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Quality of Leachate From Landfill with Reverse Osmosis Concentrate Recirculation

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ABSTRACT
The purpose of this study was to determine the changes of leachate quality on a selected municipal landfill with recirculation of concentrate from the reverse osmosis (RO) process. The research was carried out on an exploited municipal waste landfill, from which the samples of leachate, permeate and concentrate were collected in the years 2013–2015. The obtained samples were subjected to the physicochemical analysis, including the determination of general pollution parameters as well as organic and inorganic indicators. The obtained results showed that the concentrate recirculation resulted in intensification of the degradation processes in the waste body, expressed by the increase of biodegradability rate (BOD_5/ COD) and the value of conductivity and dissolved solids. Over time, the value of conductivity and dissolved solids stabilized; however, they reached the values higher than in conventional landfills. A characteristic feature of the long-recirculation landfills was a continuous increase in the concentration of ammonia nitrogen and COD values in the leachate.

Keywords: landfilling, leachate, concentrate recirculation, reverse osmosis

INTRODUCTION

The consequence of waste landfilling is the formation of leachate, the characteristic feature of which is the variable quantity and quality over time, depending on the type of waste landfilled and the method of landfill exploitation. A biological method is used to treat the leachate from young landfills sites with a high BOD_5/COD ratio. In the case of the stabilized leachate, for which the value of BOD_5/COD ratio is lower, the physicochemical methods are usually used. However, in the last decades a reverse osmosis has become an option increasingly often considered as the treatment method for landfill leachate. As a result of the RO process, the pollutants are divided into two streams: permeate (filtrate) and highly contaminated concentrate, which is usually recirculated into the waste body. It is assessed that about 20–50% of the treated leachate volume is recirculated as a highly contaminated concentrate to the waste body.

Recirculation of the concentrate from reverse osmosis process is currently the most controversial issue regarding reverse osmosis operation on municipal landfills [Wang et al. 2006, Renou et al. 2008, Liu et al. 2008, Li et al. 2008]. Such a solution, due to the increase in the waste humidity, causes a change in the rate of the occurring degradation processes, changes in the quality of leachates and a change in the leachability of individual pollutants from the waste body. The opinions about the effects of recirculation are varied, and the scientific literature on this subject is rather limited. The research results of Calabrò et al. [2010] indicate that the only change observed after the introduction of the concentrate recirculation was a moderate increase in the leachate value of COD and the concentration of nickel and zinc. According to the studies conducted by Eipper and Maurer [1999] and Heinigin [1995], the effect of concentrate recirculation on the quality of leachate is observed only in a short period of time, immediately after recirculation. On the other hand, Heyer and Stegman [2002] say that the recirculation of the concentrate for a longer period of time is unfavorable due to the generation of a larger amount of leachate and the possibility of containing some pollutants. Furthermore, Calabro et al. [2010] emphasize that the beneficial effect of recirculation is the intensification of the reduction processes in the waste body, which leads to the reduction of sulphates to sulphides, thus neutralizing the heavy metals and, as a consequence, decreasing their concentration in the leachates. Ledakowicz and Kaczarek [2002] indicate that leachate recirculation can lead to the weakening or even stopping of the methanogenesis process due to the high concentrations of organic acids and low pH. Baran et al. [2009] notes that the use of the concentrate reflux causes the introduction of additional pollutants load to the waste dump. Because the substances with short half-life period (biodegradable) decompose quicker, as opposed to the substances with long half-life one (hardly biodegradable), over time, the hardly biodegradable substances can accumulate in the landfill in large quantities [He et al. 2005, Baran et al. 2009].

The above-mentioned literature review has pointed that the concentrate recirculation into the waste may cause the change of the decomposition rate and the change of contaminants release from the waste, consequently resulting in the changes in the leachate quality. However, the literature data do not indicate unambiguously what is the effect of concentrate recirculation on the quality of leachate. There is also no detailed information on the comparison of the quality of leachate from the recycled and unrecycled landfills. Thus, the objective of this work was to assess the impact of concentrate recirculation from the RO process on the leachate quality and compare the quality of leachate from landfills using recirculation with the quality of leachates from "conventional" landfills.

MATERIAL AND METHODS

The research was carried out on real leachates from two municipal landfills in the same exploitation phase (Landfill E and Landfill H). The concentrate recirculation was carried out at the first of the landfills taken for consideration (Landfill E), while the other one was operated in a traditional manner (Landfill H). In the second, third and fourth year of exploitation of the above-mentioned landfills, the following samples were collected:

- samples of leachate and concentrate from Landfill E with a functioning reverse osmosis system and concentrate recirculation,
- samples of leachate from Landfill H without concentrate recirculation.

