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Aerobic and anaerobic biodegradation potential of leachate from old active landfill

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ABSTRACT

A common problem of biological treatment of landfill leachates is high toxicity and low biodegradability. Leachates from old landfills do not exhibit high toxicity and if the landfill is still filled by waste, they may contain sufficient amounts of biodegradable organic matter. With the aim of evaluating biotreatability of leachates from old, but active landfill, aerobic and anaerobic degradation assessment procedures were carried out. Investigated leachates showed low toxicity to aerobic and anaerobic micro-organisms. During biodegradability tests, anaerobic biological treatment did not lead to measurable production of biogas. Leachates were well degraded under aerobic condition in ready (66%) and inherent (78%) biodegradability tests, while simulation test in pilot sequencing batch reactor showed that leachates can be successfully treated, mixed with sewage up to 10% v/v.

Keywords: Biodegradability; Waste; Landfill leachate; Sequencing batch reactor; Toxicity

1. Introduction

Landfill leachates from municipal solid waste landfill sites are considered as hazardous and heavily polluted wastewaters [1]. Leachates arise when rainwater percolates through disposed waste in the landfill body. Water interacts with waste and it becomes contaminated by various waste components containing a variety of organic and inorganic pollutants in dissolved or suspended forms. Landfill leachate has a varying composition and strength of organic and inorganic compounds at each phase of municipal solid waste decomposition. In each phase of landfill lifecycle, leachate has unique characteristics that are significantly different from one phase to another [2].

Typically, leachates are classified in terms of landfill age as young, intermediate, and stabilized [3]. Young leachates are generated during early phase of landfill lifecycle (up to five years after waste placement) and they are characterized by high concentrations of biodegradable organic matters, and as a result have a high biochemical oxygen demand (BOD) and chemical oxygen demand (COD) ratio (BOD/COD > 0.5). The simplest and cost-effective biological treatment methods used for young leachates are conventional activated sludge, sequencing batch reactors (SBR) [4], biofilters [5], anaerobic up flow sludge blanket [6], anaerobic SBR [7], or a combination of anaerobic and aerobic systems [8]. With time, some of the biodegradable organics are decomposed in the stabilization process within the body of landfill and BOD/COD ratio decreases. During this period, intermediate leachate is

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generated with BOD/COD ranging between 0.1 and 0.5. Decrease of biodegradable organic fraction in leachate reduces biological treatment efficiency. However, low content of compounds inhibiting aerobic and anaerobic micro-organisms (e.g. ammonium) allows application of biological processes ideally combined with physicochemical treatment to assure high treatment efficiency [9,10]. Further, maturing of leachate poses problems to biological treatment due to decrease of biodegradable matter (BOD/COD < 0.1), increase of refractory components (e.g. humic and fulvic acids), and inhibitory compounds (e.g. ammonium) [11]. Therefore, biological treatment of stabilized leachates is recognized as not efficient and has to be replaced by physicochemical methods [12]. However, leachates from old landfill sites that are still in use and continuously filled by freshwaste may still contain high content of biodegradable organics [13]. Hence, the aim of our study was to investigate if leachate from old, but still active landfill can be treated by biological processes. At first, inhibiting impact of leachate to aerobic and anaerobic micro-organisms was investigated. Biodegradability testing was performed according to a strategy where a simple low microbial density method is applied at first, and if leachate expresses a good biodegradability, test with a higher cell density is applied. Successful biodegradability of leachate in a higher cell density test leads to performance of a simulation test.

2. Materials and methods

2.1. Characterizations of landfill leachates

The landfill site where leachates were sampled was chosen to obtain leachates from old, but active landfill. The site is located near the city of Logatec, Slovenia. The landfill was opened in 1977 and reconstructed in 2004. It has a total filling area of 1.5 ha, of which 1.45 ha had been filled by 2009. The maximum annual amount of leachate is 8,000 m³ and the maximum daily volume is 56 m³. Leachates are collected in an open retention basin of 750 m³ capacity and released into a nearby stream [14]. The composition of leachates was monitored during the years 2010 and 2011. Leachate was sampled four times at the effluent from retention basin. For evaluation of landfill leachate quality, several physicochemical parameters: pH, BOD₅, COD, DOC (dissolved organic carbon), TOC (total organic carbon), concentration of ammonium nitrogen, concentration of nitrite and nitrate nitrogen, orthophosphates, and chlorides were determined [13]. Each analysis was performed twice (two replicates) and standard deviations were calculated.

