# IMPACT ASSESSMENT OF CONCENTRATE RE-CIRCULATION ON THE LANDFILL GAS PRODUCTION

by

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Original scientific paper
DOI: 10.2298/TSCI160401137D

*This paper explores the impact of concentrate re-circulation, as a product of leachate treated by reverse osmosis plant, on the production of landfill gas at the real-scale landfill for municipal solid waste. In an effort to come up with results, experimental measurements were carried out at the landfill in city of Bijeljina, Republic of Srpska, Bosnia and Herzegovina. All performed measurements were divided into three groups. The aims of two groups of measurement were to determine landfill gas and methane yield from concentrate, and leachate in laboratory conditions (1st group), and to find out concentrations of oxidizing matters (chemical and biochemical oxygen demand) present in leachate and concentrate at different points of treatment as well as its variability over the time (2nd group), which could be used to calculate the potential of landfill gas and methane generation from concentrate by re-circulation, theoretically. The 3rd group of measurements, carried out in parallel, has goal to determine the quality and quantity of the collected landfill gas at wells throughout the landfill. The results of analysis carried out in this experimental research show the clear evidence of concentrate re-circulation impact on methane production by increasing the landfill gas flow, as well as its concentration within the landfill gas composition, at the nearby well. Although results indicated relatively high impact of concentrate re-circulation on landfill gas production, comparing to its theoretical potential, the influence on the landfill at whole, is negligible, due to relatively low volumes in re-circulation with respect to its size and objectively low potential given by organic matter present in concentrate.*

Key words: *reverse osmosis, concentrate re-circulation, methane production, renewable energy, landfill gas wells*

## Introduction

Leachate re-circulation has proven to be the most effective method of increasing biodegradation in order to achieve higher methane yields and lower costs of treatment, so the numerous researches have been done as a combination with the application of leachate re-circulation and some other methods [1]. As a general advantages of re-circulation, concerning the production of landfill gas (LFG) and leachate treatment, proven by laboratory research, are: the creation of significantly larger amounts of leachate due to faster degradation of waste, generating significantly larger quantities of LFG with a something higher methane concentration, faster enter the methanogenesis stage that favors rapid development of methanogenic bacteria and reaching peak production of methane from waste into simulated anaerobic biore-

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actors, improving quality and reducing the volume of leachate for further treatment, faster leachate and waste stabilization. Ability to remove oxidizing matter (chemical oxygen demand – COD) of leachate during the methanogenesis stage, allows the use of landfill leachate re-circulation for leachate treatment, achieving additional LFG production [2-6]. About 50% more oxidizing substances in the leachate and LFG was achieved by lecheate re-circulation in the case of fresh then the eight years old waste, unchanged composition, while the share of oxidizing matter released in leachate, by applying re-circulation, was 27% and 3.2%, in the case of fresh and old waste, respectively. Finally, it was found that 82% to 89%, out of the total quantity of oxidizing matter, release in the form of gas [3]. High efficiency of bioreactor column in which was simulated anaerobic landfill with the addition of water (analogous to precipitation in real terms) and the re-circulation, in the methanogenesis stage showed high removal efficiency of COD, which is close to 95%, while the least dependent on the COD load that is entered by leachate, as opposed to the columns of a simulated semi-aerobic conditions, as well as anaerobic but only with the addition of water without re-circulation. Effect of leachate re-circulation, in terms of its purification, is maximized when the landfill reaches a stable phase and it is proved useful to speed up the cultivation and propagation of bacterial activity on the landfill and its ability to support the treatment [4].

Laboratory research have also shown that increase of re-circulation by flow and frequency allows the generation of larger amounts of LFG, but to a certain limit, beyond which can cause a acidic environment condition due to excessive rates of hydrolysis of organic matter, which can further slow down the activity of methanogenic bacteria, significantly reducing the amount of LFG, and the concentration of methane [5, 6].

Generally, the possibility of leachate treatment by using biological processes depends on its biodegradability. The ratio between the main biological ($BOD_5$ – biological oxygen demand) and chemical characteristics of pollution, expressed as $BOD_5$/COD, which is usually used as a measure of biodegradability, tends to reduce with the age of the landfill, ranging from 0.5 for young to 0.2, and less for older and more stable leachate. Young leachate is characterized by higher proportions of organic biodegradable material which has high COD value and additionally high share of volatile fatty acids in the total organic carbon, which are with short chains with a low molecular mass and high ability rate of anaerobic degradation. As leachate is getting older, increases the share of hard-soluble and resistant to degradation, humic and fulvic acids, along with the decline of biodegradability. On the other hand, lower values of the ratio $BOD_5$/COD indicate that landfill is in methanogenesis stage [7].