The samples were collected at 3–4 month intervals. A total of 15 leachate samples were collected from Landfill H as well as 13 leachate samples and 6 concentrate samples from Landfill E. The concentrate samples were collected in the 2nd and 3rd year of the landfill exploitation due to the closure of the reverse osmosis installation after this time. In order to ensure a reliable quality of the samples, no collections of such were conducted after heavy rainfall or after prolonged periods of drought. The collected samples were subjected to the physicochemical analysis including the determination of general pollution parameters, organic and inorganic indicators as well as heavy metals. The scope of leachate and concentrate analysis included:

a) general parameters: conductivity (EC), pH, dissolved oxygen,
b) organic indicators: chemical oxygen demand (COD),
c) nitrogen ammonia ($N\text{-}NH_4^+$),
d) inorganic components: chlorides (Cl^-), sulphates (SO_4^{2-}), sulphides (S^{2-}), iron (Fe),
e) heavy metals: cadmium (Cd), copper (Cu), nickel (Ni)

All parameters were analyzed according to the standard methods for the examination of water and wastewater [Rice et al. 2012]. The obtained results were the mean value of three determinations carried out simultaneously. The analyses were performed with the use of the HACH HQ40 potentiometer, HACH DR2000 and DR 3900 spectrophotometers, iCE3400 and iCE3300 atomic absorption spectrometers, as well as Thermo Scientific ICS 5000+ ion chromatography system.

The Statistica software was used for data analysis in this study. The basic statistical analysis included calculation of minimum, maximum and mean value. The measures of variability were reported in standard deviation. In order to evaluate the changes in the leachate quality after the introduction of concentrate recirculation, the Student's t-Test was performed.

RESULTS AND DISCUSSION

Table 1 summarizes a quality of concentrate and leachate from Landfill E with recirculation and quality of leachate from Landfill H without recirculation.

The average pH value of the concentrate from landfill E was 6.7. This leachate, prior to entering the reverse osmosis system, was acidified to pH 6.0÷6.5, which affected the pH of the generated concentrate. The value of the electroconductivity in the concentrate from the landfill E was 8066 μS/cm on average. The BOD value in the concentrate from the E landfill reached an average value of 549 mg O_2/dm^3. The value of the BOD/COD indicator in the concentrate from Landfill E was 0.43. This value is comparable to the BOD/COD levels characteristic for the leachates in the intermediate stage, where the content of biodegradable contaminants is still high. The amount of nitrogen ammonia in the concentrate was 327 mg/dm^3. Among the analyzed inorganic components, chlorides and sulphates had the highest value, their average concentration in the concentrate was 5608 mg/dm^3 and 1898 mg/dm^3, respectively. The concentration of S^{2-} in the recirculated concentrate was 0.8 mg/dm^3 and iron – 6.28 mg/dm^3. The concentration of heavy metals in the concentrate from landfill E can be ordered as follows: Ni (0.73 mg/dm^3)> Cu (0.24 mg/dm^3)> Cd (0.09 mg/dm^3).

In order to compare the leachate quality from Landfill E with the concentrate recirculation and leachate quality from Landfill H without recirculation, a Student t-Test was performed and its results were presented in Table 1. The obtained results indicated that significant differences in the quality of analyzed leachate concern the value of EC, COD and the concentration in leachate of $N\text{-}NH_4^+$, SO_4^{2-}, O_2 and Fe. The characteristics of this parameter was presented in Figure 1, Figure 2, Figure 3 and Figure 4.

Leachates from the landfill E were characterized by a conductivity average value of 8,499 μS/cm (Fig. 1). The introduction of concentrate recirculation resulted in the EC increase from 3300 μS/cm at the beginning of recirculation to 6910 μS/cm at the end of this process. After closure of the recirculation system, the conductivity value continued to increase and in the fourth year of Landfill E exploitation the value of 13300 μS/cm was reached. This increase in the leachate mineralization is the result both of the concentrate recirculation as well as intensive physical and chemical processes characteristic for the young landfills at the acidic phase. At the same time, the average EC value on Landfill H was 21 584 μS/cm and increased from 17 600 μS/cm after a year of operation of the landfill to 36 000 μS /cm in the fourth year of operation.

