ARCHIVES OF ENVIRONMENTAL PROTECTION

vol. 34 no.3 pp. 63 - 70 2008

PL ISSN 0324-8461

INFLUENCE OF OZONATION ON LEACHATE QUALITY FROM LANDFILLS WITH DIFFERENT DEGREES OF SOLID WASTE PRE-TREATMENT

M. LEBIOCKA, A. MONTUSIEWICZ, M. PAWLOWSKA*, J. OZONEK, E. SZKUTNIK , M. ROSŁAN

Lublin University of Technology, Faculty of Environmental Engineering, 20-618 Lublin, ul. Nadbystrzycka 40 B, Poland
*corresponding author: tel. +48 81 5384409, fax +48 81 5381997, e –mail: m.pawlowska@pollub.pl

Abstract: Landfilling is the main method of waste disposal in Poland as well as in most countries all over the world. Leachate originating during waste deposition may be a source of ground water pollution. The aim of the paper was to characterize and compare the composition of leachate originating from three landfills in Lublin Province (Poland) and differing in their methods of waste pre-treatment. Ozonation was used in the initial trials to treat landfill leachate. Experiments were carried out to determine whether ozonation using a single dose of 1.8 gO_3/m^3 has the same effect on the efficiency of organic removal from leachates characterized by different ages and degrees of solid waste pre-treatment. From analyses (BOD_5, COD, $N\text{-}NH_4$, heavy metals) it was concluded, that excluding some fractions (glass, paper, plastics, aluminum, fine organic fraction) from the waste mass affects the leachate quality. The studied oxidizing method was found to influence the BOD_5 and COD levels.

Keywords: landfill leachate, ozonation, BOD_5, COD, heavy metals, waste pre-treatment

INTRODUCTION

Landfilling is the main method of waste disposal in Poland. In 2004, 9.5 million tons of wastes was treated, of which about 94% was deposited on landfills. Waste deposition on landfills can exert a negative influence on the environment by emissions of greenhouse gasses and odors to the atmosphere and ground water pollution by originating leachate (Al-Muzaini et al., 1995; Irene and Lo, 1996). Leachate is formed as a result of precipitation and infiltration of hydrolysates through the deposited wastes layer. Hydrolysates are products of biochemical changes of organic substances. Leachate contains dissolved substances scoured away by the rainfall, and also organic and mineral substances constituting intermediate products of the fermentation processes occurring in the wastes layer. Different types of wastes are deposited on the landfills, what causes the leachate to be generally a complex mixture of organic and inorganic compounds (Christensen et al, 2001). The composition of leachate mainly depends on the morphology of the deposited waste materials, landfill age and landfilling technology (Ward et al., 2005). According to Christensen, leachate constituents can be classified into four groups of pollutants:

1. dissolved organic matter (represented as COD, BOD_5 or TOC) including CH_4, volatile fatty acids (VFA) and refractive compounds,
2. inorganic macrocomponents : Ca, Mg, Na, K, NH_4^+, Fe, Mn, Cl, SO_4^{2-} and HCO_3^-,
3. heavy metals eg. Cd, Cr, Cu, Pb, Ni, Zn,

4. xenobiotic organic compounds (XOC) – originating from the households and industrial chemicals, present in relatively low concentrations in leachate (usually below 1mg/l of selected compounds).

In the light of the Water Law regulations regarding leachate treatment (Journal of Laws No. 115, item 1229, 2001) landfill leachate is classified as industrial wastewater. The research concerning leachate amount and composition constitutes a part of the landfills monitoring program resulting from the European Union Directive 99/31/EC (Robinson et al, 2005).

According to the literature, initial treatment of wastes consists of mechanical separation of biodegradable fractions and types of wastes that can be recycled. This step has a significant influence on leachate quality (Komilis D.P. et al, 1999, Leikam and Stegmann, 1999). Until now, the issue was not a subject of much detailed study in Poland. However, the matter has recently become very important due to the fact that knowledge of leachate composition may influence its future treatment.

