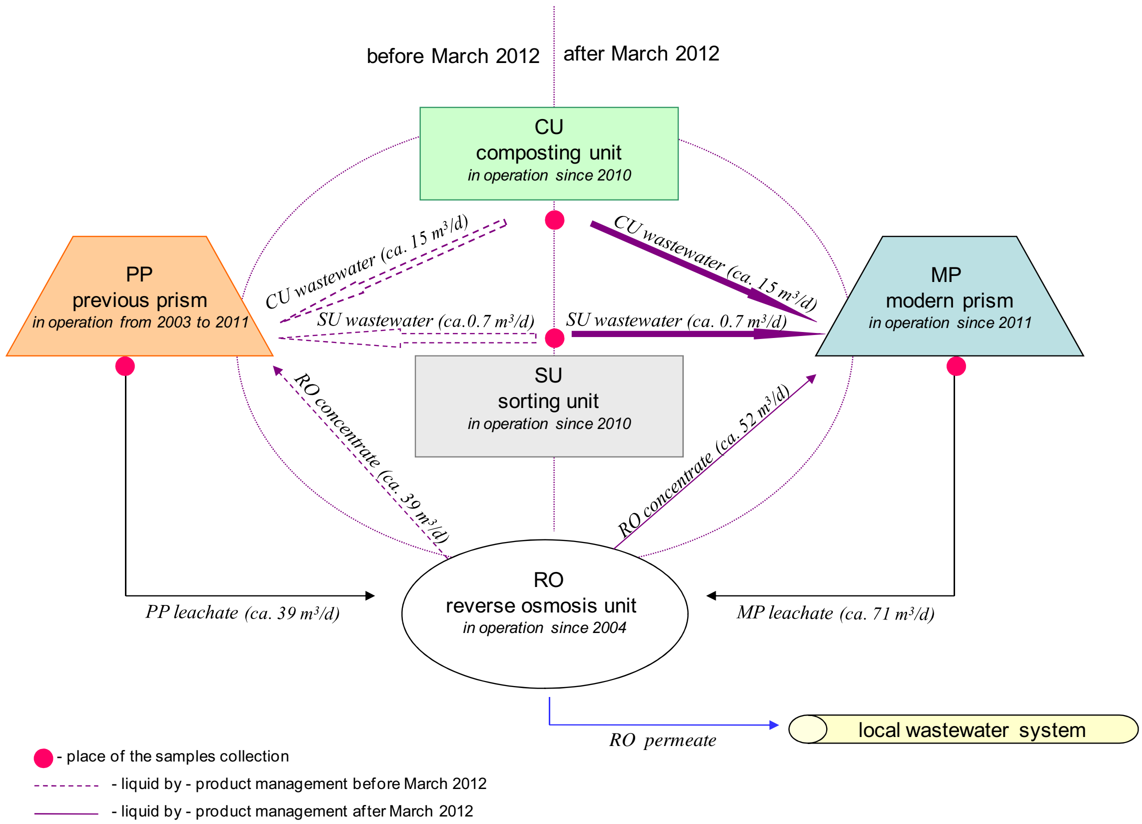

Fig. 1. Waste processing scheme at the studied MSWP, including the sampling location.

Rehabilitation was conducted until September 2015 using the stabilized waste from the CU. The MP has been in operation since November 2011 (for the MP, the operation time is equal to the study period). Both the leachates collected from the PP (ca. 39 m^3/d) and those collected from the MP (ca. 71 m^3/d) are directed to the reverse osmosis (RO) treatment unit (in operation since 2004). The RO permeate is discharged to the local wastewater system, whereas the RO concentrate (ca 52 m^3/d) is pumped back to the prisms. Until May 2012, the concentrate was directed to the PP and then to the MP along with technological wastewater from the SU and CU (Fig. 1).

Generally, the sources of the liquid waste generated by the MSWP have the following contributions: 1% for the SU, 12% for the CU, 31% for the PP, and 56% for the MP. However, these ratios may vary because the amount of leachates generated by landfill prisms is strongly influenced by rainfall; in the studied area, the average annual rainfall is 600 mm/year (Lorenc, 2005).

2.2. Physico-chemical analysis

The landfill leachate (PP and MP) and technological wastewaters (SU and CU) were sampled monthly from November 2011 to January 2015 as 24 h composite samples, kept cool and transported to the laboratory for further analysis.

The following parameters were analysed according to APHA (2005). The pH and conductivity were measured using a portable multi-parameter meter – HQ40D multi (Hach, Germany). The inorganic and total nitrogen compounds (N–NH_4, N–NO_3, N–NO_2, TN), TP, orthophosphate (PO_4–P), chemical oxygen demand (COD), chloride (Cl^-), sulphate (SO_4^{2-}) were measured using a XION 500 spectrophotometer (Dr. Lange, GmbH, Germany). The TSS, MSS and VSS were determined using the gravimetric method, and the biological oxygen demand (BOD5) was analysed using the manometric respirometric BOD OxiTop® method. The total selected heavy metals (Ni, Zn, Cr, Cu, Cd, Pb) was measured by atomic absorption spectrometry with flame atomization using an AAS Vario 6 instrument (Analytik Jena AG, Jena, German) (PN-ISO 8288:2002).

Physico-chemical analyses of leachate and wastewater were supported by meteorological and biogas generation data. The local rainfall and temperature were monitored by the automatic meteorological station that services the MSWP. The studied MSWP was also equipped with a segment for utilizing biogas and a transformer station. Degassing wells ($n = 123$) were built to collect the landfill biogas. Biogas generation monitoring data were obtained from January 2013 to January 2015. The tested MSWP biogas generally contains 50–75% CH_4, 25–45% CO_2, 10–10,000 ppm H_2S and small amounts of N_2, O_2, H_2 and NH_3 and is generated by the PP. However, in November 2014, the MP also started to generate biogas, which was initially of very poor quality and was thus burned in the flare. However, in March 2015, the MP biogas was approximately 50% methane in March 2015 and approximately 70% methane by June of that year. Thus, beginning in late April 2015, biogas from the MP has been extracted at rates of 30–36 m^3 per day (personal communication from the MSWP operator responsible for the landfill gas management).

2.3. Principal components analysis (PCA)

To analyse the physico-chemical parameters from the MP recorded over three years, PCA was conducted. Such an approach allows the reduction of a high-dimension data set of interrelated variables while retaining as much of the variation present in the data set as possible by transforming the obtained data set to a new set of uncorrelated variables (principal components, PCs). These PCs are ordered by the amount of variation explained

(Jolliffe, 2002). In this study, PCA provides a statistical interpretation of several characteristics of the MP leachates simultaneously.

2.3.1. Data preparation

The number of MP parameters and the period length were chosen to maximize the number of parameters included in the time series. In addition to the physico-chemical parameters analysed, temperature and rainfall were included in the analysis.

Before running a PCA, the data set was preliminarily prepared (Vandeginste et al., 1998). First, the outliers were removed by plotting the Mahalanobis distance vs. chi quantiles. The analysis of the CU from 15 January 2015 was excluded because certain concentrations produced values that were an order of magnitude higher than the average and caused by an unknown singular event. Second, a logarithmic transformation was applied. Taking the logarithm of the positively skewed data provides a more symmetrical distribution, smoothing extreme values. Zeros have been substituted by the detection limit. Third, to remove the influence of the measured units and the magnitudes of the values, the data were standardized by subtracting the mean and dividing by the standard deviation (Wold et al., 1987). Missing values (<1%) were substituted by the parameter mean. When performing PCA with heavy metal data, shorter periods were considered owing to a lack of measurements.

2.3.2. PCA implementation and result visualization

Many numerical computing environments are currently available for easy data manipulation. In our case, the function "princomp", available in both MATLAB (http://www.mathworks.com) and GNU Octave (http://octave.sourceforge.net/), has been used to calculate the PC coefficients, PC scores and PC variances.

A particular advantage of reducing the dimensionality of a dataset is that the first two components (PC1 vs. PC2, for instance) can be presented in a 2-dimensional plot, providing a straightforward visual representation of the data. By comparing the score plot (a plot showing sample locations along the new reference system) and the corresponding loadings plot (a plot showing the contributions of variables along the same PCs), the data can be interpreted relatively easily.