Disposal of concentrate by re-circulation in the landfill is one way for its treatment and certainly the least expensive option, but the option which split expert public opinion when it comes to the consequences of its application. There is relatively little scientific research on this topic. According to a study [8] the impact on the quality of the concentrate re-circulation of leachate is insignificant or limited to the period immediately after the start of the re-circulation, while according to the another research [9] the long-term re-circulation of concentrate expressed unfavorable due to negative effects on the production of leachate. Study of the impact of the concentrate re-circulation, coming from the treatment by reverse osmosis, on mineral and organic compounds in leachate from landfill showed that, after an initial period of re-circulation, concentration of organic compounds (BOD*,* COD) increases, in order to stabilize after six months. This indicates that the re-circulation of concentrate accelerates decomposition of organic matter, and this was particularly noticeable on the landfill cell with more fresh waste [10].

One of the biggest challenges of effective re-circulation in the field is uniform distribution of moisture within the waste at the landfill. The expected LFG emissions from the leachate at real scale landfill are significantly lower than in laboratory conditions, mainly because of channel flow through the waste which occurs at high speed and results in wetting of waste to a much lesser extent. Desired leachate trajectory is caused by differences in the local hydraulic conductivity of the waste and daily coverings that are used [11]. By exploring the impact of the flow of water through the waste of the same characteristics, but different sizes, it is shown that under laboratory conditions about 40% of the pore water participates in the transport of solutes, while the proportion is less than 0.2% of the observed real scale landfill [12].

Under stabilized conditions in methanogenesis stage, LFG contains on average 55-60% methane and 40-45% carbon dioxide with quantities of other gases in traces [13]. In open dumps methane concentration is much lower and one of the most variable factors is the outside temperature, which, with a low depth and age of waste, due to inadequate protection systems and thermal isolation, affects large seasonally and daily, as well as spacious, variations in the concentration of methane in the landfill. For example, in open dump at city of Novi Sad, Serbia, methane concentrations range from 0% to 34%, with a decrease in the concentrations to 0% during the winter season [14], while in a closed landfill dump Morelia in Mexico, based on the one-year monitoring period of 16 different locations and 49 sampling stations, average methane concentration was 45.5% [15].

The hypothesis of this study is that re-circulation of concentrate have impact on increase of methane quantity production at the landfill. In order to prove this, the main objective of this research is to link results, obtained by combining experimental measurements and theoretical calculations in order to determine concentrate LFG potential and the measurements of the actual LFG values at wells. By careful analysis, LFG production and its variability over the time should be isolated from other influential parameters that could affect it. A special benefit of this study is that it focuses on monitoring and measuring the parameters on the real scale landfill, what is lacking in the literature, unlike many of the laboratory and pilot plant researches, carried out under controlled conditions in simulators. Also, the literature reviewed did not highlight research on the real scale landfill that quantifies the effect of concentrate re-circulation on LFG production, as it is done here. Landfill, which is the subject of investigation, is the only one in the region, which has built an active LFG collection system and one of the rare with modern leachate treatment plant.

## Characteristics of landfill in Bijeljina

Landfill *Brijesnica* is located in the western part of the municipality of Bijeljina, and is at a distance of about 2 km from the outskirts of town (east side) and about 1.5 km from the nearest village to the northwest side and west side. Landfill has been in operation from the beginning of 2010, and so far deposited a more than 180,000 tons of municipal solid waste. Active landfill cells covers about 4 ha, while the available capacity of the landfill include additional two cells of about 4 ha and 3 ha, which construction is planned in the future.

Leachate is accepted in the landfill by drainage pipe system which consists of perforated polyvinyl chloride drainage pipes and layer from a natural mixture of sand and gravel at the bottom of the landfill body and drained to the receiving pool. After partial aeration of leachate, which leads to the oxidation of dissolved solids and reducing the part of organic pollution ($BOD_5$, COD) and deposition of larger suspended particles, it flows into an equalization pool and is pumped to the plant for leachate treatment by reverse osmosis (RO). The concentrate, polluted part and rest of the leachate treatment by membranes of RO, is re-circulated back into the active landfill cell.

The LFG combustion is done in an enclosed flare, after its collection by wells and transportation to the main collector. Most important measurements that are continuously done and read during regular inspection at the flare are LFG flow and its methane and oxygen concentration.

According to the described construction of the landfill and integrated system that facilitates the generation and management of leachate and LFG emissions, landfill in Bijeljina could be defined more as *controlled* than only sanitary landfill. Distribution of leachate and concentrate at the landfill is carried out directly with the outpouring of manholes via a flexible hose, which is far less favorable solution compared to typical cases which are applied in practice of bioreactor landfills.