According to the Directive 2006/12/EC of the European Parliament and of the Council it is necessary to reduce waste mass through the development of its reuse and separation of biodegradable fraction. Selective waste collection programs are being gradually introduced into the Polish system of waste management. Waste segregation allows decreasing the amount of landfilled waste and in consequence reduces the need for new or further development of existing waste disposal plants. Selective waste collection at source is the most effective type of segregation. Nevertheless, in Poland, in most cases it is still carried out on the landfills, and not at the location of waste production. The most often recovered materials are: glass, waste paper, scrap metal, plastics and sometimes biodegradable fraction.

There are many of technical applicability and the treatment performance of various advanced oxidation technologies for landfill leachate as follows ozonation, photo-Fenton oxidation, coagulation, flotation. According to literature ozonation and Fenton oxidation are the most frequently studied.Ozonation is a process often applied in the treatment technology of different media, like water, wastewater, landfill leachate (Bila et al., 2005; Marttinen et al., 2002; Monje-Ramirez and Orta de Velásquez, 2004). According to the Marttinen (2002) ozonation enhances the biodegradibility of leachate.

The aim of the work described here was a to make a comparison of the mineral forms of nitrogen, COD and BOD_5 values, heavy metal contents in leachate originating from three landfills of similar age, located in Lublin Province, as well as the effect of ozonation on the studied media.

MATERIALS AND METHODS

Characteristics of the examined objects

The studies were conducted on three landfills located in Lubelskie Province:

1) Landfill Pulawy has been in operation since 2001. The capacity of the landfill is 200000m^3 with a designed throughput of 22000 Mg of waste per year. At present about 18000 Mg of waste per year is received, mainly from the Pulawski district, including the city of Pulawy and communes: Pulawy, Janowiec, Zyzyn, Konskowola and Wawolnica. From the whole waste mass about 7000 Mg/year is stored on the landfill. The plant in Pulawy owns a waste fermentation installation, the second in Poland (the other is at Zgorzelec). The Municipal Waste Treatment Plant in Pulawy consists of: a waste sorting line, an installation for separating biofraction from the municipal wastes and an installation for preparation of biofraction suspension. The prepared suspension along with the sewage sludge originating from the municipal wastewater treatment plant is subjected to a mesophile fermentation process in the separated fermentation tanks.

The thickened fraction is transported onto the sorting line in order to separate recycling materials. The materials are collected by the mediating firms and transported to the processing plants. The remaining part of the thick fraction, that cannot be reused (so-called ballast) is directed

to the bale press, which allows thickening (about 750 g/m^3), and afterwards deposited on the landfill.

Selective collection and separating so-called thick fraction from wastes allows gaining significant amounts of recycling materials. Thanks to this procedure, the amount of wastes deposited on the landfill is decreased and in consequence the period of landfill exploitation is prolonged.

2) Landfill Stara Wies near Leczna – The construction of the Municipal Waste Treatment Plant in Stara Wies was completed in 2003. The main task of the Waste Treatment Plant is collection and utilization of municipal wastes originating from the area of Leczna (town and commune) and the neighbouring communes: Puchaczow, Milejow, Cycow. Recycling of wastes is conducted in the segregation plant situated in the area of the landfill. The total capacity of the landfill is 55500 m^3. According to data collected in 2004, the amount of wastes received annually is about 4700 Mg. The landfill in Stara Wies consists of two basins. The exploitation of the old basin is completed. At present, the second basin, formed in 2003, is being exploited.

3) Landfill Rokitno – municipal wastes and wastes similar in composition and character to those originating in households are deposited there. Rokitno landfill receives wastes from the Lublin city and surroundings. The legal owner of the Rokitno landfill is the community of Lublin and the administrative owner, who won the tender is the Municipal Water and Wastewater Company in Lublin. The Landfill in Rokitno has no waste segregation line. Every year 113000 Mg of wastes is deposited on the landfill (data from 2004). The total capacity of the landfill in the first stage is 2480000 m^3.