The main drawback of this approach is that it is difficult to interpret PCA in a meaningful way, and it is important to consider that correlation is a necessary but not sufficient condition for causality.

Plotting and statistical analyses were conducted using the free software R (http://www.R-project.org/).

2.4. Microbiological analyses

2.4.1. DNA extraction

The total genomic DNA was extracted from 7 MP leachate samples obtained in the 2nd, 5th, 8th, 13th, 19th, 25th and 37th months of the study period using the commercially available kit Sherlock AX (A&A Biotechnology, Poland) with some modifications. These dates where selected after comparing the physico-chemical results (Fig. 2) with the PCA score plots (Figs. 9 and 10). The samples 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 suggestions. The DNA concentrations of samples were determined by an ND-1000 UV–Vis spectrophotometer. The extracted DNA was stored at 4 °C for subsequent studies.

2.4.2. Real-time PCR

Real-time qPCR was performed to analyse the abundances of total bacteria and archaea, sulphate-reducing organisms and methane-producing organisms by searching for 16S rDNA, the *dsr*A gene and the *mcr*A gene, respectively. For the total bacteria and archaea quantification, 10 μM of primer pairs were used: for 16S rDNA, V4-515f/16S V4-806rB (5′-GTG YCA GCM GCC GCG GTA A-3′, 5′-GGA CTA CNV GGG TWT CTA AT-3′) (Caporaso et al., 2012); for the *dsr*A gene, DSR1F/RH3-dsr-R (5′-ACS CAC TGG AAG CAC G-3′, 5′-GGT GGA GCC GTG CAT GTT-3′) (Ben-Dov et al., 2007); and for the *mcr*A gene, mcrA-F/mcrA-R (5′-GGT GGT GTM GGA TTC ACA CAR TAY GCW ACA GC-3′ and 5′-TTC ATT GCR TAG TTW GGR TAG TT-3′) (Denman et al., 2007). The PCR reaction consisted of 10 μl of Real Time 2× HS-PCR Mix SYBR A (A&A Biotechnology, Poland), 1 μl of each forward and reverse primers (2 μl in the case of *mcr*A), and 2 μl of DNA template (1 ng/μl), with sterile double distilled water added to achieve a total volume of 20 μl. The initial denaturation lasted 3 min at 95 °C and was followed by 40 cycles of the following incubation cycle: denaturation for 15 s at 95 °C, primer annealing for 30 s at 55 °C (61 °C for *dsr*A and *mcr*A), and then product elongation for 30 s at 72 °C. SYBR Green fluorescence detection was performed at the end of each primer annealing step. Amplicon specificity was confirmed via melting curve analysis of the qPCR end products. Each run included a no-template control, and the reproducibility of the SYBR Green qPCR was assessed by running samples independently in triplicate. Analysis was performed using an Mx300P thermocycler (Stratagene).

The results were expressed as a ratio of the obtained *dsrA* and *mcrA* copy genes to the 16S rDNA copy genes and calculated as $2^{\Delta Ct}$, where $\Delta Ct = Ct_{16S} - Ct_{dsrA}$ (or $\Delta Ct = Ct_{16S} - Ct_{mcrrA}$) and Ct is the cycle threshold, assuming that the reaction yield is close to 100%.

2.4.3. Next-generation sequencing

In this study, the microbial community present in the MP samples mentioned in Section 2.4.1 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′-GTGCCAGCMGCCGCGG TAA-3; 5′-GGAC TACHVGGGTWTCTAAT-3′). Data were analysed using the 16S Metagenomics App in the BaseSpace® analysis environment.

3. Results and discussion

In this research, monthly samples of the MP and PP leachates and the SU- and CU-generated wastewaters were used as sensitive indicators of the processes in the MSWP. To enable observation of long-term changes (38 months), physical and chemical analyses were regularly conducted over three years. For the MP, the obtained multivariate data set was also tested by a PCA and supported by metagenomic analyses of bacterial and archaeal succession as well as the presence of *dsrA* and *mcrA* genes, indicators of the MP's biochemistry.

3.1. Physico-chemical results

In the first month of the MP's operation, the COD and BOD_5 values were low, at 31.1 mg O_2/dm^3 and 3.4 mg O_2/dm^3, respectively. The similar trends were then observed for the COD and BOD_5 values, and the results indicated a gradual increase for up to 5–6 months of operation, with values reaching 12,620 mg O_2/dm^3 and 8,185 mg O_2/dm^3, respectively, followed by stabilization after 33–34 months of operation, with stationary concentrations reaching approximately 1600 mg O_2/dm^3 and 400 mg O_2/dm^3, respectively (Fig. 2). The final concentrations of BOD_5 in the MP leachate were comparable to those in the PP leachate (423 ± 124 mg O_2/dm^3). In the case of COD, the final value for the

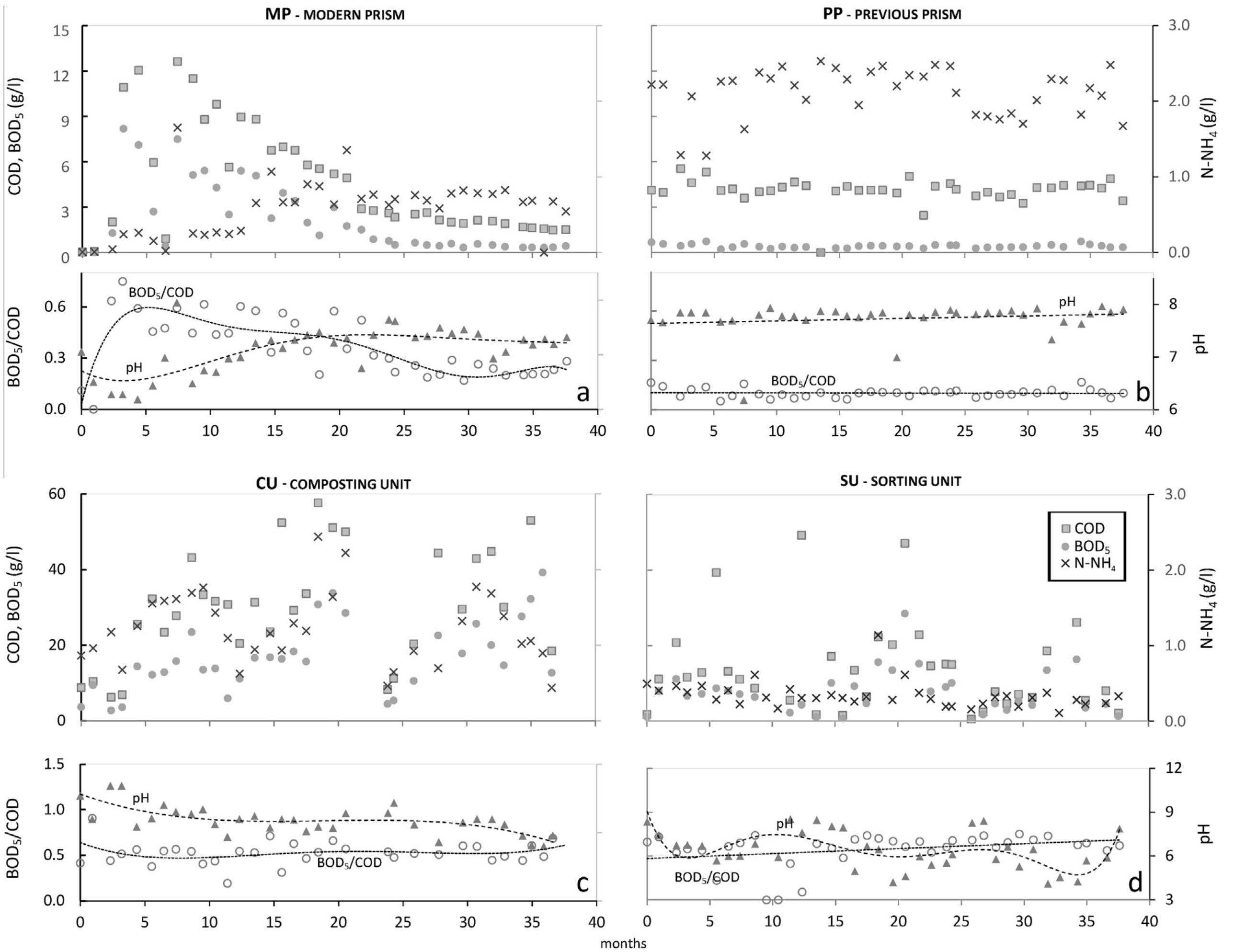

Fig. 2. Temporal evolution of the leachate generated by (a) a modern prism (MP), (b) a previous prism (PP), (c) a composting unit (CU) and (d) a sorting unit (SU) in terms of COD, BOD_5, pH, BOD_5/COD ratio and ammonia nitrogen ($N–NH_4$) concentration.