The average COD value of the leachate from Landfill E was 2514 mg O_2/dm^3 (Fig. 1). The introduction of concentrate recirculation caused the

Table 1. The characteristics of concentrate and leachate from Landfill E and Landfill H and Student's t-Test results

Parameter*	Landfill E with concentrate recirculation				Landfill H without recirculation		Student t-Test for leachate from Landfill E and H		
	concentrate		leachate		leachate				
	Average N=6	Stand. dev	Average N=13	Stand. dev	Average N=15	Stand. dev	t	df	p
pH	6.7	0.33	7.5	0.33	7.9	0.82	-1.59	26	0.12
EC	8066	6326	8499	3319	21584	7018	-5.93	26	**0.00**
O_2	1.89	2.35	0.55	1.06	6.89	5.363	-4.18	26	**0.00**
COD	1646	1429	2514	1574	6215	4766	-2.67	26	**0.01**
BOD	549	421	1100	1018	n.a.	n.a.	-	-	**-**
$N\text{-}NH_4$	327	229	321	258	981	805	-2.82	26	**0.01**
SO_4^{2-}	1898	1833	389	316	77.4	142	3.42	26	**0.00**
S^{2-}	0.83	0.96	8.43	11.7	n.a.	n.a.	-	-	**-**
Fe	6.28	3.82	2.81	2.32	10.7	10.85	-2.56	26	**0.02**
Cl^-	5608	6023	1088	556	1245	837	-0.57	26	0.57
Cd	0.093	0.065	0.020	0.023	0.09	0.078	-2.13	11	0.05
Cu	0.243	0.127	0.081	0.080	0.31	0.313	-1.89	11	0.08
Ni	0.737	0.732	0.228	0.279	0.27	0.163	-0.31	11	0.76

* All in mg/dm^3 apart EC (μS/cm) and pH; n.a. – not analyzed.

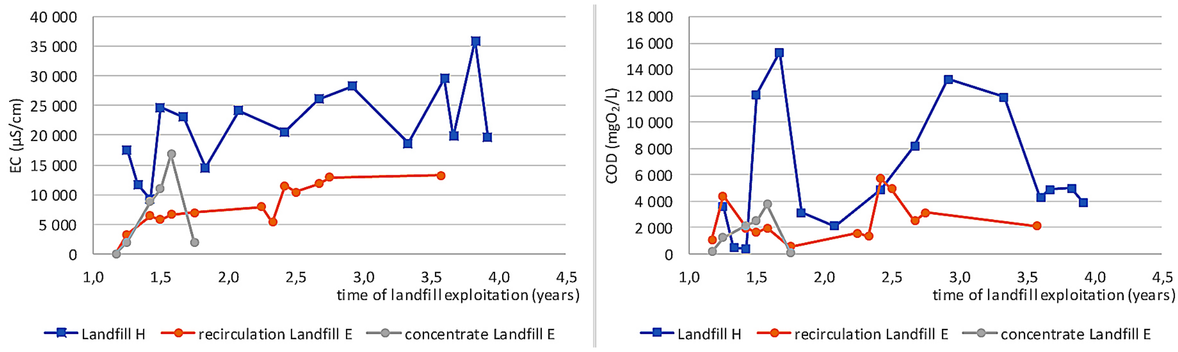

Fig. 1. EC and COD value in concentrate and leachate from Landfill H and recirculation Landfill E

increase of COD value from 1090 to 4400 mg O_2/dm^3 due to the release and hydrolysis of organic compounds supplied to the waste solid with concentrate (BOD/COD ratio of the concentrate was 0.43). The average COD value in the leachate from Landfill H (matured leachate) was 6215 mg O_2/dm^3 and throughout the research period fluctuated significantly, oscillating between 1670 to over 15000 mg O_2/dm^3.

The average concentration of N-NH^{4+} in the leachates from landfill E was 321 mg/dm^3. The introduction of concentrate recirculation on Landfill E resulted first in an increase in the leachate of nitrogen ammonia to 1000 mg/dm^3 and then decrease in its value to about 200 mg/dm^3 (Fig. 2). After stopping the recirculation process, the concentration of nitrogen ammonia was still increasing and at the end of the research period, it exceeded the value of 500 mg /dm^3. According to Kulikowska and Klimiuk [2008], the ammonium ion is released from young leachate mainly as a result of amino acids deamination in the process of organic compounds decomposition, whereas in the older leachate it is a product of the hydrolysis and fermentation of nitrogen contained in the biodegradable waste fraction. The average nitrogen ammonia concentration from Landfill H was 981 mg/dm^3 and during the whole research period, it decreased to 310 mg/ m^3. The research conducted by Christensen et al. [2001], Surmacz-Górska [2001], Bilitewskiego et al. [2006] indicated that the value of nitrogen ammonia in the leachate increases intensively in the initial phase of conventional landfill operation (without recirculation) and then gradually stabilizes. The conducted studies indicated that the recirculation of the concentrate could cause an increase in the concentration of nitrogen ammonia in the leachate. After finishing the recirculation process, the nitrogen ammonia concentration in leachate has stabilized its level stabilized.

