Environment Protection Engineering

Vol. 41 2015 No. 1

DOI: 10.5277/epe150107

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RELEASE OF HEAVY METALS FROM WASTE INTO LEACHATE IN ACTIVE SOLID WASTE LANDFILL

Mass balance of selected heavy metals (Cu, Cr, Pb, Hg, Zn, Cd) released from waste into leachate during 8 years of landfill exploitation has been calculated. The average release of all analyzed heavy metals did not exceed 1.7%. Heavy metals released in the largest quantities were Cr (0.025–1.685%), Hg (0.033–1.540%) and Cd (0.082–0.701%). Release of Pb, Cu, Zn was 0.008–0.152%; 0.012–0.085% and 0.007–0.152%, respectively. Cr and Hg do not form an insoluble sulfide precipitate and that is why they are released in a larger amount. Cd and Pb concentrations are positively correlated with the amount of the atmospheric precipitation and the amount of leachates, which means that infiltrating rainfall can leach out these metals from waste. Favorable conditions for leaching of these metals are probably a result of their precipitation as carbonates. A strong correlation between Cu, Zn, and Cr indicates they do not precipitate as carbonates and further they can be subject to desorption processes. In the case of Cu, Zn, Cr and Hg, a positive correlation with the amount of deposited waste was also observed.

1. INTRODUCTION

Although there is a growing focus on composting and incineration of waste as a better solution to an increasing waste handling problem, landfilling is still the predominant method of municipal solid waste disposal [1–3]. According to the EUROSTAT statistics from the year 2010, about 93.4 million tons of municipal waste was landfilled in the whole European Union; among these in Poland were 7.38 million tons. It means that in 2010 in European Union 186 kg of waste per capita was landfilled, while in Poland – 193 kg per capita [4]. The disposal of the municipal solid waste by landfilling is connected with a risk of pollution [5–9]. A mixed municipal solid waste is mainly a mixture of plastics, metals and various organic materials. According to Øygard et al. [1], it is inevitable that there will be a certain portion of heavy

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metals and other environmentally harmful compounds in waste. High concentrations of heavy metals can be found in waste fractions such as: food waste (Cu, Cr, Pb, Zn), plastics (Cd, Cu, Pb, Ni, Zn), coal cinders, glass (Cd, Cr, Ni, Zn), dust (Cu, Cr, Ni), and textile (Cu, Pb, Ni) [10]. The decomposition of organic matter in the waste may also produce potentially hazardous substances.

An unavoidable process occurring in a landfill is the generation of the leachate which is a result of an excessive percolating of rainwater through waste layers [3, 5, 11, 12]. Thus rainfall patterns belong to critical factors in the leachate quality [13]. Due to a complex waste composition, the landfill leachate contains a large number of compounds, some of which can be expected to create a threat to health and nature if released into the environment [14]. Kjeldsen et al. [12] summarized that the most common constituents of leachate are: dissolved organic matter, inorganic micro-components (e.g., Ca^{2+}, Mg^{2+}, K^{+}, Fe^{2+}, etc.), heavy metals, and xenobiotic organic compounds originating from households or industrial chemicals (aromatic hydrocarbons, phenols, chlorinated aliphatics, pesticides, etc.) [12, 15].