MP was approximately 2.5 times lower than the average COD in the PP leachate (4193 ± 540 mg O_2/dm^3) (Figs. 2 and 3) and was generally lower than the literature values (Table 1).

The PP, which operated from January 2003 to October 2011, received up to 90% of indistinct municipal waste. Thus, the deposition of highly biodegradable waste, as well as PP's period of operation, explains not only the aforementioned high COD and low BOD_5 concentrations but also relatively high and stable ammonia concentration (2114 ± 323 mg $N–NH_4/dm^3$) (Figs. 2 and 3). Ammonia is formed in landfill prisms during the decomposition of nitrogenous organic matter (mainly proteins) (Burton and Watson-Craik, 1998; Robinson, 1995) and is easily released with leachate, because its microbiological utilization is limited under low oxygen availability. In the studied MP, the ammonia concentration gradually but continuously increased for up to 34 months of operation (from 6 mg $N–NH_4/dm^3$ to 902 mg $N–NH_4/dm^3$). Meanwhile, during the subsequent, and final, five months of operation, a decreasing trend was observed (Fig. 2). In the case of the PP, the ammonia concentration was much higher and fluctuated around a mean value of 2324 ± 379 mg $N–NH_4/dm^3$ (Figs. 2 and 3). In both studied prisms, the ammonia constituted a major fraction of the total nitrogen (85% ± 11% of TN in MP and 91% ± 7% of TN in PP). In addition to ammonia, nitrate and nitrite, other inorganic forms of nitrogen, were also observed in the MP and PP but in negligible amounts (not exceeding 1% and 0.1%, respectively) (Fig. 3).

It should be noted that the SU and CU wastewaters were also rich in ammonia and organic nitrogen (Figs. 2 and 3). The ammonia concentrations were 344 ± 174 mg $N–NH_4/dm^3$ in the SU and 1223 ± 443 mg $N–NH_4/dm^3$ in the CU. For N_{org}, its concentration in the SU varied from 8 mg N_{org}/dm^3 to 555 mg N_{org}/dm^3 (av. 151 mg N_{org}/dm^3), whereas its concentration in the CU reached up to 2560 N_{org}/dm^3 (extreme value not shown in Fig. 3 for scale reasons), with an average of 364 mg N_{org}/dm^3. This high load of nitrogen compounds is introduced to the MP in the return stream of wastewater from the SU and CU and serves as an additional immediate and delayed (due to organic matter decomposition) source of ammonia. In Fig. 2, the temporal trends of wastewater quality can be seen for the CU and SU. In these units, the higher COD and BOD_5 values as well as the $N–NH_4$ and N_{org} concentrations (Figs. 2 and 3) are likely related to the seasonal dietary changes and increased production of garden waste. Thus, to better understand the transformations of carbonaceous and nitrogenous compounds and predict their biodegradation and release from modern prisms over time, the long-term monitoring of leachate compositions is required.

Phosphorus was detected mainly in mineral form ($P–PO_4$), especially in the PP and SU, where it constituted 76% ± 18% and 73% ± 17% of the TP, respectively (Fig. 3). In leachates generated by the MP, $P–PO_4$ constituted up to 80% of the TP, with an average of 57% ± 23%. The organic form of phosphorus was only predominant in the CU wastewater, and in average was

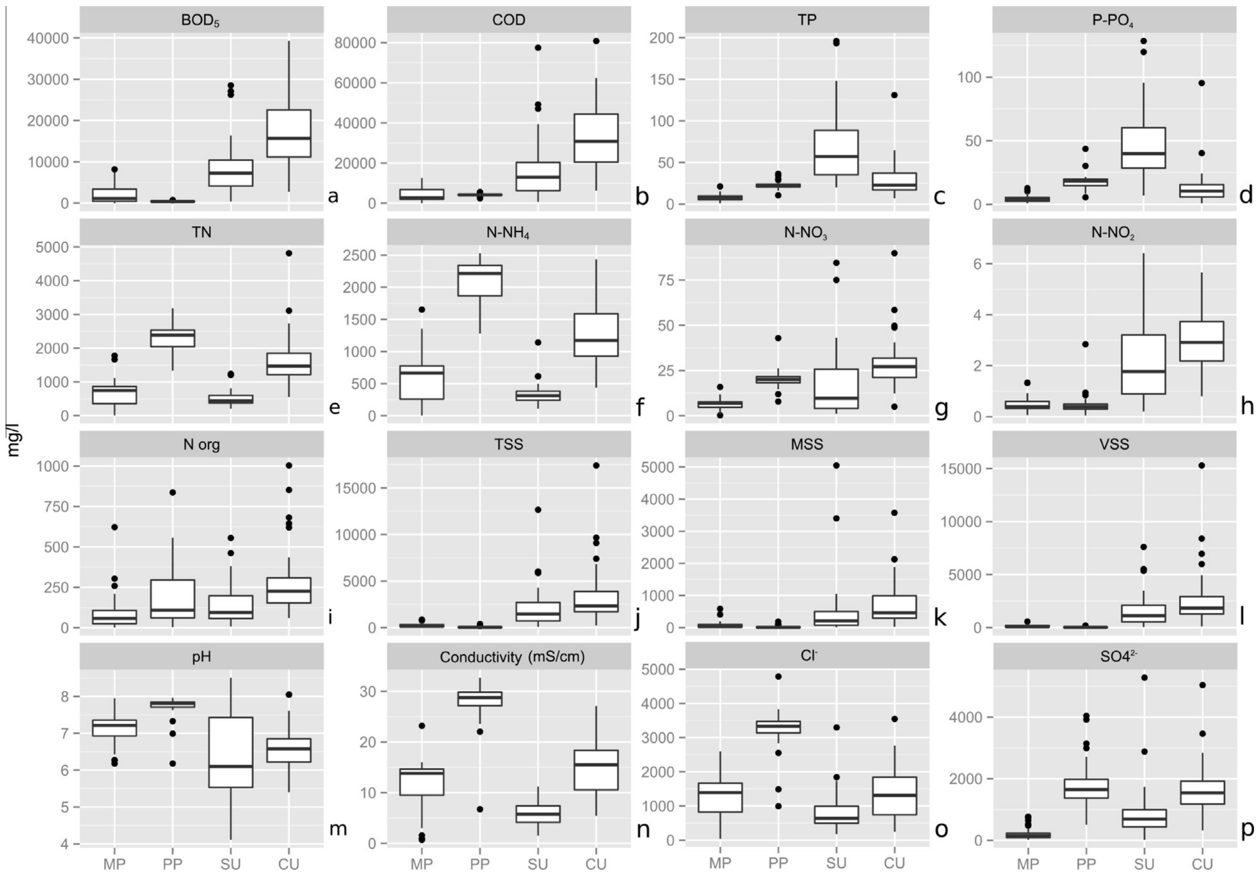

Fig. 3. Minimum, maximum, upper quartile, lower quartile, and median values of (a) BOD_5, (b) COD, (c) TP, (d) $P–PO_4^{3-}$, (e) TN, (f) $N–NH_4$, (g) $N–NO_3^-$, (h) $N–NO_2^-$, (i) N_{org}, (j) TSS, (k) MSS, (l) VSS, (m) pH, (n) conductivity, (o) Cl^-, and (p) SO_4^{2-} in the leachates and wastewaters generated by the modern (MP) and previous (PP) prisms and the sorting (SU) and composting (CU) units.