The area of the landfill is 29.22×10^4 m^2 and it is divided into two stages:

- first stage area – 11.62×10^4 m^2,
- second stage area – 17.60×10^4 m^2.

Additionally, the first stage was sub-divided into two tasks:

- task 1 – 6.03×10^4 m^2,
- task 2 – 5.59×10^4 m^2.

In 2002 the exploitation of the first basin, assigned as a task 1, was finished. Since then, the second basin, within the framework of the second task is being exploited. Completion of the second basin exploitation is expected in 2012.

All examined objects are equipped with bedrock sealing, protecting ground waters from pollution. Landfilling technology guarantees protection of surface waters and allows minimizing negative environmental effects.

Methods of examination

The value of COD and BOD_5, ammonia nitrogen and heavy metals content in leachate were examined in the laboratory of the Environmental Engineering Faculty in Lublin University of Technology.

COD determination in leachate was carried out on the basis of standard method described in Polish standard PN-74/C-04578.03. BOD_5 was also determined in accordance to the Polish standard PN-84/C-04578/05.

Ammonia nitrogen was determined using Flow Injection Analysis - FIAstar®5000 from Foss Tecator (Warsaw, Poland).

Heavy metals were determined with the use of atomic emission spectrometer coupled with plasma ICP-OES JY 238 Ultrarace (Jobin Yvon-Horriba, France). The samples for heavy metals determination were mineralized in the Microwave Oven 3000 solv Anton Paar, Austria. Maximum power of magnetron 1400W.

The oxidation (ozonation) of leachate originating from the three landfills of the Lublin Province was run in a glass reactor in which ozone was introduced from the bottom with the use of Akwatech diffuser. A water pump installed in the system allowed gas flow. The scheme of the laboratory test stand is presented in Figure 1. In each trial the reactor was filled with 2 dm^3 of

landfill leachate. The whole volume was mixed using air, and after about 5 minutes ozone was introduced. The flow of ozonized gas was 0.9dm^3/min, and the concentration of ozone was 1.8 g/m^3. The samples of 100 ml volume were taken after 5, 10, 15 and 30 minutes.

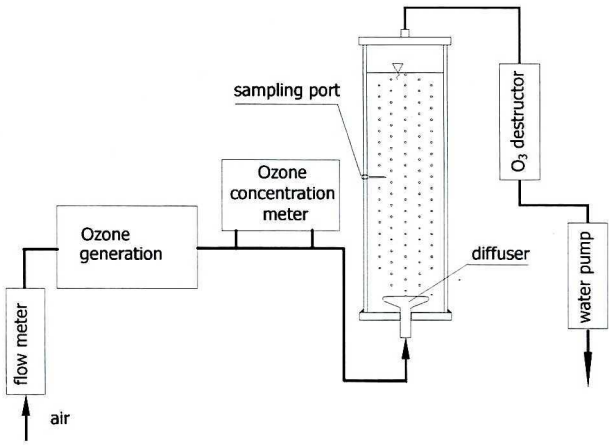

Fig. 1. Scheme of the laboratory test stand.

RESULTS AND DISCUSSION

Leachate characteristic

The values of COD shown in Table 1 correspond to the range known from the literature. The highest values were observed for the landfill in Rokitno. COD values are about seven times higher than those obtained for the landfill in Pulawy and nearly four times higher than for the leachate originating from Leczna.

Table 1. Characteristic of leachate originating from three examined landfills.