A particularly onerous and dangerous pollution in the leachate could be heavy metals. Their toxic nature disturbs the natural biological balance and inhibits self-purification processes. The composition of heavy metals in the leachate varies during the landfill activity depending on the waste composition, waste age, landfill technology and the quality of water percolating through the waste body. The evaluation of the metal release potential from a landfill needs information of the amount of a deposited refuse, its composition and historical records of the leachate quality and quantity at a given time [1, 10]. Some papers have been published on estimation of the mass balance in the leachate from landfills. Oman and Junestedt [14] report that most of heavy metals deposited remain inside landfills, only <0.02% is leached out. According to Øygard et al. [1], it is questionable if the discharge of toxic metals by the leachate actually represents an environmental risk, since a high level of retention of metals appears to take place in most municipal solid waste landfills. However, Bijaksana and Huliselan [2] suggest that the concentration of heavy metals might fluctuate as processes such as precipitation, dissolution, adsorption, and complexation might retain or mobilize metals within leachate ponds. This can pose environmental or health problems as a result. Liu and Sang [7] reported that the leaching concentration of most of heavy metals of a waste depends on the leaching amounts and it has been proven that heavy metals of a leachate can lead to a secondary pollution. Precipitation of sulfides is a common process for heavy metals (excepting Cr) immobilization, but it has been suggested that the content of sulfur in landfills is insufficient to precipitate all the metals present [11]. Martenson et al. [16] claimed that the landfills examined in their study may contain enough sulfur to precipitate only 5% of the metals present. Øygard et al. [1] elucidated the degree of release of heavy metals in sanitary landfills basing on the simulation of mass balance. The results showed that only a small portion of the yearly amount of a deposited metal was discharged into the environment through the leachate. But, as authors reported, the weakness in their investigation was the fact that the mass balance calculation was done for only one year of landfill exploitation [1].

The goal of the work was to estimate the size of the release of Cu, Cr, Pb, Hg, Zn, and Cd from a waste to the leachate during 8 years of municipal landfill exploitation as well as search for the causes/sources of their release. The effect of the rainfall volume, the amount of deposited waste and the leachate quantity in each year of the period of study were also considered in the work.

2. MATERIALS AND METHODS

Landfill site. A municipal landfill located in north-eastern Poland was chosen for the study. The climate in this region is continental with close to 50% of rainfalls occurring between May and August. The average rainfall is about 600 mm per year. The landfill has been operated since 1981 and is still active. The total area of the landfill is ca. 40 ha, including 12 ha is the area of 5 dumping fields. The oldest Field 1 (closed in 2001) is not equipped with a liner system and is situated on a constructed clay/moraine layer site to protect groundwater. In order to limit the leachate migration, a circumferential drainage around the scarp foot of the Field 1 was built. The rest of dumping fields are lined with an impermeable 2 mm thick polyethylene layer at the bottom. Leachates are collected by perforated pipes on top of the liner and flow by gravity to near-by manholes. Then, pumps transfer the leachate to a central manhole where it mixes with leachates from all fields and pumped out of the site to two retention reservoirs. The leachate amount is about 25 000 m^3 per year. In this landfill, municipal wastes are deposited, however, apart from a fluid waste, hazardous substances, and a radioactive and toxic waste. In the instruction for the landfill operation, it has been also prohibited to deposit wet organic wastes, like a sewage sludge. The total average amount of the solid waste deposited in the landfill by the end of 2011 was estimated at 261 800 t.

Sampling and analysis. Leachate samples were collected four times a year (quarterly), starting from March 2004 to December 2011. Additionally, the leachate volumes were measured 12 times a year (monthly). Regularly collected leachate samples were transported to a laboratory and stored at 4 °C. The following parameters were monitored: pH, Pb, Cu, Zn, Cr, Hg, Cd. The analyses were done in the commercial and accredited laboratory of Regional Inspectorate of Environmental Protection in Białystok. Determinations were carried out according to Polish Standards. pH was measured the same day as the samples were collected, by the potentiometric method (according to PN90/C-04540-01). Samples for the metal analyses were preserved by addition of HNO_3. Heavy metals – except for Hg – were analyzed by the atomic emission spectrophotometry ICP-OES (PN-EN ISO 11885:2009), and Hg was determined by atomic absorption spectrophotometry (PB-IN 4:04.11.2010). The results were the mean value of three determinations carried out simultaneously. The samples for metal analysis were not filtered and the test results were reported as total values.