## Measurement methodology

Experimental measurements conducted for this research, are divided into three groups. The first two groups present laboratory measurements, performed on samples of leachate and concentrate, while the third group regards the determination of the composition and flow of the LFG wells. Figure 1 shows a flowchart scheme of measuring points within the treatment plant. Sampling of leachate was conducted at the following measuring points: V100 – leachate on the outlet pipe, which comes directly from the landfill, V131 – leachate at the exit of the equalization basin and the entrance to the treatment plant, V151 – leachate after conditioning due to lower pH values, V171 – concentrate from the process of RO, V180 – permeate at the exit of the RO treatment plant, V195 – conditioned permeate with pH value close to neutral, ready for disposal in waterways.

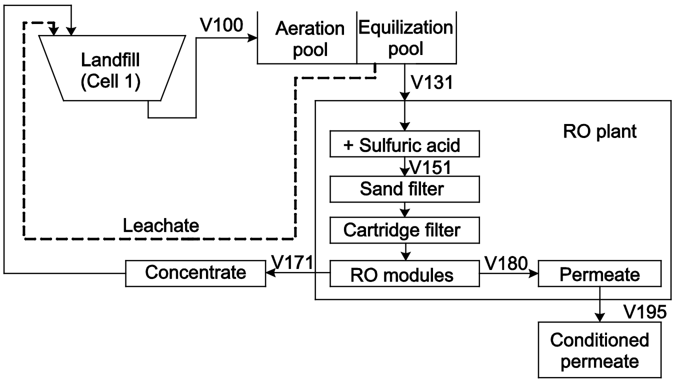


**Figure 1. Flowchart scheme of the leachate treatment plant with the sampling points**

The analysis from the first set of measurements aimed at determining the experimental LFG yield as well as to find out the degree of COD degradation and organic matter content. This analysis was done for samples obtained from measuring points V131 and V171, using method VDI 4630 [16]. Sampling was carried out on 3$^{rd}$ of December in 2015, with measurements of biogas generated in the period of one month.

The analyses in the context of the second set of measurements were performed for laboratory determination of oxidizing matter in leachate, COD, and $BOD_5$. Sampling, which covered two cycles of the RO treatment plant, was done in the period from 3$^{rd}$ to 10$^{th}$ of December in 2015, with the collection of samples on a daily basis. The ultimate objectives of this analysis were to find out concentrate biochemical degradability based on ratio $BOD_5$/COD, as well as real COD values, and its variability over time in order to assess possible LFG potential from concentrates.

The third group of measurements, concerning LFG composition and flow rate, was carried out during the period from 4$^{th}$ to 10$^{th}$ of December in 2015, as well as on 24$^{th}$ of February in 2016.

Figure 2 provides a satellite image of the active cell of the landfill with the schedule of LFG wells where measurements were performed, while the schematic view of the structure of a typical LFG well is given in fig. 3. Sampling of LFG is made through the check valve located at the outlet pipe of LFG well (fig. 3). Wells marked in grey color could not be used for measurements due to the absence of the control valve (B-6, 16, 18, 19) or because some of the wells were disconnected from the central system of LFG collecting due to waste disposal in the immediate vicinity (B-6, 13). The composition of LFG was measured by a gas analyzer, type: Gem2000 Plus, previously calibrated in the laboratory.

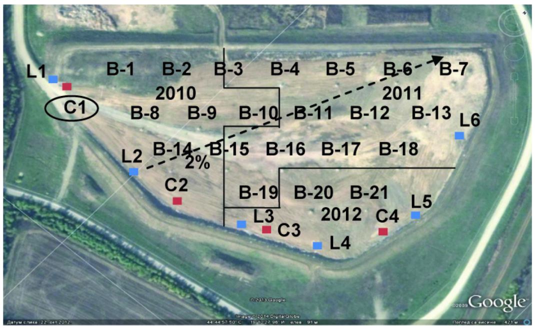


**Figure 2. Schedule of LFG wells, outflows and drainage pipe**

Given that there was no possibility for the direct measurement of LFG flow rate within the well, measurements were carried out by measuring LFG speed at the exit of the control valve when it is fully open. These measurements can provide relationship of the gas flow intensity at the wells, thanks to the fact that their geometry is identical. Speed measurements were performed using a gas appliance, type: Testo 435-1. Measurements showed that the measured gas flow at the wells varies within wide limits due to hyper-sensitivity of the measuring instrument, but also a small outlet opening, so that one could not speak of the great measurement accuracy. Additionally, the LFG flow control is performed manually via the butterfly valve, in a way that wells closer to the main pipeline and flare have valve partially open, while wells that are more distant have valve fully open, in order to achieve more uniform pressure conditions. The objectives of the third group of measurements were to assess viability of metanogenic conditions throughout the landfill and over the time, and according to analysis, which take into account local conditions and waste characteristics (composition, age, depth, temporary coverings), to find out the potential impact of concentrate re-circulation on the methane production.

