Original Research

Effect of Recirculation Method on Quality of Landfill Leachate and Effectiveness of Biogas Production

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Abstract

This study attempts to determine the effect of a dose of recirculated water and sewage added to wastes deposited on the volume and quality of leachate and biogas produced. Experiments were carried out with the use of wastes originating from the landfill in Łęgajny near Olsztyn. The research station consisted of four lysimeters; in two of them wastes were irrigated with water (2.15 and 4.30 mm/day) and the other two with sewage (2.15 and 4.30 mm/day). The research demonstrated a dependency between concentrations of pollutants occurring in the produced leachate and the volume and composition of biogas.

Keywords: leachate, recirculation, municipal solid waste

Introduction

Chemical and biochemical transformations in waste heap result in biogas and landfill leachate generation. In contrast to municipal wastewater, landfill leachate is characterized by high contents of organic and inorganic compounds, the content of a wide range of toxic substances and high variability. Concentrations of these compounds in leachate depend on the type of waste, its susceptibility to dissolution, chemical and biological degradation and the volume of water running through the waste. Landfill leachate contains mainly nitrogen and phosphorus compounds, hence it is characterized by a high chemical and biochemical oxygen demand. Concentrations of pollutants fluctuate significantly depending on the type of waste and its composition, landfill age and the method of landfill organization.

All these features are responsible for problems occurring in leachate treatment. Both in Poland and world-wide there is no rational method for landfill leachate treatment.

It is assumed that the amount of leachate ranges from 20 to 50% of annual precipitation. Lower values are reported for landfills operating with a waste press, and higher ones for landfills operating with a caterpillar bulldozer. Leachate passing through drainage and collective wells of an interior tank can be discharged to a municipal wastewater treatment plant, a local treatment plant or can be turned back on the waste heap. Leachate recirculation on the waste heap intensifies biodegradation of organic compounds under anaerobic conditions and improves a stable methanogenic phase.

Currently, landfills and processes inside the landfills insist on being treated as bioreactors with intensifying waste degradation [1, 2]. Processes that proceed inside the landfill heap are considered as in a long-term bioreactor and leachate composition is assayed [3].

The *in-situ* treatment of landfill leachate affects an increase in the production rate of biogas as a sustainable source of energy and accelerates decomposition of wastes, which may shorten the post-closure monitoring period of a landfill [4, 5].

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Onay et al. [6] carried out an experiment on a laboratory scale into the nitrification and denitrification processes as affected by leachate recirculation. A favourable impact of leachate recirculation on the waste heap and landfill operation was emphasized. The authors demonstrated that waste utilization through leachate recirculation improved biodegradation and stabilization due to a homogenous increase in water content and substrate and nitrate spreading. Consequently, leachate recirculation created in the waste environment special conditions that enabled development of denitrifying, nitrifying and methanogenic microorganisms.

In turn, Pirce et al. [7] presented the results of a study into nitrification outside the landfill in aerobic reactors and afterwards *in situ* denitrification on landfill sites.

Over the past 10 years experimental testing and field pilot studies have been conducted to develop and improve landfill techniques and designs, the goal being to control the negative effects of landfill sites on the environment [5]. Increasingly, landfills are considered to be bioreactors improving waste degradation. Hence, laboratory scale lysimeters are often used to simulate refuse degradation behaviors in landfills [8].

Experimental Procedures

The research station consisted of 4 lysimeters at the working volume of 44 l. Each lysimeter was fitted out with stub pipes. Samples of leachate and biogas were collected through the stub pipes. The volume of the biogas was measured. It enabled supplying water or leachate into the lysimeters. The scheme of the research station is depicted in Fig. 1.

Lysimeters were filled with a mixture of wastes collected from the municipal landfill located in Łęgajny near Olsztyn. Morphological and quantitative analyses of wastes were carried out before filling the lysimeters (Table 1, Table 2).

The experiment spanned for 11 months. Over the entire experimental period, the wastes in the lysimeters were supplied with water or leachate originating from the landfill in Łęgajny. The following variants were tested:

lysimeter 1 – water – 2.15 mm/d*
lysimeter 2 – water – 4.30 mm/d
lysimeter 3 – leachate – 2.15 mm/d
lysimeter 4 – leachate – 4.30 mm/d

* annual precipitation in Poland is 780 mm/year

Characteristics of the leachate used in the experiment are shown in Table 3.

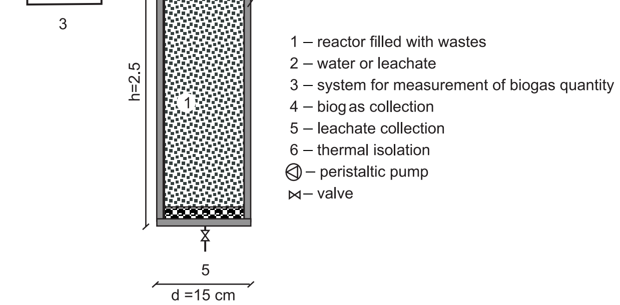

Fig. 1. The scheme of the research station.

Table 1. Sieve analysis of wastes.

Fraction [%]			
> 120 mm	120 – 40 mm	40 – 8 mm	8 – 0 mm
3.8	28.6	52.5	15.1

Table 2. Average refuse composition used in original forms (wet basis).