3. RESULTS AND DISCUSSION

The total mass of the deposited waste on the analyzed landfill since 1981 was estimated at 261 800 t. In Table 1, the estimation of the total mass of metals deposited in the analyzed landfill since starting its exploitation is given. It should be noted that there were no analyses of the heavy metal content in the municipal waste which is deposited in the analyzed landfill. Heavy metals, due to significant costs of their analyzes are rarely analyzed in Poland, so that is why the heavy metal content for analyzed waste was adopted from literature [17–20]. The range of the heavy metals content in a municipal waste is shown in Table 1. Zn, Pb, Cr, Cu are deposited in the landfill in greatest amounts, i.e. 1073, 209, 200 and 156 g/t of waste, respectively. The content of Hg and Cd is much lower and does not exceed 1.4 g/t of deposited waste for Hg and 4.0 g/t of deposited waste for Cd. From Table 1 we can range the total mass of analyzed metals in waste as follows: Zn > Pb > Cu, Cr > Cd > Hg.

Table 1

Range of metal content in municipal waste in Poland
and estimated total mass of analyzed metals deposited in waste

Metal	Range of metal content in municipal waste in Poland [g/t of waste]	Estimated total mass of metal deposited in waste [t]
Pb	8–209	2.1–54.7
Cu	24–156	6.3–40.8
Zn	47–1.073	12.3–280.9
Cr	3–200	0.78–52.3
Hg	0.03–1.4	0.008–0.32
Cd	0.47–4.0	0.12–1.05
Total mass of deposited waste on analyzed landfill 261 800 t		

Figure 1 shows the concentrations of heavy metals (mg/dm^3) in the leachate, pH and *EC* value as well as the amount of rainfall and a deposited waste in each year of landfill exploitation.

All values are within the range of leachate quality data [12, 21–24]. pH of leachate was within the range of 7.9–8.5. Since 2008 the pH values in the leachate showed an increasing trend during the next four year investigation period. Similar values of pH in leachate from landfill sites reported Tatsi and Zouboulis (7.3–8.8) [22], Kulikowska and Klimiuk (7.5–8.6) [25], Durmusoglu and Yilmaz (7.5–8.5) [5]. The pH level in the leachate – above 7.5 – indicates that leachate is stabilized and has a high content of organic matter being difficult to decompose.