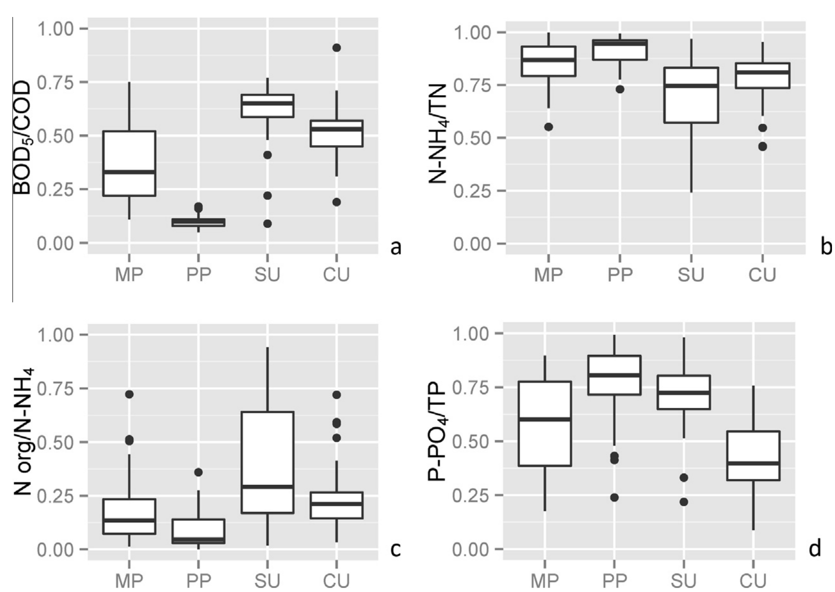

Fig. 4. Minimum, maximum, upper quartile, lower quartile, median values of (a) BOD_5/COD, (b) $N–NH_4$/TN, (c) N_{org}/$N–NH_4$, and (d) $P–PO_4$/TP ratios for the leachates generated by the modern (MP) and previous (PP) prisms and the sorting (SU) and composting (CU) units.

equivalent to 58% ± 14% of the TP. Nevertheless, both phosphorus forms (TP and $P–PO_4$) showed high variability, especially in the SU and CU, for which this variability results from seasonal changes. In the case of MP leachate, the TP and $P–PO_4$ concentrations showed an increasing trend for up to 10 months of operation, followed by a decreasing trend, with occasional maximums across the obtained data set. On average, the TP was equal to 8.0 ± 4.2 mg P/dm^3 in the MP, 23.2 ± 5.7 mg P/dm^3 in the PP, 68.2 ± 44.2 mg P/dm^3 in the SU and 28.8 ± 22.6 mg P/dm^3 in the CU (Fig. 3).

It has been suggested that the high concentration of ammonia, which is also the main form of nitrogen released with the leachate, and the low BOD_5/COD ratio are characteristic of the anaerobic phase of the landfill prism. In The BOD_5/COD ratio in the PP leachate was close to 0.1 throughout the study, whereas its average ratio in the MP leachate was 0.37 (Fig. 4). For the MP, the BOD_5/COD ratio increased up to 0.74 in the fifth month, stabilizing at 0.51 ± 0.9, and then started to decrease, stabilizing at approximately 0.2 after 25 months. A low BOD_5/COD ratio indicates that in the MP and PP, biodegradable organic matter is efficiently consumed in situ by the consortia of microorganisms or has been already been depleted. In the case of the $N_{org}/N{-}NH_4$ ratio, the lowest value was observed in the leachate generated by the PP. The results obtained for the PP together with the stable quality and quantity of its generated landfill gas (see Materials and Methods) suggest that this prism undergoes a methanogenic phase, and similar landfill biochemistry was observed by Spagni et al. (2008). In terms of the MP, it is difficult to draw conclusions based only on physico-chemical data.

In addition to the carbonaceous and nitrogenous-related pollutants found in the liquid by-products generated at MSWPs, the presence of inorganic elements, such as chloride (Cl^-), sulphate (SO_4^{2-}) and heavy metals, is of great importance. Fig. 5 shows the correlation between cumulative monthly rainfall and chloride concentrations in the studied MP and PP leachate. In the initial period (up to 16 months of operation), the Cl^- concentration in the MP leachate was directly proportional to the rainfall depth because of the leaching effect. Subsequently, the tracer concentration tended to increase up to 2198 mg Cl^-/dm^3 and was negligibly affected by rainfall. This observation can be explained by the on-site pretreatment of wastewaters generated by the SU and CU, which, along with the concentrate generated by RO units, are pumped back to the MP (Fig. 1). The SU and CU wastewaters and RO concentrates are rich in chlorides, with values equalling 851 ± 615 mg Cl^-/dm^3, 1423 ± 792 mg Cl^-/dm^3, and 3746 ± 1544 mg Cl^-/dm^3, respectively. Compared with organic substances, monovalent ions, such as Cl^-, are barely or not retained in the landfill prisms. Thus, the chemical composition of the leachate may be affected by the re-concentration of Cl^-. Such re-concentrations may influence the biodegradation of landfill prisms and result in the need for greater quantities of chemical compounds (sulphuric acid and/or antiscaling products) to run the RO process.

3.2. Heavy metals

Inside the landfill prism, the mobility of heavy metals depends on a number of factors, including the form in which they are present in the solid waste. However, it has been suggested that the metal solubility is low in anaerobic stages of waste decomposition with a pH near or above neutral, (Flyhammar et al., 1998; Huber et al., 2004). Additionally, it was calculated that deposited organic matter and sulphides have a significant capacity to bind metal ions at neutral or slightly alkaline pH (Ostman et al., 2008; van Praagh et al., 2007).

Until recently, most studies have focused on landfills managed using traditional processes with high organic content deposition (Kamaruddin et al., 2015; Kjeldsen et al., 2002; Kulikowska and Klimiuk, 2008; Rowe, 1995; Salem et al., 2008). In landfills with a limited amount of organic matter, heavy metal leaching is expected to be mainly controlled by the solubility of these metals. In this study, the total amounts of heavy metals in the PP and MP were comparable in terms of the Zn, Pb, Cu and Cd concentrations. However, the average contents of Ni and Cr in the MP leachate were approximately 5 times lower than those in the PP leachate, whereas the average contents of Zn and Cu were seven and five times higher in the MP leachate, respectively (Fig. 6). The average concentrations of heavy metals tested in the MP and PP leachates were generally lower than those found in typical municipal wastewater (Henze et al., 2008). Although both prisms had similar pH values (Figs. 2 and 3), the long-term metal-binding potential of the MP is questionable because broad parameters, such as those tested in this study (COD, BOD, pH, etc.), cannot predict this potential. A further question remains regarding whether the re-concentration of wastewater from the SU and CU may serve as an additional source of heavy metals, as mentioned above, and/or influence the mobility of heavy metal deposits within the solid waste structure. According to the obtained results, both the SU and CU are important sources of heavy metals (Fig. 6), especially Zn and Cu. The wastewater generated by the CU seems to be especially rich in Ni, Pb and Cd (1.047 ± 0.683 mg Ni/dm^3, 0.207 ± 0.189 mg Pb/dm^3, 0.019 ± 0.010 mg Cd/dm^3, respectively). In the literature, there is a lack of information on the presence of heavy metals in wastewater generated by SUs and CUs.

3.3. Microbiological results

A major concern in modern landfill prisms is their microbiology and the question of by-product generation, especially the effectiveness of methanogenesis under limited availability of biodegradable organic matter. Thus, in this study, seven samples of MP leachate (from the 2nd, 5th, 8th, 13th, 19th, 25th and 37th month of MP operation) were further subjected to next-generation sequencing and q-PCR searching for the key functional genes involved in the methanogenesis (*mcrA*) and dissimilatory sulphate reduction (*dsrA*) processes.