	Unit	Rokitno	Pulawy	Leczna
pH	-	6.9	6.8	7.1
COD_{Cr}	[mgO$_2$/dm^3]	7110 - 9610	806 - 2130	1400 – 2700
BOD_5	[mgO$_2$/dm^3]	1900 - 3950	200-700	200 - 900
$N\text{-}NH_4$	[mg/dm^3]	1800	2200	50
TOC	[mg/dm^3]	7200	405	25
Al	[mg/dm^3]	0.146	0.270	3.371
Cd	[mg/dm^3]	0.000	0.000	0.031
Co	[mg/dm^3]	0.086	0.015	0.050
Cr	[mg/dm^3]	0.122	0.182	0.181
Cu	[mg/dm^3]	0.006	0.042	0.032
Fe	[mg/dm^3]	158.166	7.921	81.675
Mn	[mg/dm^3]	4.499	0.951	12.004
Mo	[mg/dm^3]	0.000	0.000	0.029
Ni	[mg/dm^3]	0.433	0.104	0.267
Pb	[mg/dm^3]	0.031	0.000	0.139
Zn	[mg/dm^3]	1.006	0.263	5.953

The differences between COD values on the examined landfills result from the morphological composition of deposited wastes, as well as from the differing ages of the examined objects and, in turn, the different stages of chemical changes occurring in the wastes layer. Literature reports indicate that the highest COD values are characteristic of the acidic phase of waste fermentation. In the subsequent methanogenic phase the values of COD decrease (Reinhart and Al-Yousfi, 1996). In the case of the examined objects only the landfill in Leczna, because of its age, could be found in the acidic phase. However, the COD values in this case were lower than the characteristic values for this phase of biochemical changes of wastes known from the literature. The above observation may be explained by the low content of the substances, mainly organic, undergoing chemical decomposition. The organic carbon content was at its lowest in the Leczna landfill, while the leachate from Rokitno was characterized by an organic carbon content amounting to 7200 mgC/dm^3. Since the landfills in Pulawy and in Rokitno are older we assume that these landfills had undergone methanogenic phase.

The highest values of BOD_5 were obtained for the leachate from Rokitno landfill (Tab 1). The values were about five to six times higher than those measured in Pulawy and in Leczna. The values of BOD_5 in all cases were comparable to those known from the literature for the methanogenic phase (Reinhart and Al-Yousfi, 1996)

The highest value of the COD/BOD_5 ratio (ranged from 3.7 – 7.0) was measured in the leachate from Leczna. The above suggests the presence of a significant amount of decomposable inorganic compounds. The values of the ratio between COD and BOD_5 of the leachate from Rokitno and Pulawy were similar and ranged 2.5 - 4.4 and 3.2 – 5.3 respectively. The low value of this ratio in the leachate originating in Rokitno may result from the high proportion of organic substances in the leachate. According to El-Fadel et al (2002), it is possible to determine the age and biochemical stability of the landfill on the basis of the BOD_5/COD ratio. The ratio of BOD_5 to COD for all landfills fitted the range of 0.1 – 0.23, what proves that the age of the landfills is moderate and they are biochemically stable.

The values of ammonia nitrogen concentration in the leachate from the landfills in Rokitno and Pulawy correspond to the range known from the literature (Christensen et al., 2001). The lowest value of ammonia nitrogen concentration was observed in the leachate originating in the Leczna landfill. Such a low result may be explained by the fact that this landfill collects mainly wastes originating in villages, where the organic fraction is not deposited as frequently as elsewhere, but rather used as compost.

The highest concentrations of Al, Cd, Mn, Mo, Pb and Zn were observed in leachate from the Leczna landfill. The highest values of Fe, Co and Ni were noted in leachate from the Rokitno landfill. No detectible concentrations of Cd and Mo were observed in leachate from Rokitno and Pulawy landfills. The measured values of all heavy metals did not exceed the range of permissible concentration of heavy metals in the industrial wastewater discharged into the sewerage system (Journal of Laws, No 136, item 964), except for the Zn concentration in leachate from Leczna and the Fe concentration in leachate from Rokitno and Leczna.