Composition	Weight percentages
Food wastes	49.4
Paper	15
Plastics	3.9
Cloth	3.5
Glass	6.7
Metals	1.7
Rest mineral fraction	3.3
Fine fraction	16.5
Total	100
Water (moisture)	38.1

Table 3. Physical and chemical properties of the leachate used in the investigations.

	Mean	Range
Reaction (pH)	8.87	8.19 – 9.08
Conductivity (mS/cm)	8.2	7.4 – 9.9
COD (mg/dm^3)	3915	2665 – 4352
Total P (mg/dm^3)	20.8	16.6 – 31.8
Total N (mg/dm^3)	442	385 – 490
Sulfate (mg/dm^3)	274	238 – 327
Dry residue (mg/dm^3)	8618	7344 – 10628
Residual after combustion (%)	79	73 – 85
Loss at combustion (%)	21	15 – 27

The experiment was carried out at room temperature (about 20°C). Biogas generation was measured every day in calibrated vessels filled with saline. Analyses of biogas quality were done four times over the experimental period. Biogas was assayed for the following gases: methane, carbon dioxide, nitrogen, oxygen and hydrogen.

Analytical Methods

Leachates were assayed once a month and the following analysis were carried out:
- pH (pH-meter *HI 8818*);
- concentration of organic compounds expressed as COD (dichromate method, PN-74/C-04578/03);
- total nitrogen (Kjeldahl method, *Buchi*);
- ammonium nitrogen (distillation method, *Buchi*);
- concentration of nitrate with the colorimetric method [PN –73/C-04576/087]
- total phosphorus [Photometer SQ-118 Merc]

The analyses of biogas composition were carried out at the Institute of Mining and Gas Oil in Warsaw. Biogas was assayed for the following gases: methane, carbon dioxide, nitrogen, oxygen and hydrogen.

Results and Discussion

The chemical composition of landfill leachate depends on a variety of parameters and processes taking place inside a waste heap. Locations of landfill, waste type, landfill construction and the method of its management affect the qualitative and quantitative composition of leachate.

An organic fraction of waste exerts an influence on waste degradation on the landfill site, and on leachate composition. Substances toxic to microorganisms may especially inhibit biodegradation, and indirectly the amount of leachate and its composition. The content of inorganic compounds in leachate depends on the contact of waste with water, reaction pH and chemical balance on the surface of distribution between the solid state and liquid. Most metals are leaching from waste mass at acidic pH. Reaction is the basis of mass transfer in the waste-leachate system in precipitation, dissolution, and sorption.

Changes in leachate composition and pollutants leaching from waste are interrelated with landfill age. Landfill age determines the characteristics of leachate and the type of stabilization process. It can be described by the regulation of changes in waste composition and does not depend only on waste age but also on the level of waste stabilization and the volume of water that infiltrates inside the waste heap. Maximal pollutant loading in leachate is observed in the initial years of landfill operation (2-3 years), and gradually decreases in subsequent years. This trend is appropriate in relation to organic compounds, especially to COD, BOD, TOC, mainly to microbiological pollutants and, first of all, to inorganic ions (heavy metals, Cl, SO_4, etc.).

Water content in wastes collected on landfills is one of the elements influencing waste degradation rate. The vol-