The analysis of 16S rDNA provided information on the bacterial and archaeal community in the tested samples. According to the obtained results, bacteria were predominant in the leachate during

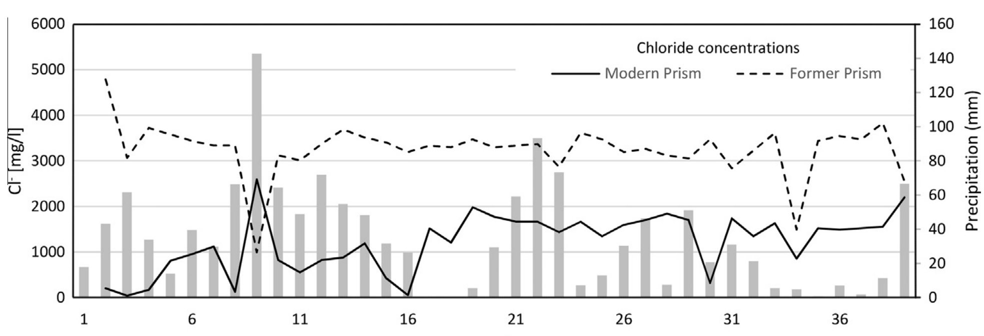

Fig. 5. Cumulative rainfall and chloride concentrations in the leachate generated by the modern prisms (MP) (continuous line) and previous prisms (PP) (dotted line). The horizontal axis indicates the number of months after the landfill opening. The left vertical axis indicates the concentration of chloride ions (black line), and the right vertical axis indicates the depth of monthly cumulative rainfall (grey bars).

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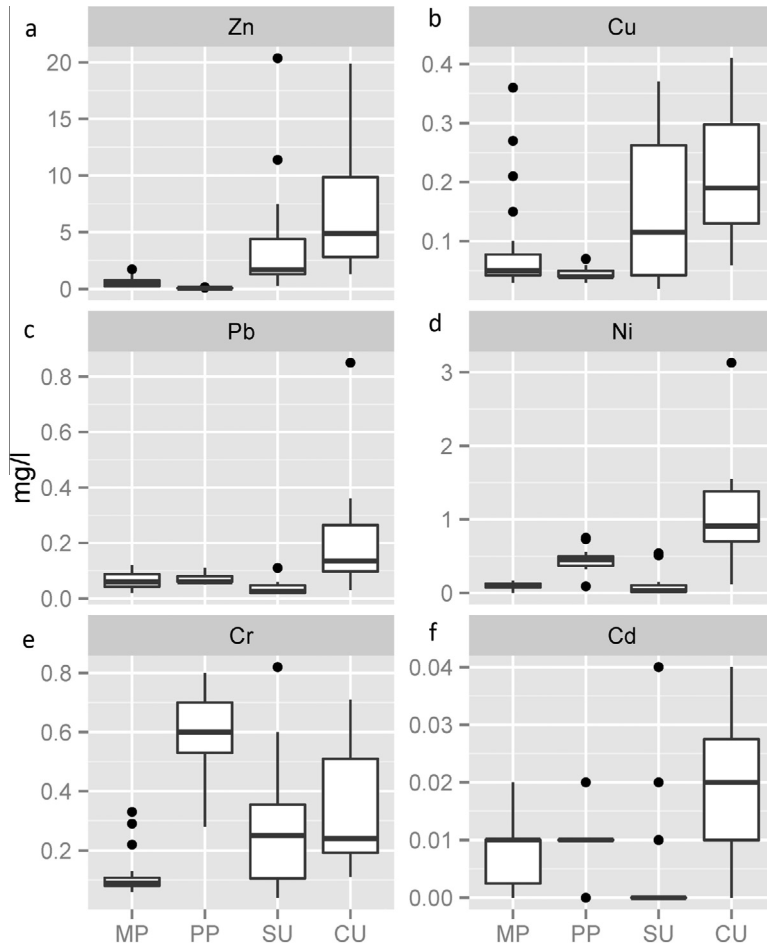

Fig. 6. Heavy metal concentrations of (a) Cd, (b) Cr, (c) Cu, (d) Ni, (e) Pb, and (f) Zn found in the leachates generated by modern prisms (MP) and previous prisms (PP) and the sorting (SU) and composting (CU) units.

the 37-month study period for the MP operation, whereas the relative abundance of archaea was below 0.5% of the total reads during the first 8 months and, at the end of study, started to increase, reaching 23.08% (Fig. 7). For the domain level, more than 99% of the total reads were classified; however, for lower taxonomic levels, the classification rate dropped (see Fig. S1).

The archaeal community mainly consisted of *Euryarchaeota* phylum (from 0.42% at the beginning to 23.04% of total reads in the last month of the study), with incidental occurrence of *Crenarchaeota* (<0.05% of the total reads in the tested samples). A large number of *Euryarchaeota* produces methane (CH_4) in anaerobic conditions. In this study, the phylum *Euryarchaeota* was represented by two classes, both methanogens: the predominant *Methanomicrobia* (from 70% to 99% of *Euryarchaeota*-linked sequences) and the less frequent *Methanobacteria* (Fig. 7, Table 2). *Methanomicrobia* was represented by 5 families of the order *Methanomicrobiales* (*Methanocalculus*, *Methanoregulaceae*, *Methanocorpusculaceae*, *Methanospirillaceae*, *Methanomicrobiaceae*) and 2 families of the order *Methanosarcinales* (*Methanosaetaceae* and *Methanosarcinaceae*) (Table 2). Similar results for the methanogen community at the order level were observed in the laboratory-scale reactors, simulating the MSWP bioreactors, where methanogens affiliated with *Methanomicrobiales* were consistently the most abundant (Bareither et al., 2013). In this study, for each tested MP leachate sample, the sequences affiliated with methanogens were also represented mainly by the order *Methanomicrobiales*, family *Methanocorpusculaceae* (up to 95% and up to 91% of methanogens reads, respectively). The family *Methanocorpusculaceae* presented increasing abundance over time, from 0.07% of the total reads at the beginning to 18.06% of the total reads at the end of observation (Table 2). *Methanocorpusculaceae* contains only one genus, *Methanocorpusculum*, of very small, irregular cocci, characterized by complex nutritional requirements. Substrates for methanogenesis are frequently formate and sometimes alcohols, but methane can also be produced via the hydrogenotrophic pathway ($4H_2 + CO_2 = CH_4 + 2H_2O$) (Whitman et al., 2006).

In addition to the order *Methanomicrobiales*, and increasing abundance was also noticed for the order *Methanosarcinales*, but it did not exceed 2.5% of the total reads (Table 2). The *Methanosarcinales* was represented by two families, *Methanosaetaceae* and *Methanosarcinaceae*, and three genera, *Methanosaeta*, *Methanimicrococcus* and *Methanosarcina*. As a substrate for methanogenesis, *Methanosaeta* uses acetate, and *Methanimicrococcus* uses methylamines, whereas *Methanosarcina* can produce methane using all three known methanogenesis pathways (methylamines, acetate and hydrogenotrophic) (Whitman et al., 2006). The incidental occurrence of acetoclastic methanogens (e.g., *Methanosaetaceae*) in this study (Table 2) can be explained by their inhibition by high ammonium concentrations, as suggested by Karakashev et al. (2005). In MP leachate, the ammonia concentration gradually increased during the MP operation. Archaeal sequences, particularly those affiliated with the family *Methanosaetaceae*, were observed by Mori et al. (2003) in landfill leachate obtained from a prism filled with wastes mainly consisting of incineration ash.

In this study, the increasing abundance of methanogens is correlated with the increased relative abundance of the *mcrA* copy

Fig. 7. Classification of NGS results by taxonomic level.

gene, expressed as a ratio to the 16S rDNA copy genes (Fig. 8). *mcrA* is ubiquitous among known methanogens and was used in several studies as a biomarker for the effectiveness of methanogenesis (Morris et al., 2014). Methanogens, as obligate anaerobes, require a redox potential of less than −300 mV for growth; otherwise, denitrifying, sulphate-reducing or iron-reducing bacteria may

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Table 2
Methanogens of the *Euryarchaeota* phylum detected during MP operations.