The effect of ozonation on leachate quality

The results of ozonation show that the COD values in the leachate from three examined landfills decrease. In Figure 2 the values of COD before and after the oxidation process are presented.

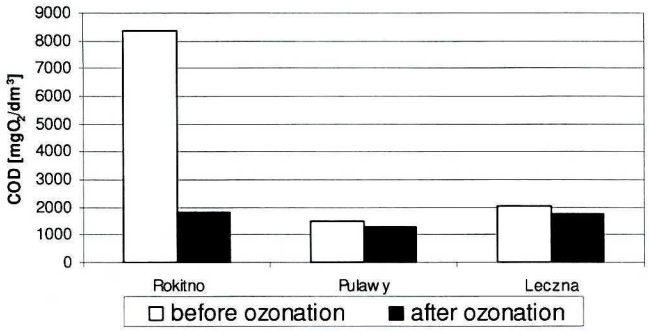

Fig. 2. Average COD values before and after 30 minutes ozonation. Dose: 1.8 g O_3/m^3.

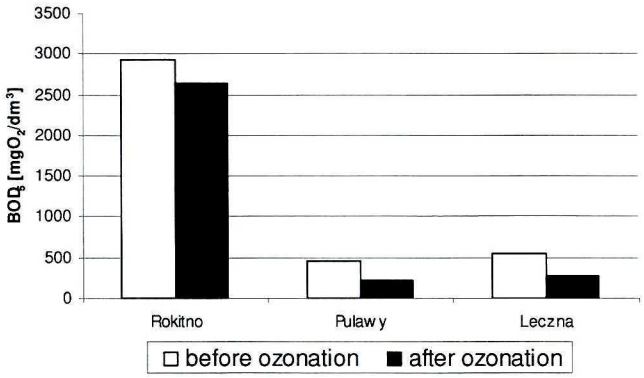

Fig. 3. Average BOD_5 values before and after 30 minutes ozonation. Dose: 1.8 g O_3/m^3.

Table 2. Average COD and BOD_5 values before and after ozonation.

	Unit	Rokitno		Pulawy		Leczna	
		before ozonation	after ozonation	before ozonation	after ozonation	before ozonation	after ozonation
COD_{Cr}	[mgO_2/dm^3]	8360	1839	1486	1263	2050	1742
BOD_5	[mgO_2/dm^3]	2925	2632	450	225	550	275

Ozonation appears to influence the COD and BOD_5 values on all landfills. Figures 2 and 3 depict the degree of BOD_5 and COD reduction. The highest reduction of COD (of about 78%) is observed for the leachate in Leczna where the initial value of COD was the lowest. The reduction of COD in leachate from Rokitno and Pulawy was similar and oscillated around 15%. The reduction of BOD_5 is similar for the landfills in Pulawy and Leczna (about 50%) but in the case of leachate from the Rokitno landfill the reduction was very low and amounted only to 10%.

The values of $N\text{-}NH_4$ after ozonation did not show significant changes. Marttinen et al. (2002) stated that ozonation process had no influence on the ammonia concentration, in agreement with the present work.

CONCLUSIONS

Our results allow us to conclude that ozonation is promising as a technique for biodegradation of organic substances present in leachate.

However, its effect was dependent on the initial composition of leachate determined by the landfill conditions. The highest degree of reduction in COD and BOD_5 was observed in cases when the initial values of these parameters were low.

The results indicate that waste segregation influences the leachate quality originating on the landfill. Separation of the organic fraction and recyclable wastes on the landfills at Pulawy and Leczna, causes reduction of BOD_5 and COD values. $N\text{-}NH_4$ and heavy metals concentrations were also decreased. The same parameters at the Rokitno landfill, where no segregation was carried out, are significantly higher.

The examined leachate originated from landfills of similar age, thus allowing comparison of their composition and to affirm the influence of segregation. Leachate was collected only in springtime. Samples were not collected in other seasons, which may affect the overall results because of the influence of temperature, rainfall etc.