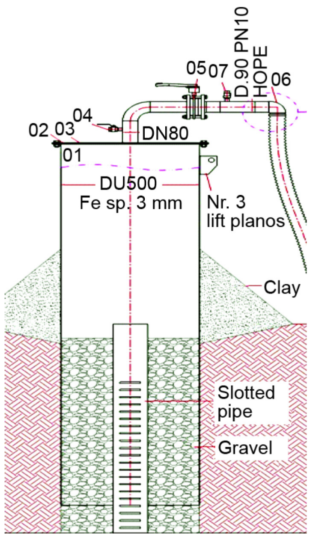


**Figure 3. Scheme of LFG well**

## Analysis of the results

### *Laboratory measurements*

The results of analysis, which belong to the first group of measurements, showed that the leachate and concentrate samples are characterized by relatively low dry matter (DM) content and organic dry matter (ODM), fraction subject to degradation and biogas production, which was 0.5% and 0.13% for the leachate (V131) and 1.75% and 0.41% for the concentrate (V171). Relationship between DM and ODM represents the concentration factor, expressed by the ratio of volume of leachate entering the treatment plant and the concentrate that comes out of the plant, multiplied by the organic matter removal efficiency of the plant. According to 2$^{nd}$ group of measurements, average removal efficiency of conductivity and COD values amount to 97.4% and 99.4%, respectively. The results were correlated with results obtained in a similar survey conducted in real conditions of the landfill, where the efficiency of removing pollutants for eight consecutive cycles in RO treatment plant were from 96.3% to 98.2% and from 92.8% to 99.9% for conductivity and COD, respectively [17].

Due to the extremely low content of ODM, it was not possible to set-up an experiment in accordance with the applied method [16], as inadequate volumes of used reactor, where is going exactly the prescribed quantity of ODM sample and inoculum that initiates the degradation. Even after minor modifications to the experimental method used, results showed that the yield of biogas not exists and that it is not possible to produce biogas from that substrate.

As it is also shown by the 2nd group of measurements, average COD value of the leachate from landfill (V100) and concentrate (V171) samples were only 1,347 $gO_2/m^3$ and 3,055 $gO_2/m^3$, respectively. It is very low comparing, for example, to the average COD values of wastewaters of food industry in province of Vojvodina, Serbia, which amounts from 29.33 $gO_2/m^3$ to 995.41 $gO_2/m^3$ in production of cocoa, chocolate and confectionery products to production of starches and starch products, respectively [18].

Based on the values of the indicator $BOD_5$/COD, given in tab. 1, leachate, which comes from the landfill, could be classified as middle-aged [7], while low values of COD and $BOD_5$ concentrations are closer to the old landfills [19]. The obtained values indicate that the biodegradability of concentrate is higher than from the leachate coming from the landfill, which, along with increased BOD and COD values, means greater potential to generate methane.

**Table 1. The parameters of organic pollution leachate and indicator $BOD_5$/COD**

| Parameter | Sampling data | Measurement points with description | | | |
|---|---|---|---|---|---|
| | | V100 | V131 | V151 | V171 |
| | | Outlet pipe | Inlet to plant | Inlet to module | Concentrate |
| COD [$gm^{-3}$] | December 3, 2015 | 1,328 | 995 | 1,000 | 2,366 |
| $BOD_5$ [$gm^{-3}$] | | 394 | 224 | 604 | 900 |
| **$BOD_5$/COD [–]** | | **0.30** | **0.23** | **0,60** | **0.38** |
| COD [$gm^{-3}$] | December 9, 2015 | 1,511 | 1,326 | 1,325 | 3,458 |
| $BOD_5$ [$gm^{-3}$] | | 314 | 850 | 930 | 1.016 |
| **$BOD_5$/COD [–]** | | **0.21** | **0.64** | **0.70** | **0.29** |

Equalizing the stoichiometric oxidation reaction of methane it turns out that the oxidation of 1 mole of methane requires 2 moles of oxygen molecules, which ultimately provides a balance that from 1 g COD it is possible to obtain 350 ml of methane. As it is known from practical experience that about 10% of the load converted oxidizing substances is spent on the creation of new biomass, it can be assumed that under practical conditions of complete degradation of 1 g COD could be produced about 320 ml of methane [16].

On the basis of the known COD value of concentrate, it is possible to calculate the amount of biogas or LFG, that could be theoretically obtained [16], using the expression:

$$D_{\mathrm{COD}} = \frac{V_{\mathrm{LFG}} x_{\mathrm{CH_4}}}{320\, m_{\mathrm{c}}\, \mathrm{COD}} 100 \tag{1}$$

where $D_{COD}$ [%] is the degree of COD degradation, $V_{LPG}$ [$ml_N CH_4 g^{-1}$] – the volume of LFG, $x_{CH_4}$ [%] – the proportion of methane in biogas, $m_c$ [g] – the mass of concentrate, and COD [$mgO_2 l^{-1}$] – the chemichal oxygen demand of concentrate.