Months of MP operations	2	5	8	13	19	25	37
Domain							
Archaea	0.46%	0.26%	0.41%	2.36%	6.34%	7.98%	23.08%
Phylum							
Euryarchaeota	0.42%	0.26%	0.41%	2.35%	6.34%	7.98%	23.04%
Class							
Methanomicrobia	0.33%	0.21%	0.39%	1.62%	6.26%	7.94%	23.00%
Methanobacteria	0.09%	0.04%	0.01%	0.73%	0.08%	0.04%	0.03%
Sum	0.42%	0.26%	0.41%	2.35%	6.34%	7.98%	23.03%
Order							
Methanomicrobiales	0.31%	0.18%	0.38%	1.43%	6.17%	6.75%	20.82%
Methanosarcinales	0.02%	0.03%	0.01%	0.19%	0.09%	1.18%	2.17%
Methanobacteriales	0.09%	0.04%	0.01%	0.73%	0.08%	0.04%	0.03%
Sum	0.42%	0.26%	0.41%	2.35%	6.34%	7.98%	23.02%
Family							
Methanocorpusculaceae	0.07%	0.20%	0.37%	1.29%	5.86%	6.25%	18.06%
Methanomicrobiaceae	0.04%	0.02%	0.00%	0.11%	0.24%	0.46%	2.38%
Methanoregulaceae	0.09%	0.01%	0.01%	0.01%	0.05%	0.03%	0.33%
Methanospirillaceae	0.11%	0.00%	0.00%	0.01%	0.01%	0.01%	0.02%
Methanocalculus	0.00%	0.00%	0.00%	0.00%	0.00%	0.01%	0.07%
Methanosarcinaceae	0.02%	0.03%	0.01%	0.19%	0.08%	1.06%	0.87%
Methanosaetaceae	0.01%	0.00%	0.00%	0.00%	0.00%	0.12%	1.29%
Methanobacteriaceae	0.09%	0.04%	0.01%	0.73%	0.08%	0.04%	0.03%
Sum	0.32%	0.25%	0.39%	1.62%	6.25%	7.94%	23.03%
Genus							
Methanocorpusculum	0.07%	0.16%	0.37%	1.29%	5.86%	6.24%	17.98%
Methanoculleus	0.00%	0.01%	0.00%	0.08%	0.10%	0.16%	1.35%
Methanogenium	0.02%	0.00%	0.00%	0.00%	0.02%	0.22%	0.65%
Methanoplanus	0.00%	0.00%	0.00%	0.01%	0.09%	0.03%	0.20%
Methanofollis	0.01%	0.01%	0.00%	0.02%	0.02%	0.04%	0.16%
Methanospirillum	0.11%	0.00%	0.00%	0.01%	0.01%	0.01%	0.02%
Methanocalculus	0.00%	0.00%	0.00%	0.00%	0.00%	0.01%	0.07%
Methanolinea	0.00%	0.00%	0.01%	0.01%	0.00%	0.00%	0.01%
Methanosarcina	0.02%	0.03%	0.01%	0.19%	0.07%	1.05%	0.85%
Methanosaeta	0.01%	0.00%	0.00%	0.00%	0.00%	0.12%	1.29%
Methanimicrococcus	0.00%	0.00%	0.00%	0.00%	0.01%	0.00%	0.02%
Methanobacterium	0.09%	0.02%	0.00%	0.19%	0.04%	0.02%	0.01%
Methanobrevibacter	0.00%	0.02%	0.01%	0.54%	0.04%	0.02%	0.01%
Methanosphaera	0.00%	0.00%	0.00%	0.01%	0.00%	0.00%	0.01%
Sum	0.32%	0.25%	0.40%	2.35%	6.28%	7.94%	22.63%

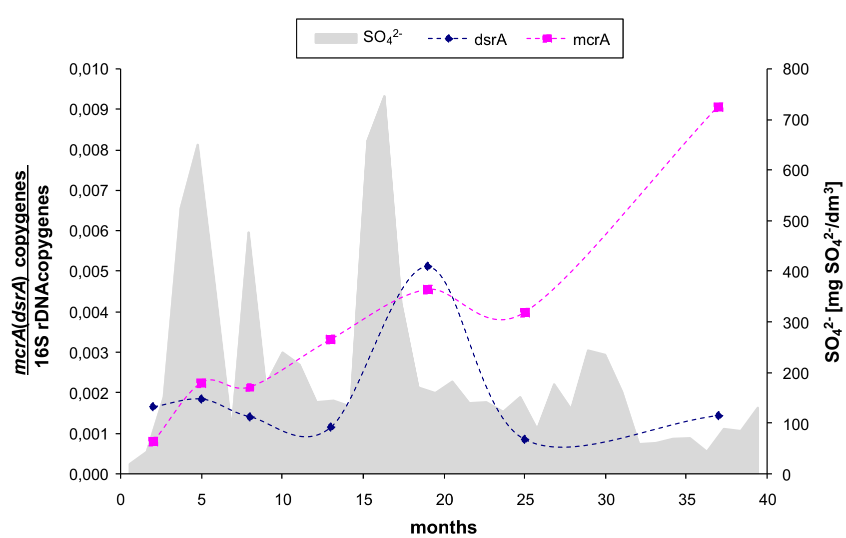

Fig. 8. Relative abundance of the mcrA and dsrA copy genes expressed as a ratio with the 16S rDNA copy genes.

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outcompete them. Thus, their presence is limited when electron acceptors other than CO_2 are present (O_2, NO_3^-, Fe^{3+}, and SO_4^{2-}). Fig. 8 shows the relative abundance of the *mcrA* copy gene as well as the SO_4^{2-} concentration.

For methanogens, the synthesis of methane is the major source of energy; however, they are only able to utilize a small range of substrates: CO_2/H_2, acetate, and methylated C1 compounds (Whitman et al., 2006). Therefore, in the landfill prism, methane production depends on the waste composition and complex degradation processes conducted by a variety of microbial consortia, including hydrolytic bacteria (producers of extracellular enzymes from the hydrolases group), acidifying bacteria, and acetate-forming and hydrogen-producing bacteria. The final products of these transformations are used by the methanogens. In this study, besides the methanogenic phylum *Euryarchaeota* described above, the bacterial clones were affiliated with 29 phyla, mainly *Proteobacteria*, *Bacteroidetes*, and *Firmicutes*, followed by *Actinobacteria*, *Cyanobacteria*, *Fibrobacteres*, *Spirochaetes*, *Synergistetes*, *Tenericutes*, *Verrucomicrobia* and others, whose relative abundance did not exceed 1% in the tested samples of MP leachate (Fig. 7). Similar phyla were detected in reactors treating organic household waste, with *Firmicutes*, *Proteobacteria*, and *Bacteroidetes* being again predominant (Cardinali-Rezende et al., 2009).

In this study, the *Proteobacteria* were the most abundant at the beginning of the MP operation (73.08% of the total reads) and were represented mainly by β-proteobacteria, whereas the presence of other classes (α-proteobacteria, γ-proteobacteria, δ-proteobacteria, ε-proteobacteria) did not exceed 10% of the total reads. Interestingly, at the beginning of the MP operation, the β-proteobacteria were mainly represented by the families *Comamonadaceae* and *Rhodocyclaceae* (21.9% and 11.2% of total reads), characterized by versatile (aerobic or denitrifying) metabolic capabilities. Later, their abundance as well as the abundance of β-proteobacteria did not exceed 1% of the total reads. During MP operation, the abundance of *Proteobacteria* also decreased, stabilizing after 19 months at 11.4–14.9% of total reads. In addition to β-proteobacteria, decreases in the relative numbers of α-, γ- and ε-proteobacteria were noticed, for each class from approximately 7% of total reads at the beginning to less than 2.5%, 1.6% and 0.4%, respectively, after 37 months of MP operation.

Among the *Proteobacteria*, the δ-proteobacteria were the most stable in number and were mainly represented by the sulphate-reducing bacteria from the families *Desulphobulbaceae* and *Desulphobacteraceae* (from 0.3% to 4.2% and from 0.01% to 2.15% of total reads, respectively). In addition to δ-proteobacteria, sulphate-reducing bacteria were also represented by *Desulphosporosinus*, *Desulphurispora*, and *Desulphotomaculum* from the class *Clostridia* of the phylum *Firmicutes*, but their abundance did not exceed 0.8% of total reads in the tested samples. Additionally, incidental (0.02% of total reads) occurrences of sulphate-reducing bacteria from the phyla *Thermodesulphobacteria* and *Nitrospirae* were detected in the MP leachates, but sulphate-reducing archaea were not found.