The concentration of iron in the leachate from Landfill E was 2.81 mg/dm^3, while the average concentration of this metal in the leachate from Landfill H reached the value of 10.7 mg/dm^3 (Fig. 2). High concentrations of iron, together with a value of nitrogen ammonia, indicate the anaerobic conditions prevailing in the waste body [Staton et al. 2004].

The average concentration of sulphates in the leachate from landfill E was 389 mg/dm^3 (Fig. 3). The value of sulphate in the leachate from Landfill H was lower and amounted to 77.4 mg/dm^3. High concentrations of SO_4^{2-} in the leachate from Landfill E result from recirculation of a concentrate with high content of sulphates (average

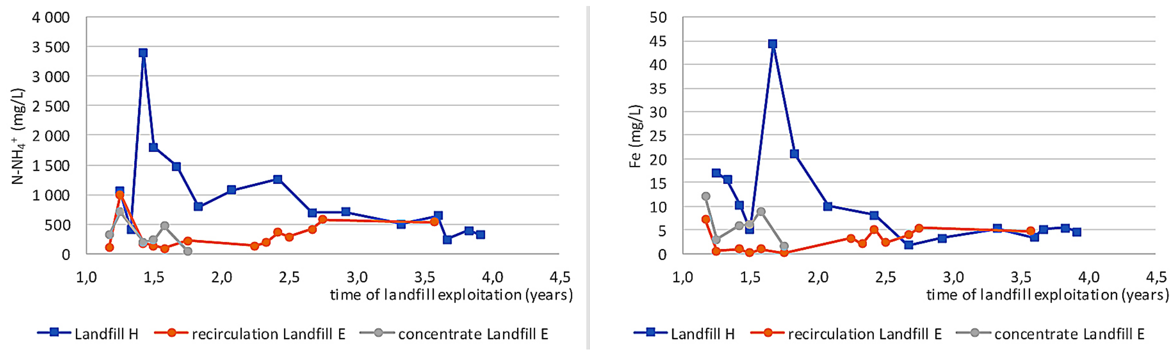

Fig. 2. Concentration of N-NH_4^+ and Fe in concentrate and leachate from recirculation Landfill E and Landfill H

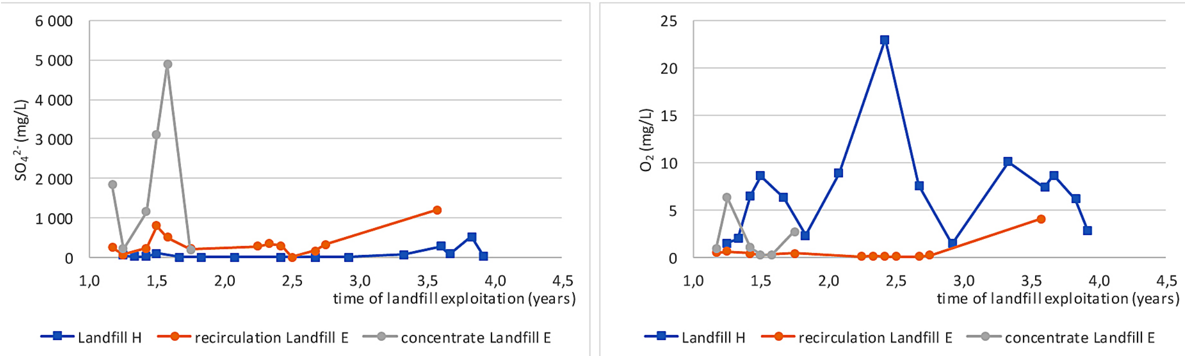

Fig. 3. Concentration of SO_4^{2-} and O_2 in concentrate and leachate from recirculation Landfill E and Landfill H

1898 mg/dm³) into the waste body; they come from leachate acidification by sulphuric acid before the reverse osmosis process. According to Christensen et al. (2001) the concentration of sulphates in the leachates can be reduced only by dilution and their reduction to S^{2-} [Durmusoglu and Yilmaz, 2006]. Therefore, the concentration of sulphates in the leachate negatively correlates with the sulphides concentration due to the conversion of SO_4^{2-} into S^{2-} under reducing conditions or the transformation of S^{2-} into SO_4^{2-} in the oxidizing environment.