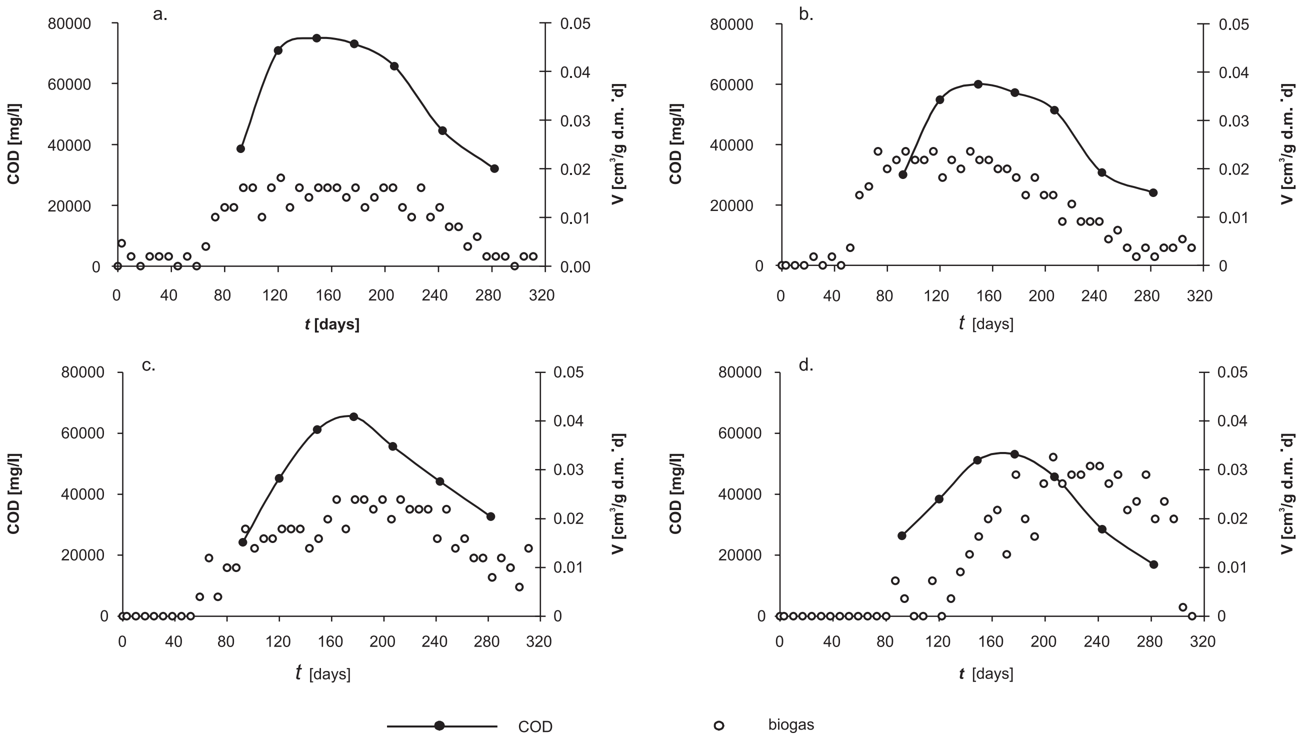

Fig. 2. Relationship between COD concentration in leachate and biogas production. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter 4.

ume of leachate and its composition also result from the moisture content of wastes. Bibliographic data indicate that normally moisture content of waste ranges from 20% to 40% [9]. Water content of waste used in the experiment was 31.8%, thus within the common range.

Solid waste hydrolysis, biological degradation and dissolution of the soluble salts originating from waste are the mechanisms responsible for the regulation of mass transfer from waste to leaching water, i.e. leachate.

Figs. 2-6 show variations of pollutant concentration in leachate. Our results demonstrated that the highest COD concentration in landfill leachate – 75,000 mg/l (Fig. 2) was in lysimeter 1. It was a lysimeter supplied with water at a dose of 215 mm/d, which corresponds to average annual precipitation. Concurrently, biogas production in this lysimeter was the lowest compared to other lysimeters tested. Methane was not detected (Fig. 7). Analogous results were described by He et al. [10], who also obtained leachate with the highest COD values (70,000 mg/l) and the lowest gas production in the case of a lysimeter supplied with water.

In the reported experiment, the lowest COD concentration – 50,000 mg/l, was observed in lysimeter 4. In all lysimeters, the same tendency was observed for COD changes, i.e. an increase of up to ca. 180/day and then a decline in the concentration of pollutants expressed in COD values. This is consistent with findings of other authors, who reported first the highest increase in COD concentrations in leachates after ca. 100 days of the experiment (125 [11], 150 [12], 160 [13]) and then their decline.

The highest biogas production was observed in lysimeter 4 supplied with leachate – 4.30 mm/d, which corresponds to double annual precipitation. Biogas contained up to 50% of methane. Lower concentrations of pollutants in lysimeter 4 resulted from the stable methanogenic stage.

Recirculation of leachates may exert a variety of effects on changes proceeding in wastes. Leachates added may, to a considerable extent, enhance refuse stabilization and yield early methanogenesis. Their occurrence might be attributable to the excess of methanogenic bacteria and/or the higher pH of leachate (Table 3) recycled back with the leachate. The leachate recycled to column with a mature landfill layer would carry with it excess methanogenic bacteria, but may need time to propagate downward with the leachate flow to stimulate methanogenesis [14, 15, 10]. Also in our experiment, a tangible delay in the appearance of biogases was observed in lysimeters 3 and 4, as compared to lysimeters 1 and 2 supplied with water.

The results of changes of nitrogen compounds (Fig. 3-5) in leachate from the four lysimeters tested indicated that in the lysimeters supplied with water (Fig. 3a, b) the highest total nitrogen concentration was in time of the highest biogas production. A decrease in the volume of biogas generated was accompanied by a decrease in the total nitrogen concentration in leachate. In the lysimeters supplied with leachate, the maximum total nitrogen concentration in leachate (Fig. 3c, d) was achieved just before maximum biogas production. High biogas generation in