Another influence of concentrate re-circulation on methane production could be seen from the pH value disturbance of concentrate and leachate that come from the landfill. Figures 4 and 5 show the change conductivity *EC* and pH value for the leachate, concentrate and permeate since the plant was put in operation. The increase in conductivity with concentrate re-circulation, which was followed by reduction of pH values, is in accordance with the results obtained in the study [10].

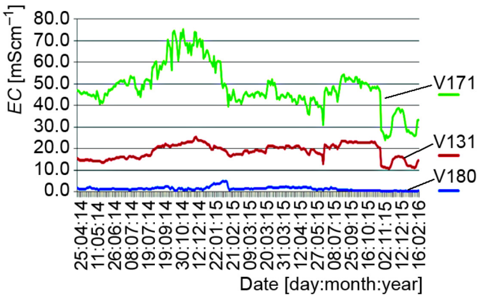


**Figure 4. Conductivity changes over time**

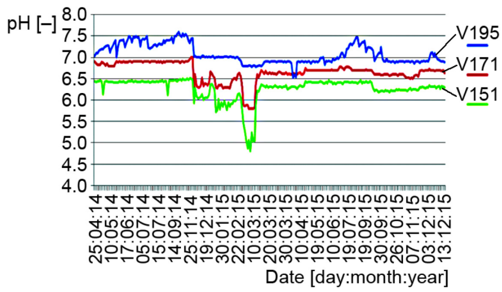


**Figure 5. The pH value changes over time**

Changing of the leachate pH values, based on measurements at the outlet pipe from the landfill during the regular quarterly laboratory analysis, is visible in fig. 6. It could be noted a significant drop in pH value of the leachate from the average value of about 9 to the value of 6.56 in the 4$^{th}$ quarter in year 2014 (laboratory analysis performed on 28$^{th}$ November in 2014), which could be consequence of mentioned drop in concentrate pH value. Return of leachate pH value in the next quarter of over 9 and remaining on this level afterwards, is also in line with return of concentrate pH value to a higher level.

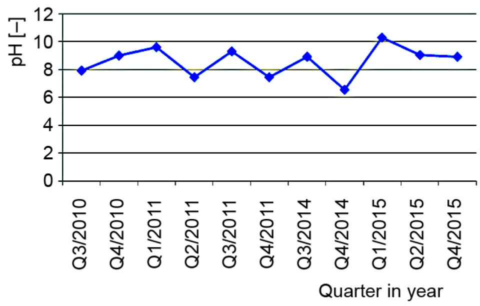


**Figure 6. Leachate pH value changes over time**

Decrease of pH value could be also due to the stimulation of the hydrolytic and fermentative bacteria in the deposited waste, which have resulted in carboxylic acids. In younger landfills the process of dissolution and mineralization occurs to a significant extent as a result of intensive transformations which are characteristic for an initial period of landfill operation (acidogenesis stage) and the re-circulation of concentrate [10].

The imbalance of methanogenic and acidogenic bacteria could affect the possible reduction of LFG production over time due to existence of acidogenic conditions, probably in part of the landfilled waste, which is in opposite to theoretical LFG potential from the concentrate. The optimum level, stated in the literature as favorable for methanogenesis, is in the range of 6.4 to 7.2 [20], while authors [21] take the optimal pH range for the real-scale landfill between 7.5 and 9.0, giving especially bad marks on the methane production influence for pH values below 7.2. As it could be seen from fig. 7, noticeably low LFG flow from landfill occur during the

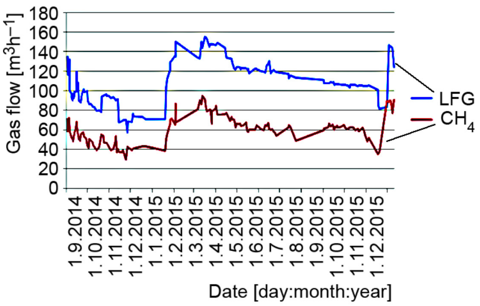


**Figure 7. The LFG and $CH_4$ changes over time**

period of pH decrease, although precipitation, as one of the most influencing parameters were similar during the same period in 2015.

Substituting the average COD value of the concentrate, obtained during the experimental measurements, results in the methane potential from the concentrate of 1.0 $m^3CH_4/m^3$ concentrate. It is theoretical potential which could be achieved only if entire COD quantity is involved in methane production. The real impact of concentrate on methane production depends on real biochemical degradability of concentrate, methanogenic conditions being set-up within the landfill and macroscopic concentrate flow through the waste at the landfill. Ratio between real and potential methane quantities could be defined as COD removal efficiency.