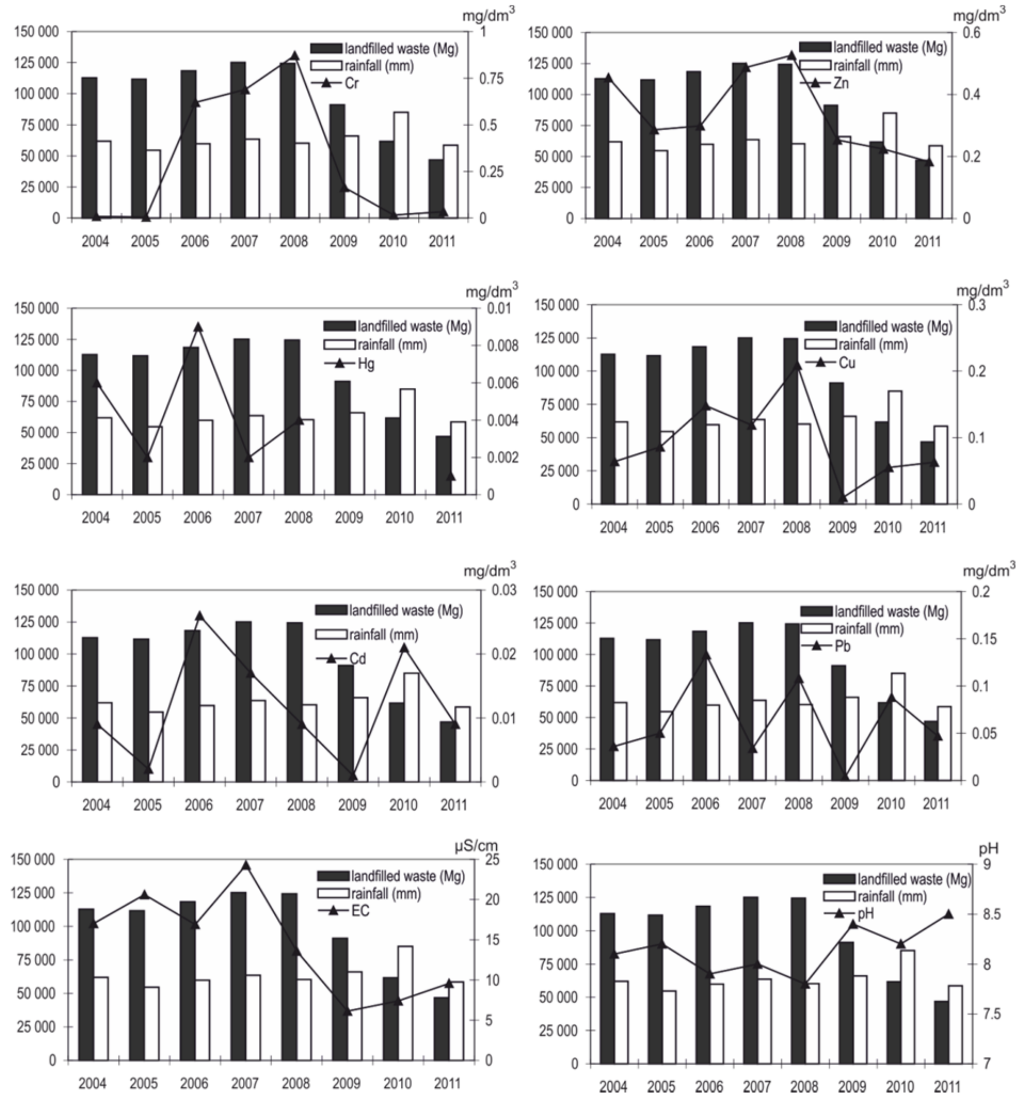

Fig. 1. pH, *EC* value and concentration of heavy metals in leachate from the analyzed landfill. Rainfall in mm×10^2

The concentrations of all analyzed heavy metals were variable over 2004–2011. Till 2008, the Cr, Zn and Cu concentrations were growing and amounted for

Cr – 0.871 mg/dm^3, for Zn – 0.526 mg/dm^3 and for Cu – 0.209 mg/dm^3. After 2008, a drop in the Cr, Zn and Cu concentration was noticed. The distribution of the concentration of these metals is close to the distribution of the amount of the landfilled waste with that amount highest in 2008. The Hg, Cd and Pb concentrations are subject to stronger and less clear fluctuations. Lowest values of metals under study were recorded in 2009, when also a high value of the atmospheric precipitation (660 mm) and a high pH level were recorded (8.4).

In order to evaluate the degree of the release of heavy metals from a municipal waste, the total mass of metals deposited in waste and the total mass of metals in the leachate in the 2004–2011 study period have been calculated (Table 2).

Table 2

Release of analyzed heavy metals through the landfill leachate [kg] in 2004–2011 years