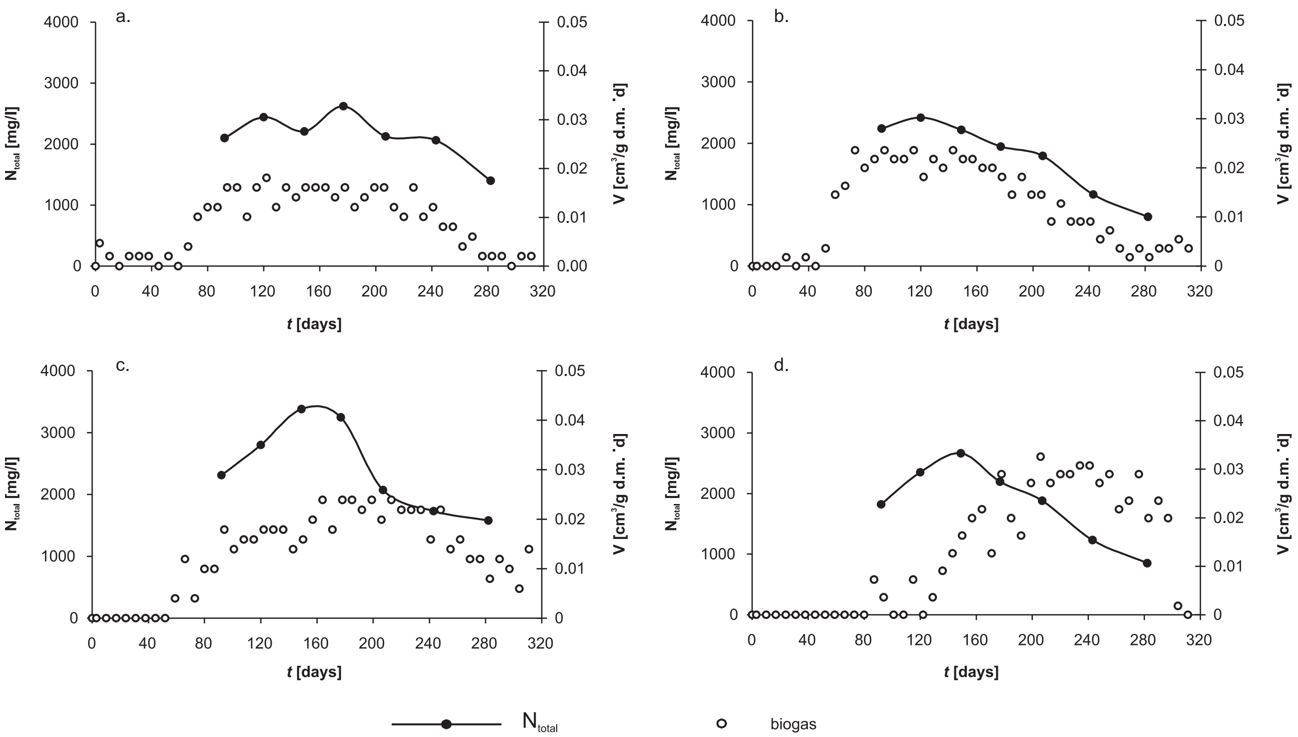

Fig. 3. Relationship between N-total concentration in leachate and biogas production. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter 4

these lysimeters was related to the decrease in total nitrogen content of leachate.

An opposite tendency was observed in the case of ammonium (Fig. 4). In lysimeters supplied with water or leachate in the amount of 2.13 mm/d (Fig. 4a, c), ammonium concentration in leachate over the entire experimental period ranged from 1400 to 1500 mg/l. A twofold increase in the amount of water or leachate supplied (4.30 mm/d) resulted in a decrease in ammonium concentration in both lysimeters after 120 days of the experiment (Fig. 4b, d). The high concentration of ammonium nitrogen in leachate, i.e. 1200 – 1500mg/l, results from decomposition of organic matter containing nitrogen, such as protein and amino acids. As a result of degradation of these nitrogenous organics the ammonia-nitrogen concentration is so high. Decreasing ammonium concentration results from the fact that ammonia was consumed by the anaerobic bacteria to develop their cellular components [16].

Fig. 5 depicted changes in nitrate concentration in leachate. In three lysimeters tested, the increase in nitrate concentration in leachate was accompanied by a decrease in biogas production. Nitrate concentration in lysimeter 4 was found not to affect the increase in biogas production.

This is consistent with results described by Pirce et al. [7]. The authors proved the impact of nitrate on biogas production. The rate of methane generation decreased suddenly in all reactors concurrently with the presence of nitrate. Further studies showed that when nitrates were not supplied into the reactors the rate of methane production achieved the level from before nitrate addition. The authors suggested that methanogenic microorganisms were limited by the high nitrate concentrations. This fact is especially important in the case of landfill management. If we want to use methane generated at the landfill site we cannot design the landfill as a bioreactor with simultaneous methane production and nitrogen compound transformation.

An opposite tendency was observed for total phosphorus (Fig. 6.). The highest concentrations of P_{og} were achieved at the beginning and at the end of the experimental period. In time of the highest biogas production phosphorus concentration was observed to decrease.

Fig. 7 shows the composition of biogas generated in lysimeters The results obtained indicate that biogas composition depended on both medium supplied into the lysimeter (water or leachate) and on its dose. In lysimeters 3 and 4, supplied with the leachate, a higher percentage content of methane was observed in biogas, in contrast to lysimeters 1 and 2, supplied with the same amount of water. Only in the lysimeters supplied with water, a high hydrogen content was observed in the initial days of the experiment. Variations in CO_2 and N_2 in all lysimeters tested were analogous. However, in lysimeter 1 supplied with the higher dose of leachate, a decrease below 9% was observed for N_2 just after 210 days, whereas while in the other lysimeters the percentage content of nitrogen in biogas ranged from 40 to 30%. The positive effect on biogas generation with a high content of methane was revealed, taking into consideration the amount of medium supplied