### *The LFG measurement*

The measured values of the methane concentration and average LFG velocity at the outlet of wells are shown in tab. 2. The obtained values show the following: (1) concentration of methane in all wells are quite high, with total average values from 60.5% to 62.6%, which

**Table 2. Measured concentrations of methane and flow of LFG through wells**

| Well code | Measurement date | | | | | Average $CH_4$ [%] | $v$ [$ms^{-1}$] | $H$ [m] | $H/v$ [–] |
|---|---|---|---|---|---|---|---|---|---|
| | 12/4/15 | 12/6/15 | 12/8/15 | 12/10/15 | 02/24/16 | | | | |
| B-1 | 64.1% | 64.1% | 64.3% | 64.3% | 64.5% | 64.3% | 2.2 | 4.5 | 0.48 |
| B-2 | 63.0% | 62.9% | 63.3% | 63.3% | 63.2% | 63.1% | 2.7 | 4.8 | 0.56 |
| B-3 | 62.8% | 62.9% | 63.2% | 63.0% | 63.6% | 63.1% | 2.0 | 5.1 | 0.38 |
| B-4 | 63.2% | 63.0% | 63.1% | 62.9% | 50.6% | 60.6% | 0.8 | 5.4 | 0.14 |
| B-5 | 63.2% | 63.3% | 63.6% | 63.4% | – | 63.4% | 4.3 | 5.7 | 0.75 |
| B-7 | 60.1% | 60.3% | 60.9% | – | 58.2% | 59.9% | 1.7 | 10.0 | 0.17 |
| B-8 | 63.0% | 63.3% | 63.3% | 63.4% | 63.5% | 63.3% | 2.0 | 5.5 | 0.36 |
| B-9 | 63.1% | 63.3% | 63.5% | 63.4% | 63.8% | 63.4% | 2.7 | 5.0 | 0.53 |
| B-10 | 60.7% | 61.7% | 60.5% | 59.2% | 63.4% | 61.1% | 1.1 | 5.3 | 0.20 |
| B-11 | 63.2% | 64.1% | 64.5% | 63.5% | 48.1% | 60.7% | 2.3 | 5.7 | 0.39 |
| B-12 | 61.5% | – | – | – | – | 61.5% | – | 6.0 | – |
| B-14 | 62.9% | 63.2% | 63.1% | 37.4% | 63.1% | 57.9% | 1.0 | 6.0 | 0.17 |
| B-15 | 59.1% | 59.4% | 60.0% | 59.4% | 60.5% | 59.7% | 2.0 | 6.5 | 0.31 |
| B-17 | 61.6% | 61.2% | 61.7% | 62.2% | 61.1% | 61.6% | – | 7.5 | – |
| B-20 | 63.1% | 62.4% | 62.7% | 62.7% | 62.4% | 62.7% | 2.3 | 6.3 | 0.36 |
| B-21 | 61.1% | 61.3% | 61.6% | 61.5% | 61.1% | 61.3% | 2.4 | 6.6 | 0.36 |
| Average: | 62.2% | 62.4% | 62.6% | 60.7% | 60.5% | 61.7% | 2.1 | 6.5 | 0.37 |

$v$ – gas speed at control valve, $H$ – waste height; date format: month/date/year is well above the literature values [13], (2) the concentration of methane are fairly uniform throughout the surface of the landfill, and (3) there are very small variations of methane concentration in the wells during the time. The drastic decline in the concentration of methane, which is recorded in several wells, resulted in the temporarily intake of external air through the waste near the wells. These data indicate that the entire landfill is actually in a stable methanogenic stage. Figure 2 shows imaginary lines which divide the landfill into areas depending on the calendar year when the first layer of waste was deposited. As it could be seen, waste was buried in that way which results in different height within the landfill. At the part with the oldest waste, which has the highest altitude, disposed waste is only 4 m to 5 m height, while it reaches about 10 m in parts with the lowest altitude.

Results of the average values of methane concentration depending on the age of waste are: 63.4%, 61.1%, and 61.8% of methane in parts of landfill where waste disposal started in 2010, 2011, and 2012, respectively. This clearly indicates that the older parts of the landfilled waste give slightly higher methane concentrations. It could be seen that the amount of waste, according to its height, does not follow an increase in LFG flow rate at the wells. The LFG at well B-5 had convincingly the highest speed, and the reason could be found in the fact that the well at the time of measurement was surrounded by puddles of water. This fact could affect humidity of waste locally, and therefore, provide a greater LFG production.