In this study, the dissimilar sulphate processes were also analysed by the presence of *dsrA*, a key gene involved in the sulphate respiration pathway. According to the obtained results, *dsrA* was detected in each tested sample, and the ratio of *dsrA* copy genes to 16SrDNA copy genes showed a trend similar to the SO_4^{2-} concentration in the MP leachate (Fig. 8). The decrease in *dsrA* abundance observed after 19 months of MP operation was in contrast to the almost steady presence of sulphate-reducing bacteria noted after 13 months of MP operation (5.54% ± 0.3% of total reads). However, the obtained results are consistent with the suggestion that sulphate-reducing bacteria may exhibit high metabolic flexibility depending on the environmental conditions (Muyzer and Stams, 2008). This is especially important in low-redox niches, such as landfill prisms, where sulphate-reducing bacteria and methanogens form consortia that can compete or grow syntrophically, depending on the availability of sulphate.

In the tested MP leachate, in addition to *Proteobacteria*, the other most abundant phyla were represented by members of *Firmicutes* and *Bacteroidetes*, which are common in anaerobic natural and artificial niches rich in organic matter due to their ability to ferment complex polysaccharides (Fig. 7).

The *Firmicutes* were represented mainly by the classes *Clostridia* (from 5% to 42% of total reads) and *Bacilli* (from 1% to 9% of total reads), whereas the other classes did not exceed 1% of total reads (Fig. 7). The most abundant *Clostridia* are known to produce a variety of extracellular enzymes capable of degrading large organic molecules, such as cellulose, lipids, and proteins. The same species of the genus *Clostridium* are important cellulose digesters (Schwarz, 2001). Detected in this study, *C. alkalicellulosi* and *C. termitidis* can utilize cellulose and a variety of sugars as a sole carbon source, whereas *C. caenicola* can utilize the cellobiose that is released during cellulose hydrolysis (Schwarz, 2001).

The third most abundant phylum was *Bacteroidetes* (from 7.1% to 49.2% of total reads), represented by three classes, mainly *Bacteroidia* (from 0.5% to 36.6% of total reads) and *Sphingobacteriia* (from 4.8% to 10.8% of total reads), with *Flavobacteriia* being in the minority (from 0.5% to 3.0% of total reads). *Bacteroidetes*-like sequences were also detected in landfill leachates obtained from laboratory-scale (Bareither et al., 2013) and field-scale bioreactors (Cardinali-Rezende et al., 2009; Huang et al., 2004). *Bacteroides*, similarly to *Clostridia*, secrete proteases to hydrolyse proteins to the amino acids, which are further degraded to fatty acetate, propionate, butyrate, and ammonia. Klocke et al. (2008) suggested the phylum *Bacteroidetes* to be a part of microbial biogas consortia with *Bacteroides* detected as a predominant genus. In this study, however, *Bacteroidetes* were represented by members of three families: *Bacteroidaceae* (from 0.1% to 6.8% of total reads), *Porphyromonadaceae* (from 0.1% to 7.2% of total reads), and *Prevotellaceae* (from undetected to 24% of total reads). Interestingly, the highest number of sequences affiliated with the above-mentioned families was noted 5 months after the MP operation, e.g., 24% for *Prevotella*-like sequences, 6.8% for *Bacteroides*-like sequences, 2.5% for *Parabacteroides*-like sequences, 1.8% for *Porphyromonas*-like sequences and 1% for *Dysgonomonas*-like sequences. The number of total reads affiliated with the *Bacteroides–Parabacteroides–Dysgonomonas–Prevotella* consortium, even in the following months, accounted for 2.5% to 11.9%. All genera are characterized by the ability to metabolize complex organic matter; thus, their growth was probably supported by a high organic load that was introduced to the MP by the wastewater generated by the SU and CU (see Fig. 1 for details). A similar tendency was detected among other members of the phylum *Bacteroidetes*: *Sphingobacterium* and *Pedobacter* from the family *Sphingobacteriaceae* as well as *Flavobacterium* from the family *Flavobacteriaceae*.

It should be noted that the MP leachate sampled after the 5th month of MP operation contained a relatively high number of total reads affiliated with *Fibrobacter* (up to 8.2%), a highly cellulolytic genus from the phylum *Fibrobacteres*, as well as *Treponema* (up to 1.6%) and *Sphaerochaeta* (up to 3.0%), carbohydrate-fermenting genera from the phylum *Spirochaetes*.

In the tested MP leachate, some sequences of a metagenomic dataset remained unidentified (Fig. S1), indicating that the anaerobic degradation processes in landfill prisms may be conducted by unknown species. Nevertheless, the next-generation sequencing and q-PCR data suggest that the modern landfill prisms are still inhabited by a complex microbial community and that the collaborative consortia are efficient in the degradation of deposited organic compounds, despite their limited availability and biodegradability.

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3.4. PCA results

A PCA was performed for the data matrix obtained for the MP, which was composed of 17 parameters. To identify the relations within the multivariate data set, the loading plots and score plots were analysed.

3.4.1. Waste descriptor and recursive clusters

The contribution of the first two PCs to the total variance was 64.7%. The loadings show that the first PC (PC1) has a clear positive relationship with almost every other component, being similar to a weighting function of the leachate characteristics (Fig. 9). For this reason, it is suspected that PC1 is representative of the leachate composition. The loading plots show the recursive groups of the parameters, such as suspended solids (MSS, TSS, VSS), nitrates and nitrites (N–NO_3, N–NO_2), ammonia and total nitrogen (TN, N–NH_4), orthophosphates and total phosphorous (P–PO_4, TP) and biological and chemical oxygen demands (BOD_5 and COD). The organic compounds (COD, BOD_5) demonstrate good covariance with polyatomic anions (PO_4^{3-}, SO_4^{2-}, NO_3^-).

3.4.2. Influence of landfill age

The score plots represent leachate observations and can be used to show the possible evolution of the leachate. By simultaneously considering the score and loading plots from the MP, it is possible to note some trends. Fig. 9 plots the scores of the first two PCs (PC1 vs. PC2). This pair explains 64.7% of the total variation of the dataset. It is possible to recognize two phases in the leachate composition: a first, shorter phase of up to 7 months and a second, longer phase continuing until the end of observations (Fig. 9b). The first phase can be linked to the initial and transitional phases, in which the organic biodegradable components begin to undergo aerobic bacterial decomposition. In this phase, the samples are characterized by an increasing trend of the following parameters: suspended solids, SO_4^{2-}, BOD_5. In the second phase, these parameters tend to decrease, as BOD_5 is assimilated more rapidly by the increased biological activity, and as the landfill becomes anaerobic, nitrate and sulphate are used as electron acceptors in the biological process.

Changes in the MSWP technology and the metagenomic analyses confirm the highlighted trends. Next generation sequencing revealed an increasing participation of microorganisms capable of metabolizing complex organic matter after the 5th month of

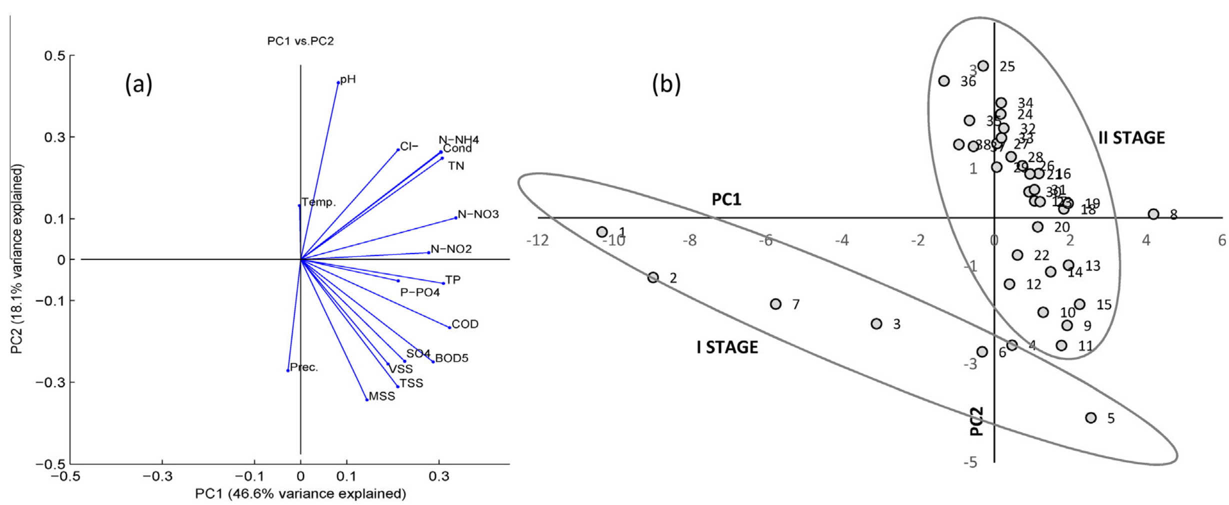

Fig. 9. Loading plot (a) and score plot (b) of the PC1 vs. PC2 for modern prisms (MP).

