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Impact of concentrated leachate recirculation on effectiveness of leachate treatment by reverse osmosis

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ABSTRACT

The effect of concentrate recirculation on leachate quality and effectiveness of their treatment by reverse osmosis (RO) was investigated. The concentrate from reverse osmosis was characterized by a high concentration of sulphides (average of 828 μg/l), sulphates (1898 mg/l), chlorides (5608 mg/l), electro-conductivity (8066 μS/cm), moderate chemical oxygen demand (COD) value (1646 mg/l) and low pH (6.7). Analysis of the leachate quality data showed that concentrate recirculation landfill produces stronger leachate especially in terms of inorganic compounds. Concentrate recirculation accelerated waste decomposition, therefore the concentration of COD, biochemical oxygen demand (BOD), ammonia nitrogen and COD/COD_{max} ratio value in leachate increased in first three months of recirculation. Changes in the leachate quality can influence the effectiveness of leachate treatment by RO. Correlation analysis has shown a negative correlation (−0.75) between iron concentration and COD removal, which is caused by fouling and bio-fouling induced by Fe. A negative correlation between the leachate pH and BOD removal (−0.78) was also observed, which was the result of electrostatic effects between the membrane surface and macromolecules of some organics.

1. Introduction

Disposal of leachate is recognized as one of the most difficult task associated with the landfill operation. Different on- and off-site treatments of the leachate are nowadays proposed and adopted to meet different requirements of each landfill (Wiszniowski et al., 2006; Renou et al., 2008a,b; Calabrò et al., 2010). A conventional biological process could be effective for the removal of organic substances, suspended solids and nutrients (Yahmed et al., 2009; Theepharaksapan et al., 2011). To remove recalcitrant compounds advanced treatment processes such physico-chemical and membrane technologies are required (Kurniawan et al., 2006; Theepharaksapan et al., 2011; Ahmed and Lan, 2012; Smol et al., 2014). Reverse osmosis seems to be one of the most promising methods among the new processes for landfill leachate treatment. Several studies were performed, both at lab and on industrial scale, to investigate RO performances on the separation of pollutants from landfill leachate (Bodzek et al., 2006; Chan et al., 2006; Liu et al., 2008; Li et al., 2009; Theepharaksapan et al., 2011; Smol et al., 2015). One of the most debated options, when the RO treatment is adopted, is a recirculation of concentrated leachate (Renou et al., 2008a,b; Liu et al., 2008; Li et al., 2009). Opinions on the consequence of this practice are not unanimous and specific studies present in a scientific literature are rare. According to Bilgili et al. (2007) the advantages of leachate recirculation include distribution of nutrient and enzymes, pH buffering, dilution of inhibitory compounds, liquid storage and evaporation opportunities. Sponza and Ağdağ (2004) reported that the leachate recirculation also shortens the time required for stabilization from several decades to 2–3 years. On the other hand Ledakowicz and Kaczarek (2002) maintain that leachate recirculation can lead to the inhibition of methanogenesis as it may cause a high concentration of organic acids which are toxic for methanogenesis. The concentrated leachate recirculation can change the leachate quality, causing an increase in the concentration of inorganic compounds (due to cyclically leaching/washout) and organic compounds (due to enhanced biodegradation processes in waste body). This can result in a change of the RO effectiveness, as it is limited among other things by fouling and scaling. Organic constituents contribute directly to organic fouling or provide carbon sources for the development of biofilms on the membrane surfaces (Wend et al., 2003). Inorganic compounds can cause gradual build-up on the membrane surface, which may consist of colloidal particles, iron and/or manganese oxides or may be due to inorganic scale formation (Ameen et al., 2011). Scaling is a result of deposition of particles on a membrane, causing it to plug. It

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occurs due to precipitation of sparingly soluble gas, such as calcium carbonate, calcium sulfate, barium sulfate and strontium sulfate.

Heavily contaminated leachate from a concentrate recirculation landfill can disturb the RO operation and be conducive to the process of fouling and scaling. The membrane fouling and scaling with a consequent reduction in a specific flux can lead to changes in the removal rate and may be a limitation of the membrane filtration process. In spite of many experimental works reported in the literature, at lab and on industrial scale, a systematic investigation of the impact of the RO concentrate recirculation on the RO performance has not yet been performed. The objective of this study is to evaluate the impact of the concentrated leachate recirculation on the effectiveness of the RO process in relation to selected contaminants.

2. Materials and methods

2.1. Landfill characteristics

Samples for analysis were taken from municipal landfill in Warminsko-Mazurskie Voivodeship, in north-eastern part of Poland. The landfill began its operation in 1983 opening of a 7.7 ha Area A for waste disposal. The Area A was not equipped with the liner system. This landfill cell is sealed with a natural 31-m clay substrate functioning as a geological barrier. In order to protect from uncontrolled leachate migration a circumferential ditch – around scarp foot of the Area A – was built. After 28 years of landfilling on the Area A it was closed and reclaimed. Then a new Area B of 5.1 ha started its operation in 2011. Area B was constructed with 2 mm geomembrane and leachate collection system. Table 1 summarizes the main properties of Area A and Area B.