Year	Pb	Cu	Zn	Cr	Hg	Cd
	Total mass of metal in deposited waste [kg]					
2004	902.4–23.575	2.707–17.597	5.302–121.034	338.4–22.560	3.4–157.9	53.0–451.2
2005	893.6–23.345	2.681–17.425	5.250 –119.853	335.1–22.340	3.4–156.4	52.5–446.8
2006	947.0–24.740	2.841–18.466	5.564 –127.014	355.1–23.675	3.6–165.7	55.6–473.5
2007	1.001.1–26.155	3.003–19.522	5.882–134.278	375.4–25.029	3.8–175.2	58.8–500.6
2008	995.0–25.995	2.985–19.403	5.846–133.456	373.1–24.875	3.7–174.1	58.5–497.5
2009	729.0–19.045	2.187–14.216	4.283–97.778	273.4–18.225	2.7–127.6	42.8–364.5
2010	493.1–12.883	1.479–9.616	2.897 –66.140	184.9–12.328	1.8–86.3	29.0–246.6
2011	374.2–9.776	1.123–7.297	2.198 –5.188	140.3–9.355	1.4–65.5	22.0–187.1
	Total mass of metal in leachate [kg]					
2004	0.55	0.74	5.98	0.08	0.08	0.13
2005	1.09	2.31	5.67	0.12	0.04	0.04
2006	2.02	2.17	4.46	7.18	0.10	0.36
2007	0.59	3.12	10.36	17.42	0.07	0.57
2008	1.91	4.00	9.96	16.80	0.06	0.15
2009	0.07	0.22	5.65	4.23	0.00	0.02
2010	2.31	1.04	5.21	0.39	0.01	0.49
2011	1.87	1.92	5.58	0.69	0.03	0.37
	Total release discharge of heavy metal [%]					
2004	0.002–0.061	0.004–0.027	0.005–0.113	0.000–0.023	0.052–2.414	0.028–0.237
2005	0.005–0.122	0.013–0.086	0.005–0.108	0.001–0.035	0.023–1.091	0.010–0.083
2006	0.008–0.213	0.012–0.077	0.004–0.080	0.030–2.023	0.063–2.917	0.077–0.655
2007	0.002–0.059	0.003–0.104	0.008–0.176	0.070–4.639	0.040–1.866	0.114–0.970
2008	0.007–0.192	0.021–0.134	0.007–0.170	0.068–4.504	0.037–1.733	0.030–0.259
2009	0.000–0.010	0.002–0.010	0.006–0.132	0.023–1.548	0.003–0.155	0.006–0.049
2010	0.018–0.468	0.011–0.070	0.008–0.180	0.003–0.209	0.007–0.347	0.198–1.683
2011	0.019–0.093	0.026–0.171	0.011–0.254	0.007–0.495	0.039–1.797	0.196–1.669
Average	0.008–0.152	0.012–0.085	0.007–0.152	0.025–1.685	0.033–1.540	0.082–0.701

The total mass of metals in the waste in the landfill in 2004–2011 was calculated by multiplying the maximum and minimum content of the metal by the amount of the deposited waste in each year. The total mass of metals in the leachate was calculated by multiplying the quarterly average of the metal concentration in the leachate by the quarterly leachate volume. Then, the quarterly values for each analyzed year were summed.

The release of heavy metals is affected by both the leachate quality and the amount of landfilled waste and generated leachates. High volume of landfilled waste in 2007 and 2008 had an impact on higher total discharge of metals. A similar effect on the amount of released metals had high quantity of leachate in 2010. The presented data show that the conditions on the landfill may promote Cr and Hg to release. The total discharge of Cr was 0.025–1.68% and Hg 0.033–1.54%. This can be due to the fact that Cr does not create sulfide precipitates – which are one of the reasons for immoblization of heavy metals, and Hg forms it only to a very small extent. The increased complexation capacity of oxidized humic acids in relation to reduced humic acids or temporary oxidation of metal sulfides to metal sulfates (only in the case of Hg) could increase the concentration of metals in the leachate.

The total discharge of Cd, Pb and Cu was 0.082–0.701%, 0.008–0.152% and 0.012–0.085%, respectively. The lower concentrations of Cd, Pb and Cu can be related to organic matter. According to Christensen et al. [11], organic complexes make up a significant part of the total content of heavy metals: Cd 85%, Pb 71–91%, Cu 59–95%. Complexation of Zn plays a smaller role, i.e. it is 16–36% which is reflected in a higher Zn concentration in the studied leachate. In addition, sulfide precipitation is expected to be of importance in the attenuation of heavy metals. Cr is an exception because it does not form an insoluble sulfide precipitate and that is why its total discharge is higher than those for Pb, Cd, and Cu.