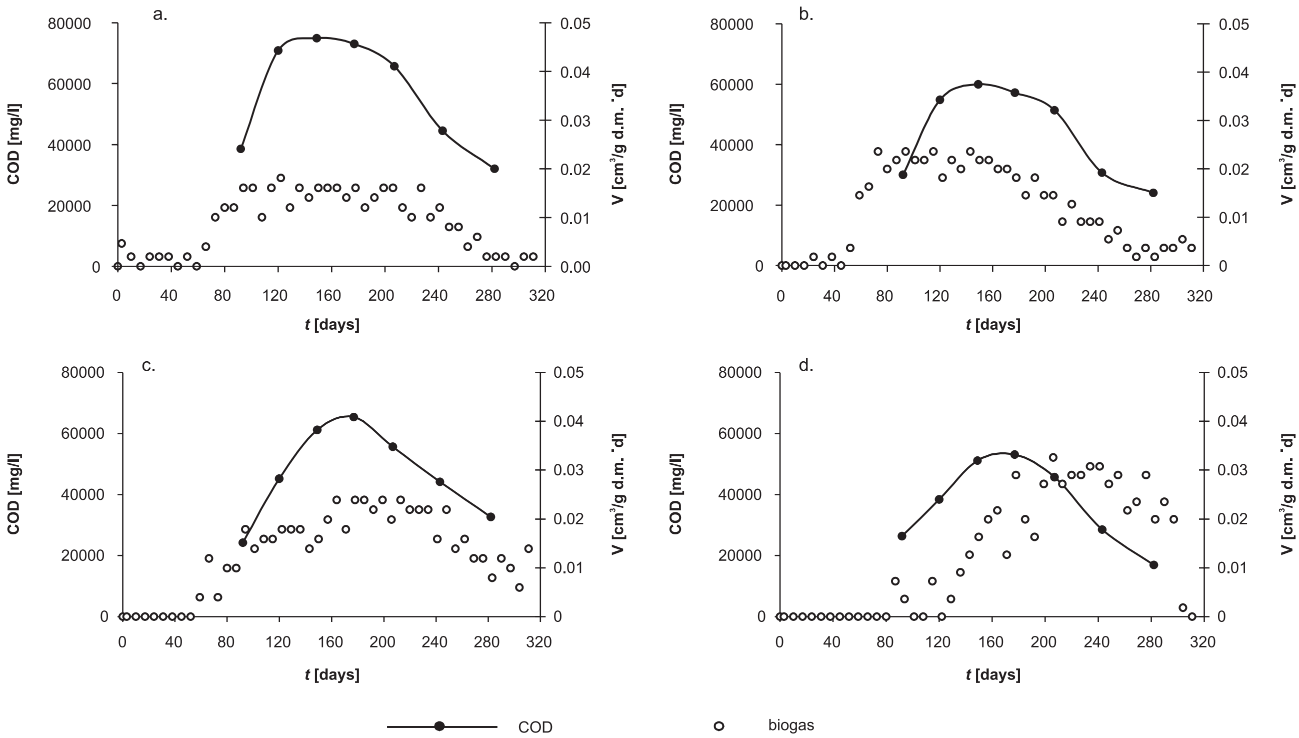

Fig. 4. Relationship between NH_4-N concentration in leachate and biogas production. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter 4.

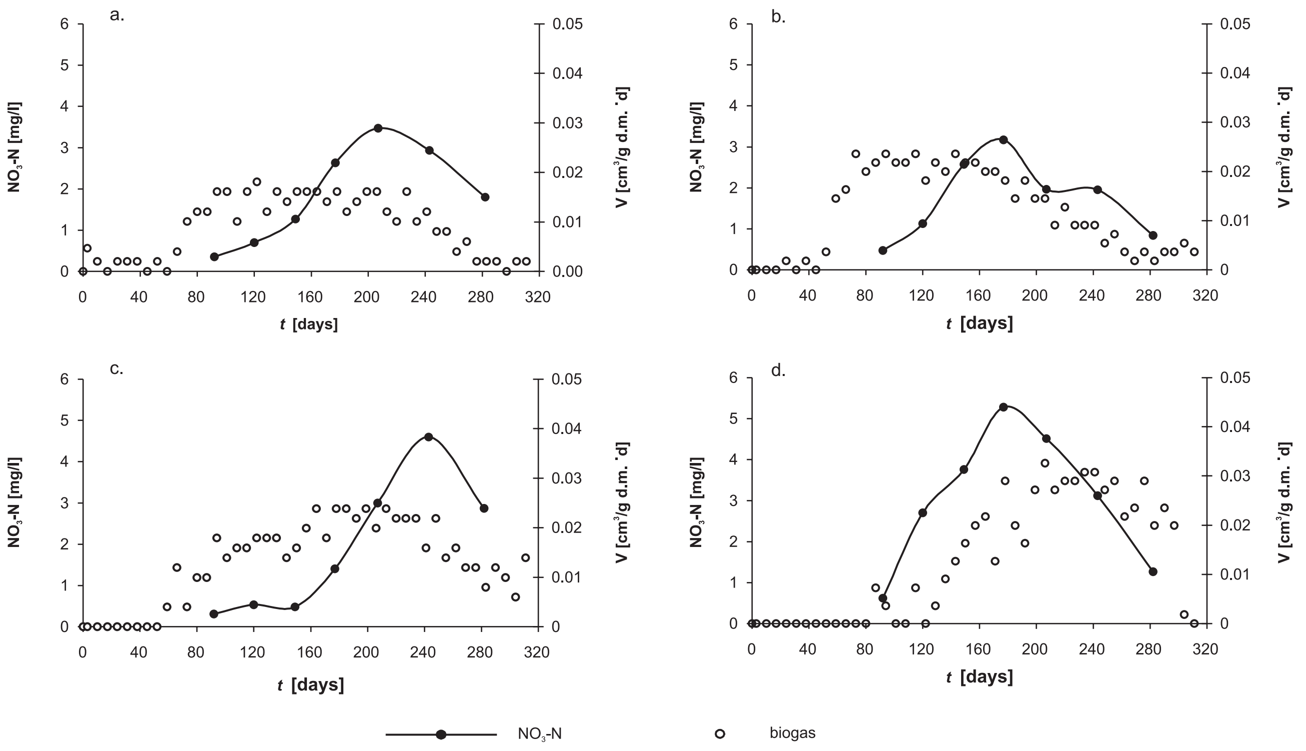

Fig. 5. Relationship between NO_3-N concentration in leachate and biogas production. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter.