The average oxygen concentration in the leachate from the recirculation Landfill E amounted to 0.55 mg/dm³ and was over twelve times lower than the oxygen concentration in the leachate from Landfill H (6.89 mg/ m³). The reduction conditions favored precipitation of sulphides, the concentration of which in the leachate from the recirculated Landfill E was 0.83 mg/dm³. As it was said, the concentrate recirculation introduces additional quantities of sulphates and sulphides coming from leachate acidification by sulphuric acid before the RO process. Martensson et al. [1999] pointed out that the waste body does not contain enough sulphur to immobilize all heavy metals present in the waste. Consequently, recirculation of the concentrate may be advantageous for heavy metal immobilization and the decrease in their concentration in the leachate. This is confirmed by the data presented in Table 1. According to Durmusoglu and Yilmaz [2006], the concentration of heavy metals in leachates depends on the intensity of such processes as: complexation, redox reactions, sorption and precipitation, wherein sorption and precipitation are considered as the main mechanisms responsible for their immobilization.

Table 2 compares the quality of the analyzed leachates from Landfill E and Landfill H with the literature data.

The research results indicate that at the beginning of concentrate recirculation, the values of the analyzed pollutant indicators are within the ranges typical for young municipal landfills. A similar relationship was found by Reinhardt and Al-Yousif [1996], Morris et al. [2003] and Bilgili et al. [2007] during their studies on landfills with recirculation of leachates. However, it should be pointed that the concentrate recirculation in a longer period of time may cause accumulation of inorganic compounds in leachates and generate more leachates, as noted by Heyer and Stegman [2002] in their research. The accumulation of chlorides and nitrogen ammonia may also be a disadvantageous aspect of recirculation of leachate in the long term [Robinson 2005, Francois et al. 2007, Calabro et al. 2010]. Moreover, the research of Baran et al. [2009] and He et al. [2005] indicate the possibility of large amounts of hardly biodegradable substances accumulating over time in the leachate. All these observations, however, require confirmation in further research.

CONCLUSIONS

The conducted research indicated that one of the characteristic features of the leachate from recirculation landfill is the low value of oxygen, which promote the reductive conditions in the waste body. Another characteristic is the high concentration of sulphates coming from leachate acidification before their direction to the RO system. In this anaerobic environment, sulphates are reduced to sulphides, which favors heavy immobilization in the waste body. The conducted research showed that the concentrate recirculation

Table 2. Leachate quality on Landfill E and Landfill H against literature data

Parameter*	Landfill E with recirculation	Landfill H without recirculation	Young landfills (below 3 years of exploitation)		Stabilized landfills (over 10 years of exploitation)	
	range	range	average	range	average	range
pH	7.2÷7.9	6.1÷9.5	-	6.2÷7.8	-	7.2÷9.0
EC	3.3÷6.91	9.2÷36	17	2÷50	17	2÷50
COD	520÷4400	390÷15310	10000	500÷40000	2 700	460÷8300
BOD	200÷850	n.a.	1000	600÷2000	200	20÷700
$N\text{-}NH_4^+$	90÷999	235÷3400	800	10÷3000	1000	10÷2000
S^{2-}	0.1÷39.3	n.a.	-	-	-	-
SO_4^{2-}	60÷800	0.0÷525	250	30÷1000	200	10–500
Fe	0.1÷7.2	1.8÷44.25	100	20÷2000	25	1÷200
Cl-	302÷1124	50÷2650	1000	100÷5000	2000	100÷5000
Cd	0.02÷0.06	0.002÷0.19	-	0.001÷0.5	-	0.001÷0.5
Cu	0.06÷0.18	0.080÷0.98	-	0.005÷0.6	-	0.005÷0.6
Ni	0.12÷0.82	0.070÷0.56	-	0.01÷1	-	0.01÷1

* All in mg/dm^3 apart EC (mS/cm), and pH; n.a. – not analyzed.
Source: Szymański 1987, Szymański 2009a, 2009b, Lipniacka-Piaskowska 2010, Fudala-Książek 2011, Bilitewski et al. 2006, Szymański and Nowak 2012.

affects the lower concentrations of all analyzed heavy metals in comparison to the leachates from conventional landfills.

Despite the differences in the leachate composition, the quality of leachate from landfills in the first years of concentrate recirculation is within the ranges accepted for conventional landfills.

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