Since starting the operation of RO plant, only chamber C-2 was exploited, which has been substituted for C-1 just before the first measurement. Concentrate is transported through the hose and disposed inside the trench, made in the waste surface, dimension $A \times B \times H = 1\ \mathrm{m} \times 0.5\ \mathrm{m} \times 1\ \mathrm{m}$, at a distance of about 15 m from the well B-1. As shown in fig. 2, C-1 is much better located then the C-2, in means to avoid a short cycling of concentrate to the inlet of drainage pipe and thus providing better chances for COD removal and energy efficiency of methane production. This is supported by the fact that the leachate conductivity at the entrance to the RO plant (V131) reduced sharply from the value of 20.1 mS/cm to a value of 12.3 mS/cm after changing location of the concentrate disposal on 25$^{th}$ of November in 2015. This was the biggest recorded drop in conductivity (fig. 4), which at the same time reached the lowest value since the beginning of the concentrate re-circulation. It is unlikely that precipitation could affect the reduction in conductivity by dilution of leachate from the waste, given that a comparative analysis of rainfall and conductivity during the previous period did not reveal anything similar. Average methane concentrations in the first measurement period (from 4$^{th}$ to 8$^{th}$ of December in 2015) at wells B-14 and B-15, which are respectively closest to the C-2, amount to 63.1% and 59.7%, respectively. This may indicate the influence of concentrate to increase the concentration of methane in the well B-14, but this influence is difficult to prove, given that well B-14 is surrounded with older waste, which has already been discussed. Also, if increased methane concentration at the well B-14 is the result of concentrate impact, its decrease would be probably recorded during the measurement on February 24$^{th}$ in 2016, but it was not the case.

The most significant result of the LFG composition measurements, which could be firmly attributed to the influence of concentrate, is increased methane concentration at the well B-1. With an average value of 64.2% and 64.5% in the period of 2.5 months, it is 1.8% more than the average value of the respective nearby wells. It could be also noted that the re-circulation of the concentrate showed its influence on the increase of methane concentration already after ten days of its application.

Another, very important influence of concentrate disposal is that it supports waste to degrade by bringing additional moisture. Optimal moisture content allows for the transportation of nutrients, microorganisms, and intermediate products for enhanced biodegradation of waste to produce methane, as well as dilutes biodegradation inhibitors such as sulphates and heavy metals. Hence, moisture content has an effect on all other LFG indicators in a facilitator role. Dominant influence of moisture on methane production is also explained in the research where authors used multi-criteria analysis method for LFG prediction, where it was assigned the highest weight value [21].

If it is observed ratio of gas velocity, as a measure of gas flow, and the height of waste, as a measure of the amount of locally deposited waste, could be notified that the relative gas flow to the amount of waste in the well B-1 is among the highest of all wells or about 30% greater than the average (tab. 2). There are other parameters influencing lager amount of LFG at well B-1 then the average, such as a better waste coverage with earth, which contributes to the greater collecting efficiency of LFG because of less fugitive emissions. On the other hand, around the well B-1 is noticeable lush vegetation, which could affect higher moisture vapor from the surface and a smaller inflow of available moisture into the waste, which could lead to less moisture and consequently smaller amounts of gas.

Taking into account average COD value of the concentrate and disposed volume over the first measurement period, potential increase of the methane production from the concentrate, theoretically, if entire quantity would be released by the waste in a gaseous form, is about 8.7%. Real increase gives COD removal efficiency of 20%. This value is reasonable, bearing in mind that this is a real scale landfill. It is in line with research of leachates re-circulation in pilot plant [22], with COD removal efficiency 63-70% in total, while 40-48% was after first day of re-circulation (twice a day re-circulation regime). The COD value of landfill leachate was in range between 3500 g/m$^3$ and 5000 g/m$^3$, while the ratio $BOD_5$/COD was from 0.2 to 0.37, which is similar to the value of concentrate used in this research.

## Conclusions

This experimental research showed that the re-circulation of the concentrate obtained from the treatment of leachate by reverse osmosis, can generate additional quantities of LFG and methane from waste degraded under methanogenic anaerobic conditions. Numerical results obtained by laboratory analysis of concentrate organic matter and the LFG composition measurement at wells, show that 20% of total concentrate organic matter participate directly in methane production. Due to the relatively small amounts of discharged concentrate and low content of organic matter, its influence is negligible compared the total quantities produced at the landfill, with total share of only 0.07%. More important influence is assessed to be from moisture that is supplied to the landfill especially in climate areas with low precipitation as in this case. The current moisture impact is roughly assessed at about 0.9% of total LFG quantities.

According to pH values of leachate and concentrate over time, caused by managing of concentrate re-circulation, it could be noted that it may affect methane and LFG production temporarily, in terms of its reduction because of possible imbalances in population of methanogenic bacteria due to drop of pH value below optimal level.

In order to improve waste degradation with increase in methane production, further investigation should be directed towards direct leachate re-circulation. Increasing of leachate re-circulation should probably require improvement of its distribution within the waste, so the waste could be wetted to a greater extent which would create more favorable conditions for the methane generation by increasing the LFG flow rate, regardless of the lower COD content of leachate. Also, significant participation of concentrate organic matter in methane production, highlighted by this research, supports an idea for re-circulation of stronger leachate which could be obtained from the new landfill cells, which will be constructed in the future, to the existing cell. Proper management of leachate re-circulation could be valuable, not only in achieving greater, but also more uniform, methane quantities during the year, which would significantly improve the energy efficiency of methane production, as well as its beneficial use as a renewable energy source. In turn, landfilled waste and leachate would be much more stabilized, followed by less detrimental impact on the environment.