However, the release of heavy metals from the analyzed landfill is not high. Both sorption and precipitation are believed to be a significant mechanism for the metal immobilization and the subsequent low leachate concentration. Waste contains soils and organic matter, which, especially at the neutral to basic environments prevailing in methanogenic leachate, has a significant sorptive capacity. In addition, the solubility of many metal sulfides and carbonates is low and sulfide and carbonate anions are typical of landfills and landfill leachates. A sulfide is formed through sulfate reduction during waste decomposition in landfills, and the sulfide precipitation is often cited as an explanation for low concentrations of heavy metals (except for Cr) [11, 12]. pH of the analyzed leachate is conducive to the sulfide precipitation which is generally induced under near neutral conditions, i.e., at pH of 7–8 [5].

The total mass of heavy metals in the leachate and the quarterly leachate volume in 2004–2011 are shown in Fig. 2. The maximum value of the metal release/transfer corresponds to an increased leachate quantity. There is no clear relation between the heavy metals content and the season of the year. In Table 3, the correlation analysis of heavy metals content in the leachate, the leachate quantity and the value of rainfall are presented.

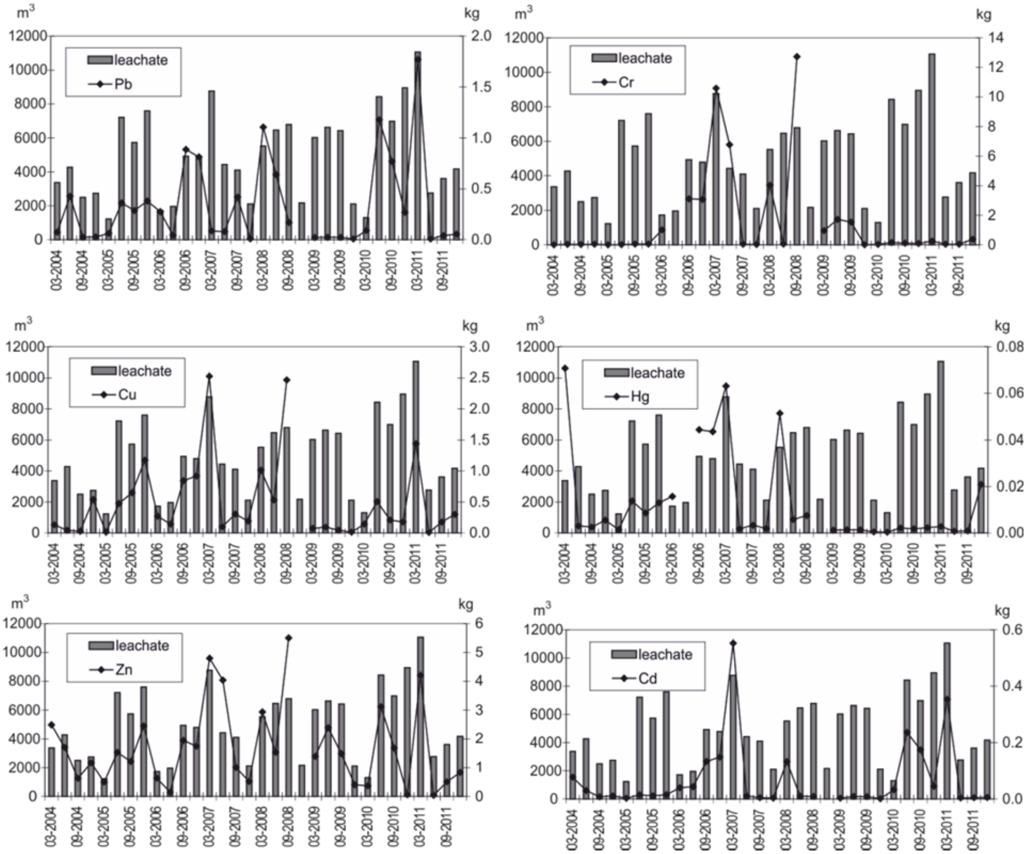

Fig. 2. Total mass of metals in leachate and leachate volume in 2004–2011 (month-year)