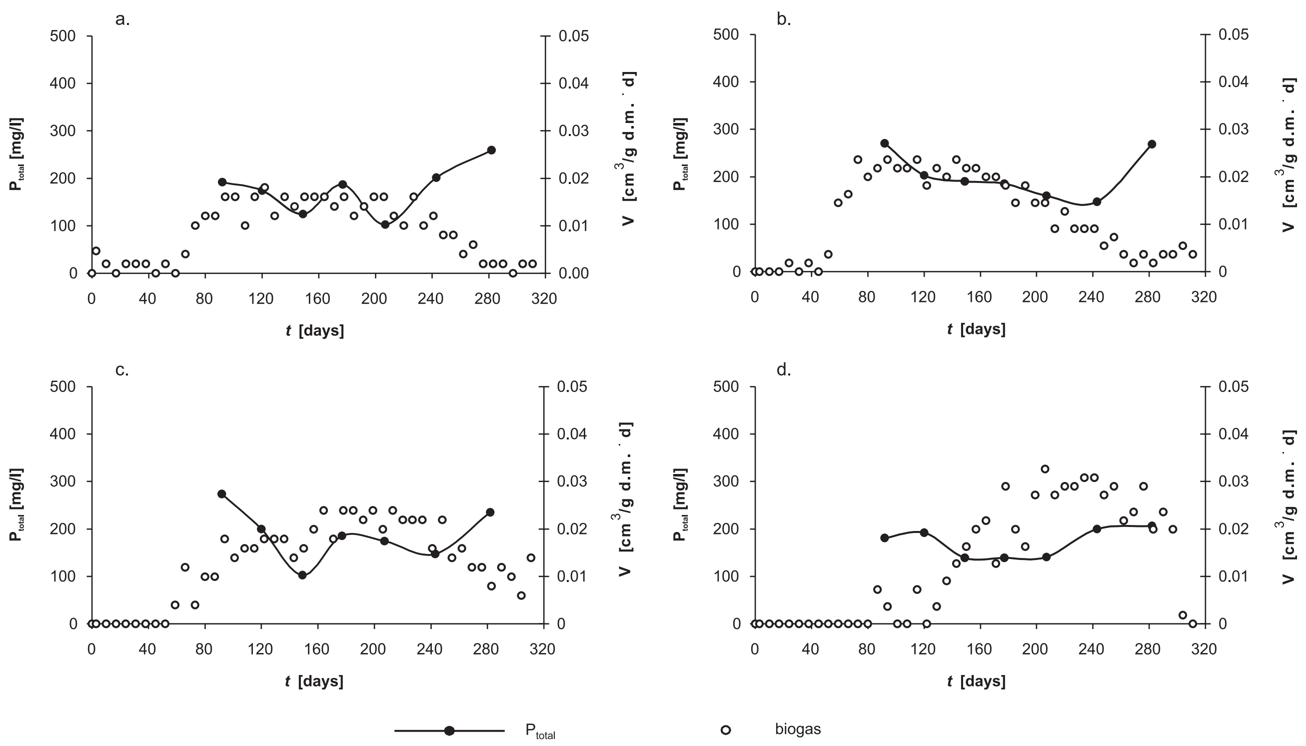

Fig. 6. Relationship between P_{total} concentration in leachate and biogas production. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter

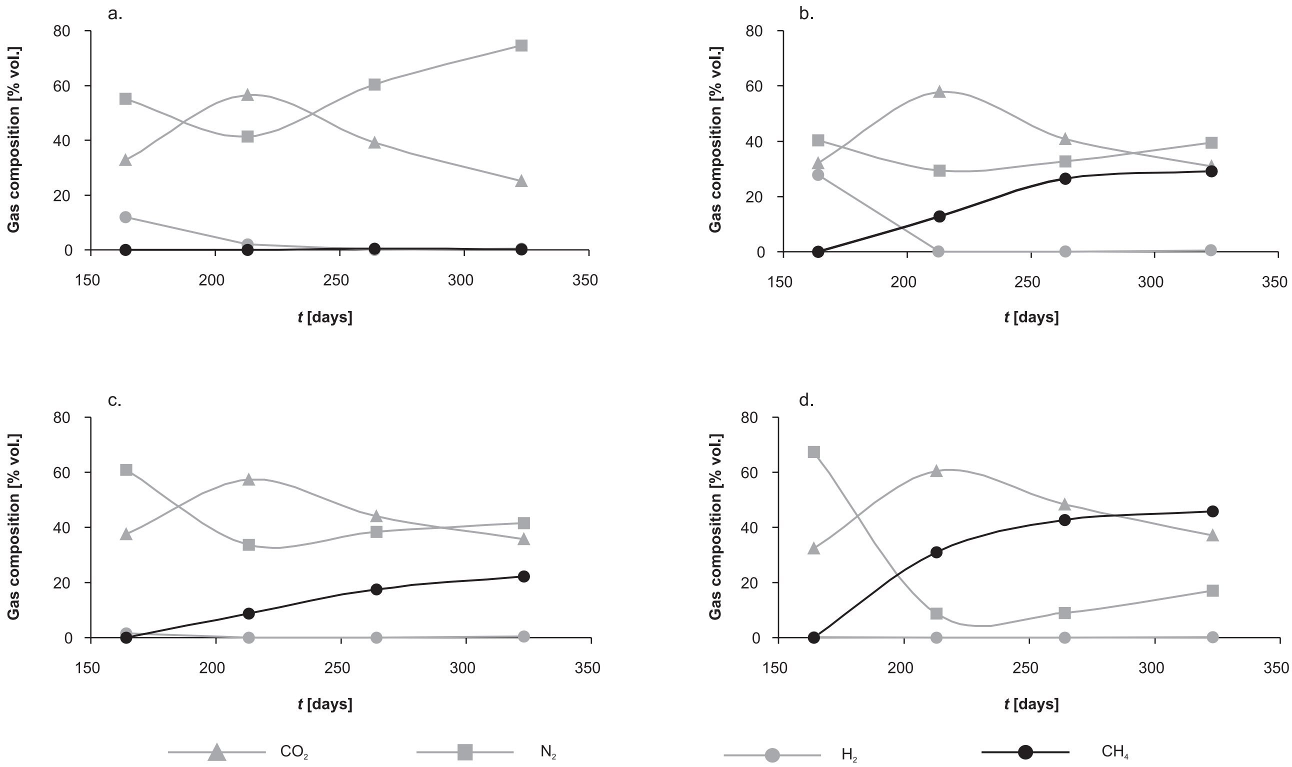

Fig. 7. Gas composition. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter 4.

into the lysimeters. A comparable content of methane in biogas, about 25%, was obtained in lysimeter 2 supplied with water – 4.30 mm/d, and in lysimeter 3 supplied with two times lower amount of leachate – 2.15 mm/d. In lysimeter 4 supplied with 4.30 mm of leachate per day, the content of methane in biogas was found to account for 46%; however, in lysimeter 1 supplied with water – 2.l5 mm/d, methane content was under detection, whereas nitrogen and carbon dioxide were present.

In this study, in the four lysimeters tested, the pH value was observed to decrease to pH 5.4 around day 210 of the experiment, and next to increase to the value of 6.8 – 7.09 (Fig. 8). The results obtained are consistent with findings of other authors. In studies of Sponza and Agdag [11] and He et al.[10] pH values ranged from 5.35 to 6.0, whereas San and Onay [17] obtained pH of 5.5. Such a pH value, however, does not facilitate the process of methanogenesis. Recent studies have shown that methanogenesis is enhanced at pH ranging from 6.4 to 7.2 [18]. An increase in pH value between day 240 and 310 of the experiment had a substantial effect on methane concentration in the biogas produced (except for lysimeter 1, where no methane was detected in the biogas). The maximum production of biogas, at simultaneously the lowest value of pH, is likely to indicate that it is not the pH that inhibits and reduces biogas production. This has also been confirmed by works of other authors addressing a comparison of the process of biogas production from wastes and sludges [11, 19].

A decrease in pH at high COD values under anaerobic conditions may also enhance the production of ammonium- nitrogen (NH_4-N) from degradation of nitrogenous organic compounds. In this study, the highest COD values, ranging from 75,900 to 53,100 mg/l, were reported on ca. day 180 of the experiment, and this was also the time when the highest concentrations of NH_4-N (from 1,200 to 1,600 mg/l) were recorded in leachates.

High volatile fatty acids formation and high temporary pressure of CO_2 may result in reaction pH decrease. In the presented experiment,, an increase in CO_2 content in biogas generated in all lysimeters was observed after 210 days. The content of CO_2 constituted about 60% of biogas in all lysimeters (Fig. 7).

Our experiment showed that leachate taken from the landfill was characterized by high concentrations of COD and ammonium. COD concentration, depending on medium supplied into the lysimeters and the dosage, ranged from 53,000 to 75,000 mg/l, and the highest value was observed in lysimeter 1, supplied with water at a dose of 2.15 mm/d. However, the lowest value of COD content was reported in lysimeter 4 supplied with a higher dose of leachate. COD concentrations appeared to be higher in lysimeters with lower dosage of water or leachate – 2.15 mm/d. The two times higher dose of medium caused a decrease in COD content. An analogous relationship was observed in the case of total nitrogen, the concentration of which ranged from 2,400 to 3,400 mg/l, depending on the dosage of medium supplied into the lysimeters. A higher total nitrogen concentration was obtained in lysimeters supplied with the lower doses of medium, both of water and leachate (Fig. 3).