The results of the present study allow the conclusion that increasing the degree of waste segregation on the landfill contributes to a more uniform waste mass, and this will help to design the processes of wastewater treatment plants more precisely.

Acknowledgement

This work was supported by the Ministry of Science and Higher Education of Poland (Ministerstwo Nauki i Szkolnictwa Wyższego), PBZ-MEiN-3/2/2006.

REFERENCES

[1] Al-Muzaini, S., Beg, M.B., Musimani, K., 1995. Characterization of landfill laechates at a waste disposal site in Kuwait. Environmental International. 22, 399-405.

[2] Bila, D.M., Montalvão, A.F., Silva, A.C., Dezotti, M., 2005. Ozonation of landfill leachate: evaluation of toxicity removal and biodegradability improvement. Journal of Hazardous Materials. B 117, 235-242.

[3] Christensen, T.H., Kjeldesen, P., Bjerg, P.L., Jensen, D.L., Christensen, J.B., Baun, A., Albrechtsen, H.J., Heron, G., 2001. Biogeochemistry of landfill leachate plumes. Applied Geochemistry. 16, 659-718.

[4] Directive 2006/12/EC of the European Parliament and of the Council of 5 April 2006 on waste.

[5] El-Fadel, M., Bou-Zeid, E., Chahine, W., 2002. Temporal variation of leachate quality from pre-sorted and baled municipal solid waste with high organic and moisture content. Waste Management. 22, 269-282.

[6] Irene, M., Lo, C., 1996. Characteristics and treatment of leachates from domestic landfills. Environmental International. 22, 433-442.

[7] Komilis, D.P., Ham, R.K., Stegmann, R., 1999. The effect of municipal solid waste pretreatment on landfill behavior: a literature review. Waste Management and Research. 17, 10-19.

[8] Leczna Commune Development Plan for years 2004 – 2013.
[9] http://www.leczna.pl/.../Z2Z4L2xlY3puYS9wbC9kZWZhdWx0X29waXN5LzI2LzEvMQ/planrozwoju_lokalnego_leczna_2004_2013.pdf, date of access April 2007.
[10] Leikam, K., Stegmann, R., 1999. Influence of mechanical-biological pretreatment of municipal solid waste on landfill behaviour. Waste Management and Research. 17, 424-429.
[11] Marttinen, S.K., Kettunen, R.H., Sormunen, K.M., Soimasuo, R.M., Rintala, J.A., 2002. Screening of physical – chemical methods for removal of organic material, nitrogen and toxicity from low strength landfill leachate. Chemosphere. 46, 851-585.
[12] Monje–Ramirez, I., Orta de Velásquez, M.T., 2004. Removal and transformation of recalcitrant organic matter from stabilized saline landfill leachates by coagulation-ozonation coupling processes. Water Research. 38, 2359-2367.
[13] Pilipczuk, I., Jakubowski, M., Szabatkiewicz, W., 2002. Ecological Review for Rokitno Landfill. MPWiK Lublin. unpublished data.
[14] Polish Water Law, Journal of Laws. No 115, item 1229.
[15] Pulawy Solid Waste Management Plan. 2004. unpublished data.
[16] Reinhart, D.R., Al-Yousfi, A.B., 1996. The impact of leachate recirculation on municipal solid waste landfill operating characteristics. Waste Management and Research. 14, 337-346.
[17] Robinson, H.D., Knox, K., Bone, B.D., Picken, A., 2005. Leachate quality from landfilled MBT waste. Waste Management. 25, 383-391.
[18] The permissible concentrations of heavy metals in the industrial wastewater discharged into sewerage systems. Journal of Laws. No 136, item 964.
[19] Ward, M.L., Bitton, G., Towsend, T., 2005. Heavy metals binding capacity (HMBC) of municipal solid waste landfill leachates. Chemosphere. 60, 206-215.

Received: September, 2007; accepted: June, 2008.