## References

[1] Reinhart, D. R, *et. al*., The Bioreactor Landfill – Its Status and Future, *Waste Management and Research, 20* (2002), 2, pp. 172-186
[2] Chan, G. Y. S., *et. al*., Effects of Leachate Recirculation on Biogas Production from Landfill Co-Disposal of Municipal Solid Waste, Sewage Sludge and Marine Sediment, *Environmental Pollution, 118* (2002), 3, pp. 393-399
[3] Francois, V., *et. al*., Leachate Recirculation Effects on Waste Degradation: Study on Columns, *Waste Management, 27* (2007), 9, pp. 1259-1272
[4] Wang, Q., *et. al*., Research on Leachate Recirculation from Different Types of Landfills, *Waste Management, 26* (2006), 8, pp. 815-824
[5] San, I., Onay, T., Impact of Various Leachate Recirculation Regimes on Municipal Solid Waste Degradation, *Journal of Hazardous Materials, 87* (2001), 1-3, pp. 259-271
[6] Delia Sponza, T., Agday, O. N., Impact of Leachate Recirculation and Recirculation Volume on Stabilization of Municipal Solid Wastes in Simulated Anaerobic Bioreactors, *Process Biochemistry, 39* (2004), 12, pp. 2157-2165
[7] Chian, E. S. K., DeWalle, F. B., Sanitary Landfill Leachates and Their Treatment, *J. Environ. Eng. Div., 102* (1976), *EE2*, pp. 411-431
[8] Eipper, H., Maurer, C., Purification of Landfill Leachate with Membrane Filtration Based on the Disc Tube DT, *Proceedings,* 7th International Waste Management and Landfill Symposium – Sardinia 99, CISA, Cagliari, Italy, 1999
[9] Heyer, K. U., Stegman, R., *Landfill Management: Leachate Generation, Collection, Treatment and Costs*, IFAS – Hamburg, Hamburg, Germany, 2002
[10] Talalaj, I. A., Mineral and Organic Compounds in Leachate from Landfill with Concentrate Recirculation, *Environ Science and Pollution Research*, *22* (2015), 4, pp. 2622-2633
[11] Kjeldsen, P., Beaven, R., Landfilling: Hydrology, in: *Solid Waste Technology & Management* (ed., T. Christensen), A John Willey and Sons, London, UK, 2011, pp. 709-733
[12] Fellner, J., *et. al*., Comparing Field Investigations with Laboratory Models to Predict Landfill Leachate Emissions, *Waste Management, 29* (2009), 6, pp. 1844-1851
[13] Barlaz, M., *et. al*., Bacterial Population Development and Chemical Characteristics of Decomposition in Simulated Sanitary Landfill, *Appl. Environ. Microbiol., 55* (1989), 1, pp. 55-65
[14] Vujić, G., *et. al*., Influence of Ambience Temperature and Operational-Constructive Parameters on LFG Generation – Case Study Novi Sad, *Thermal Science, 14* (2010), 2, pp. 555-564
[15] Gonzalez, C., *et. al*., Robles F., Effect of Solid Wastes Composition and Confinement Time on Methane Production in a Dump, *Journal of Environmental Protection*, *2* (2011), 10, pp. 1310-1316
[16] ***, VDI 4630 Fermentation of Organic Materials, Characterisation of the Substrate, Sampling, Collection of Material Data, Fermentation Tests
[17] Talalaj, I. A., Biedka, P., Impact of Concentrated Leachate Recirculation on Effectiveness of Leachate Treatment by Reverse Osmosis, *Ecological Engineering, 85* (2015), Dec., pp. 185-192
[18] Kukić, D., *et. al*., Potentials of Biogas Production from Wastewaters of Food Industry, *Proceedings*, 5th Regional Conference Industrial Energy and Environmental Protection in South Eastern European Countries IEEP '15, Zlatibor, Serbia, 2015, pp. 62
[19] Bilgili, M. S., *et. al*., Influence of Leachate Recirculation on Aerobic and Anaerobic Decomposition of Solid Waste, *J. Hazard Mater, 143* (2007), 1-2, pp. 177-183
[20] Farquhar, G. J., Rovers, F. A., Gas Production During Refuse Decomposition, *Water, Air, and Soil Pollution, 2* (1973), 4, pp. 483-495

[21] Emkes, H., *et. al.*, A Decision Support Tool for Landfill Methane Generation and Gas Collection, *Waste Management, 43* (2015), Sep., pp. 307-318
[22] Rodriquez, J., *et. al.,* Removal of Non-Biodegradable Organic Matter from Landfill Leachates by Adsorption, *Water Research, 38* (2004), 14-15, pp. 3297-3303

Paper submitted: April 1, 2016
Paper revised: May 12, 2016
Paper accepted: May 13, 2016