In order to solve the leachate problem a reverse osmosis (RO) system for leachate treatment was implemented at the end of 2012 year. Leachate from both A and B areas of the landfill are directed to the buffer tank with total volume of 370 m^3 and mixed there. Pressurized leachate from the buffer tank is fed into the ROCHEM CD9-RO disc-tube modules with the amount of 1500 m^3 per month. Before entering into the reverse osmosis module sulphuric acid (98%) is dosed to the leachate to maintain the pH value at 6.0–6.5 and to increase the solubility of inorganic salts. Then, the leachate is fed into a sand filter, which is used to remove suspended particles larger than 50 μm in size. Having passed the sand filter, leachate is directed into a cartridge filter, which is applied for a further removal of suspended solids larger than 10 μm.

The leachate treatment installation by the RO process is equipped with a thin-film composite membrane made of polyamide. The system works under an operating pressure of 65 bar, at a temperature of 0–45 °C. The filtration area is 9 m^2 with a package 194 m^2/m^3. The membrane flux is up to 50 l/m^2 h. The capacity of this system is 72 m^3/day with a recovery rate of 75%. Salt rejection rate is 99%. To ensure a long-term membrane life and operation a procedure of membrane cleaning is launched in case of permeate flow decrease by 10–15%, decrease of permeate quality or increase of applied pressure by about 10–15%.

The generated liquid concentrate from the RO system is recirculated and sprinkled onto the surface of the Area B to ensure proper humidity of the waste and to enhance the waste degradation. The produced concentrated leachate represents about 25% of the total incoming leachate, i.e. up to 375 m^3 per month.

The climate in this region is continental with close to 50% of rainfall occurring between May and August. The average rainfall is about 619 mm per year, the average annual temperature is 7 °C.

Table 1
Properties of Area A and Area B.

	Area A	Area B
Cell opened	1983	2011
Cell closed	2010	Exploited
Area (ha)	7.7	5.1
Final mass (Mg)	550 000	280 000
Time of exploitation	28 years	2 years
Time of concentrated leachate recirculation	–	1 year
Total volume of recirculated concentrate (m^3/r)	–	1990
Volume of generated leachate (m^3/r)	7000	33 000

The average amount of the leachate produced per day is 60–90 m^3.

2.2. Leachate collection and analysis

In order to assess the effect of concentrate recirculation on the leachate quality and the RO effectiveness, leachate, permeate and concentrate samples have been collected. The samples were collected eight times, from February 2013 (directly after starting the RO process) till April 2014.

During sampling a raw leachate from the Cell B, permeate and concentrate from the concentrate tank were taken. Collected samples were immediately transported to the laboratory and analyzed within 48 h. Three groups of parameters were determined: (a) general parameters such as the pH, the electroconductivity (EC) and total suspended solids (TSS); (b) organic indicators such as the biochemical oxygen demand (BOD_5), the chemical oxygen demand (COD), BOD/COD ratio, (c) inorganic parameters such as the concentration of nitrogen ammonia ($N\text{-}NH_4$), cyanide (CN^-), chloride (Cl^-), sulphates (SO_4^-), sulphides (S^-), boron (B) and iron (Fe).

The electroconductivity and the pH were measured on-site by a conductivity and potentiometric method, respectively, using a portable pH meter (HACH HQ40). Total suspended solids were determined by a mass balance method after a well-mixed sample filtration through a FILTRAK cellulose fiber filter, and the residue retained on the filter was dried for a 1 h at 103–105 °C in drying oven (to constant weight). The chemical oxygen demand was analyzed using a calorimetric method with a HACH spectrophotometer (620 nm) after a 2-h reactor digestion (a $K_2Cr_2O_7$ method) and the biochemical oxygen demand – using an OxiTop (WTW) measuring system based on a pressure measurement.

Cyanide was determined by a pyridine-pyrazolone method. The results were measured with a HACH spectrophotometer at 612 nm. Chloride was analyzed by an iron(III)-thiocyanate method (468 nm), sulphates – by a barium sulphate method (450 nm), sulphides – by a methylene blue method (665 nm), and ferrous – by a phenanthroline method (510 nm), where for all these determinations a HACH spectrophotometer was used. Boron was measured by its reaction with carminic acid in the presence of sulfuric acid (605 nm) using the HACH spectrophotometer. Nitrogen ammonia was determined with N Tube Vials on the HACH spectrophotometer (655 nm) using a salicylate method. The obtained results were the mean value of three determinations carried out simultaneously.

For data analysis a Statistica software was used in this study. The basic statistic analysis included calculation of minimum, maximum and mean value. Measures of variability were reported in standard deviation. To assess the effectiveness of RO a removal ratio was calculated for each contaminant. For detailed analysis an additional parameters were calculated such as BOD/COD ratio and COD/COD_{max} ratio.

3. Result and discussion

3.1. Concentrate and leachate characteristics

The investigation on leachate and concentrate composition from analyzed landfill were started just after the RO process started and