The obtained results indicate that contents of all analyzed heavy metals in the leachate are negatively correlated with the pH value. A strongest negative correlation occurs in the case of Hg ($r = –0.74$) and Cr ($r = –0.66$). There is a strong positive correlation between Cu and Zn ($r = 0.72$), Cu and Cr ($r = 0.76$), Zn and Cr ($r = 0.87$), thus the release of these metals can be caused by the same reasons/causes. Both Cr, Cu and Zn are not subject to precipitation as carbonates; additionally Cr is not subject to precipitation as sulfides and this may be the main reason for the release of these metals. Only in the case of Cd and Pb, a positive correlation between the release value and the rainfall quantity were found ($r = 0.44$ for Cd and $r = 0.26$ for Pb). The correlation of these metals with the leachate quantity was 0.25 for Pb and 0.12 for Cd. In this case, the carbonate fraction can be a substantial source causing release of metals. In contrast to the other analyzed metals Cd and Pb may precipitate as a carbonates. Metals associated with this fraction are easily washed out [26]. Therefore for Cd and Pb, higher release may result from their dilution and leaching from waste. The release of Hg is also connected with the quantity of landfilled waste ($r = 0.72$), like in case of Cr ($r = 0.61$), Zn ($r = 0.5$) and Cu ($r = 0.47$).

Table 3

Correlation analysis between heavy metals content in leachate, leachate quantity and the value of rainfall

Subject	Metal content in leachate						Deposited waste	pH	Leachate quantity	Rainfall
	Pb	Cu	Zn	Cr	Hg	Cd				
Pb	1									
Cu	0.36	1								
Zn	–0.18	0.72	1							
Cr	–0.03	0.76	0.87	1						
Hg	0.05	0.44	0.23	0.42	1					
Cd	0.43	0.25	0.2	0.29	0.14	1				
Deposited waste	–0.34	0.47	0.5	0.61	0.72	–0.22	1			
pH	–0.31	–0.67	–0.46	–0.66	–0.74	–0.17	–0.76	1		
Leachate quantity	0.25	0.05	0.11	–0.07	–0.85	0.12	–0.55	0.37	1	
Rainfall	0.26	–0.4	–0.14	–0.16	–0.47	0.44	–0.44	0.1	0.48	1

4. CONCLUSIONS

The conducted analyses have shown that the release of heavy metals in a municipal landfill is fairly low. Hg and Cr were released to a larger extent but their discharge never exceeded 2%. The discharge of Pb, Cu, Zn and Cd was below 1%. A key role in the immobilization of these metals may play the content of organic matter and sulfide precipitation. Cr does not form insoluble sulfide precipitates and that is why it is released in larger amounts.

There was no clear relationship between the amount of the heavy metals transfer and the season of the year. In the case of all heavy metals, the value of their transfer was negatively correlated with the pH value. The reason for Cr, Cu and Zn leaching is that they are not subject to the precipitation as carbonates and – in case of Cr – as sulfides. For Pb and Cd the value of their transfer is affected mostly by the leachate quantity and the rainfall. The release of Hg is positively correlated with pH of leachate and the quantity of the deposited waste.

It should be noted that some of the leachate can migrate in the form of uncontrolled leakage through the landfill bottom or during the transfer to retention reservoir.

Thus, the release of heavy metals may be, in fact, slightly higher than those presented in the work. In addition, in this work the content of metals in municipal waste was adopted from literature. The real range of the metal content in the waste from the analyzed landfill is most likely within these limits, but can undergo fluctuation in the course of years.

Nevertheless, conducted analyses have shown that a landfill can function as a long term or permanent reservoir for many metals. To maintain such a landfill function, a reducing environment and thus retention of metals in landfill should be ensured.

ACKNOWLEDGEMENTS

The author wishes to thank Regional Inspectorate of Environmental Protection in Białystok as well as managers of the analysed landfill for their help in collecting data.

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