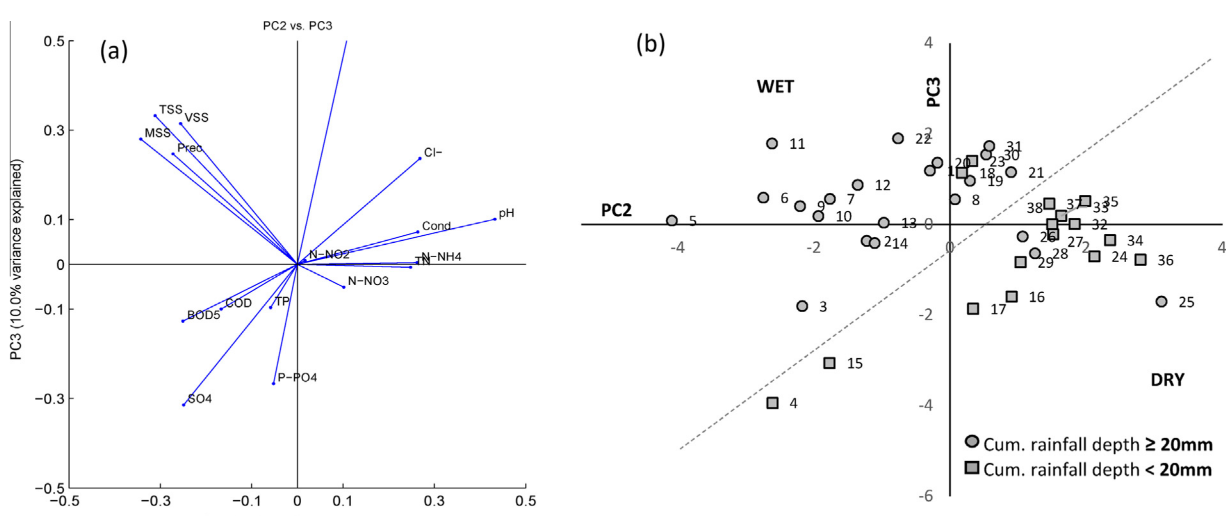

Fig. 10. Loading plot (a) and score plot (b) of the PC2 vs. PC3 for the leachates generated by the modern prism (MP); where □ represents "dry periods" and ● represents "wet periods".

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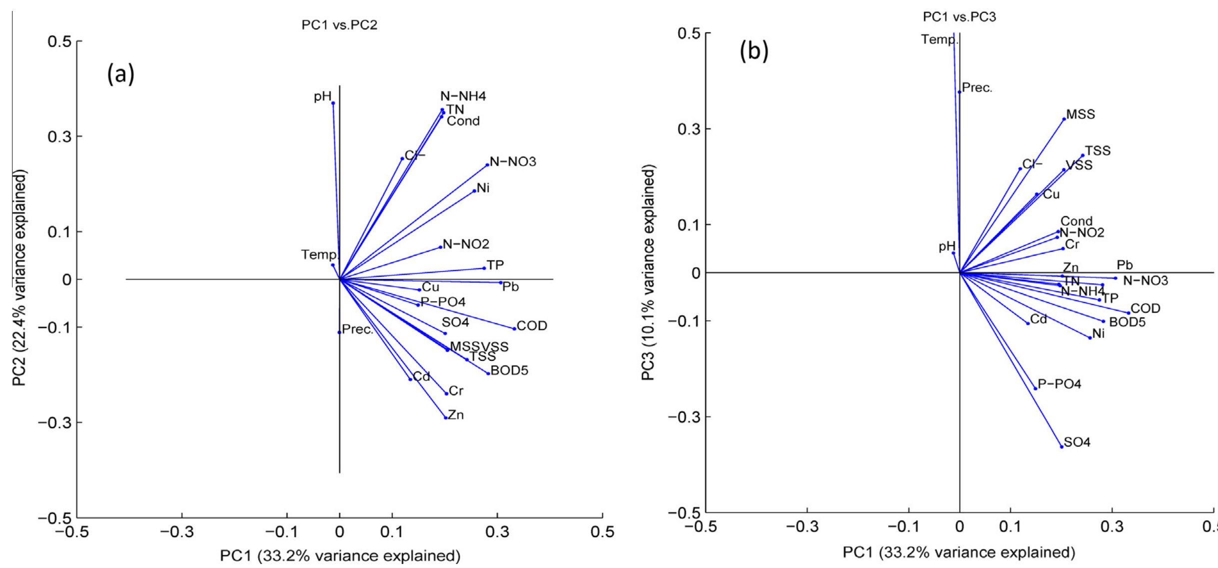

Fig. 11. Loading plot (a) of the PC1 vs. PC2 and (b) the PC1 vs. PC3 for the modern prism (MP); analyses for a limited period with heavy metals.

MP operation. At the same time, untreated wastewaters from the SU, CU and RO were injected back into the MP, creating suitable conditions for their growth (Figs. 1 and 7).

3.4.3. Influence of rainfall

The influence of rainfall was analysed by the second and third PCs (PC2, PC3). "Dry" and "wet" periods were defined, with the former including periods in which the cumulative rainfall was less than 20 mm and the latter including all other periods.

For the MP, even the relatively low variance explained by PC2 and PC3 (equal to 28.1%), from the score plot (Fig. 10), it is possible to observe samples clustering into two distinct regions. The "dry" cluster, as previously defined, properly collects 86.7% of the samples. The loadings show that rainfall has a good positive correlation with suspended solids, which can be linked to the leaching process. Moreover, electrical conductivity and pH are inversely correlated with rainfall. An excess of water dilute leachate, decreasing conductivity and pH was also reported by Tränkler et al. (2005) and Vadillo et al. (1999).

3.4.4. Fate of heavy metals

Concentrations of heavy metals were added to the analysis. To perform PCA with additional data, a different restricted period (February 2012–February 2014) was considered as a result of missing data. Together, the first two PCs explained 55.6% of the data variation, which is lower than the corresponding result from the PCA in Section 3.4.1 (64.7%), which omitted the heavy metals data and used a longer period (Fig. 11).

The loadings showed that PC1 is positively correlated to the metal release. As reported by Jean and Fruget (1994), pH appears to be opposed to metal ion concentrations (Fig. 11). Heavy metal clustering is inversely correlated to pH values, and this result can be explained by the fact that lower pH values (acidic conditions) will increase the solubility of metals. Heavy metals are grouped in the lower-right part of the plot, along with the suspended solids (MSS, VSS, TSS) and organic part of the leachate (COD, BOD). Baun and Christensen (2004) reported that Cd, Cr, Cu, Ni, Pb and Zn found in the colloids of landfill leachate were mostly associated with organic matter. To better understand the heavy metal leaching, their physical (particulate, colloidal, dissolved) and chemical (free metal ions, organic and inorganic complexes) fractionation is needed, especially for leachates generated by MPs in terms of their toxicity and treatment options.

4. Conclusions

The results suggest that MP with a limited deposition of biodegradable carbon may reach maturity faster than PP. The expected reduction of greenhouse gas emissions may not be achieved because of the injection of liquid by-products generated by CUs and SUs to the mass of landfilled waste. Therefore, metagenomic analyses may be useful because they provide superior assessments of the biochemistry of MP. It should also be noted that the heretofore neglected wastewaters generated by SU and CU contain high pollutant loads and should be monitored in future treatment programs. Thus, PPs and MPs as well as CUs and SUs act as hotspots for emerging pollutants, which is in *de facto* contradiction to the 'zero waste' philosophy. Thus it is suspected the real environmental costs of implementing the waste hierarchy in MSWPs may be underestimated.

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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.2016.01.022.

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