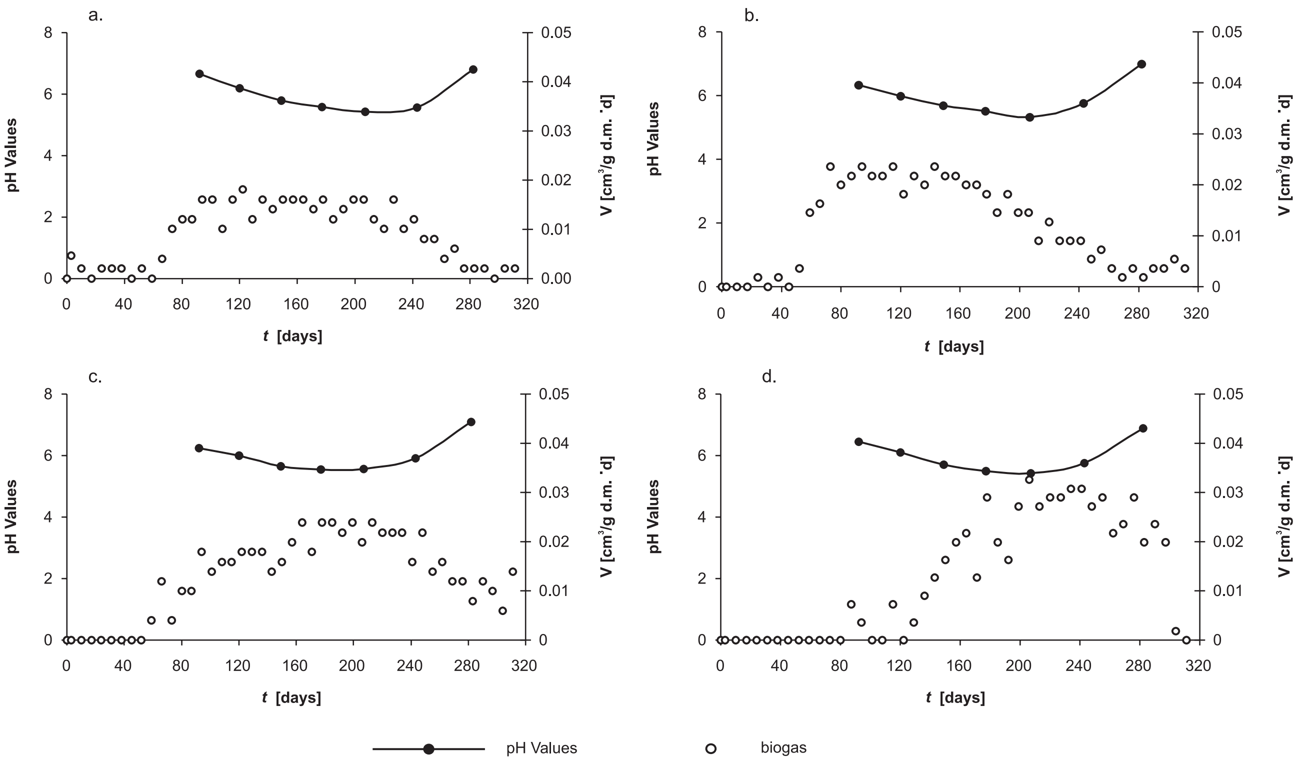

Fig. 8. Changes of leachate pH values over time. a – lysimeter 1, b – lysimeter 2, c – lysimeter 3, and d – lysimeter 4.

Ammonium concentration after 160 days of the experiment was high in all lysimeters and reached 1500 mg/l.

Similar results were achieved by Robinson (1989) [20], leachate from the landfill being in this phase was characterized by a high concentration of BOD_5 (above 10,000 mg/l), a high ratio of BOD/COD (>0.7), acid reaction (pH 5-6), and high ammonium content (often 500 – 1000 mg/l).

The results obtained indicate that the concentrations of pollutants present in leachate were not constant, and were closely related to the system of waste collection and period of waste landfilling. Transformations of nitrogen compounds in leachate are of high relevance. There is a necessity for precisely monitoring not only the amount of generated leachate but also the type and concentration of the pollutants. Accurate characteristics of landfill leachate determine the method of waste disposal.

Conclusions

The effect of leachate recycling on leachate quality and biogas production was experimentally explored in four landfill lysimeters. The addition of water and sewage to leachate originating from the landfill in Łęgajny near Olsztyn appeared to affect both the quality of leachate obtained and the composition of biogas produced.

The study demonstrated that over the entire experimental period, the lowest pH values, i.e. 5.33 – 5.55, were noted on ca. day 180 of the experiment. Since ca. day 200, an increase was observed in pH values up to ca. 7.0 in all lysimeters.

For all the variants examined, the highest COD values were found on ca. 180 day of the study, that were followed by a tangible decrease. In lysimeters to which sewages were recirculated, the concentration of pollutants expressed in COD was lower by ca. 15% as compared to those supplied with water. The lowest COD values were reported in lysimeter 4, i.e. a decrease to 16,000 mg/l.

The study confirmed the inhibiting effect of the presence of nitrate nitrogen NO_3-N on the efficiency of gas production. A decrease in the concentration of NO_3-N was also found to affect an increase in methane concentration in the biogas produced.

The highest efficiency of biogas production was noted in the lysimeter supplied with leachate at a dose of 4.30 mm/d. Simultaneously, biogas obtained from that lysimeter was characterized by the highest concentration of methane – 50%.

Despite the biochemical basis of the process taking place inside the waste heap of landfill, there is no possibility of precise correlation of leachate quality with the sort of waste collected at the landfill site with methods of waste management and landfill age. Leachate quality is strictly dependent on physical, chemical and biochemical processes proceeding inside the landfill. Especially leachate from the landfill being in the stable methanogenic phase involves lower concentration of pollutants.

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