2.2. Biodegradability tests

2.2.1. Aerobic biodegradation

First of all, inhibition of oxygen consumption by nitrifying and heterotrophic micro-organisms in activated sludge was measured. As a result, an effective concentration (EC₅₀) was calculated. It is the concentration of added landfill leachate that after 30 and 180 min of incubation under given condition brings a specified effect in 50% of the test micro-organisms. EC₅₀ was calculated according to standard procedure [15]. Definitive test was repeated twice and standard deviations were calculated.

The first applied biodegradability test was used for determination of ready biodegradability and it provides information about biodegradability of leachate under the most common environmental conditions. The concentrations of leachates in the system were chosen to avoid any toxic impact to the activated sludge (inoculum in biodegradability tests) and to assure initial COD of 100 mg L⁻¹, and DOC of 40 mg L⁻¹. The inoculated medium was stirred in a closed flask and biodegradability was evaluated by DOC analysis [16]. Biodegradation of leachates was monitored for 28 d.

In the second test, inherent biodegradability was determined to evaluate biodegradability of leachate under conditions which appear in a biological treatment plant. Only leachates which were well degraded in the ready biodegradability tests were used for evaluation of inherent biodegradability in the Zahn-Wellens test [17]. Biodegradability of the leachate was determined in a presence of high-density inoculum $(1,500 \text{ mg}_{\text{MLVSS}}\text{L}^{-1})$ and concentration of landfill leachate was 10% v/v in 5 L reactors with continuous stirring and aeration. Reduction of DOC and COD, concentration of oxygen, and pH was monitored daily for a period of 25 d in the test mixture, blank, as well as in the abiotic test (without inoculum added).

The last test was chosen to evaluate biotreatability of leachates in a lab-scale SBR [18]. The SBR is operated mainly under aerobic conditions when organic compounds (COD) are reduced and ammonium is oxidized into nitrate *via* nitrification. During each SBR cycle, one longer settling phase was included to achieve anoxic conditions for possible conversion of nitrate into gas nitrogen *via* denitrification [19]. For the laboratory experiments, two glass cylindrical reactors of total volume 2 L were used. One of them was fed with synthetic sewage (blank test), while in the second one the landfill leachate was introduced. Synthetic sewage was prepared according to standard procedure [18]. Peristaltic pump was employed for influent feed and effluent withdrawal, while the excess of sludge was removed manually to maintain its constant concentration $(3.2 \pm 0.3 \text{ g L}^{-1})$ during the performance of the system. Nineteen cycles (days) were run. During this time, each cycle (24 h) contained the following phases: filling (0.25 h), aerating (5.0 h), settling (5.0 h), aerating (13.0 h), settling (0.5 h), and removal of treated leachate (0.25 h). In the first four cycles, reactor was acclimated with synthetic sewage, in the next seven cycles, 10% v/v of the landfill leachate was added in the synthetic sewage, while in the next four cycles its concentration was increased to 20% v/v. In the last four cycles, 40% v/v of landfill leachate was added. The second reactor (blank) was operated under the same conditions, but it was fed only with synthetic sewage [20].

To evaluate possible loss of ammonium nitrogen by air stripping during aeration phase of the SBR cycle, additional batch experiment was setup: 200 mL of the leachate was diluted by synthetic sewage (20% v/v of the leachate) and the mixture was aerated for 24 h.

2.2.2. Anaerobic biodegradation

For evaluation of biodegradability of leachates under anaerobic conditions, a method for determination of ultimate biodegradability of complex organic substrates and biogas production [21] was used with some minor modifications. The method is based on exposure of the tested sample to anaerobic microorganisms from digested sludge at constant temperature 39°C (\pm 1°C), in the dark and in a sealed stirred system (OxiTop system, WTW Germany, 2008) for up to 10 d.

First of all, the toxicity of the leachates to microorganisms of anaerobic sludge was determined by measurement of increased pressure in the vessels resulting from the production of CH₄ and CO₂ (biogas) in the system with different concentrations of the landfill leachate, inoculum (anaerobic sludge, 6.6 g L^{-1}), and glucose (60 mg L^{-1}). Any toxic effect caused by the leachate was indicated by a lowering of the biogas production. Simultaneously, the blank test and the reference test (only glucose) were accomplished. At the end of the experiments, 2.5 mL of 6 M NaOH solution was injected in the rubber sleeve of the reaction vessel and the CO₂ in the gaseous phase was absorbed. The residual pressure in the system pertained to produced methane. The inhibition of biogas production was calculated according to the standard procedure [21].

Biodegradability experiments were prepared in the same way as the experiments for determination of toxicity, but the test mixture (landfill leachate and inoculum) did not contain glucose, and thus the production of biogas was caused only by the degradation of the landfill leachate.

3. Results and discussion

3.1. Characterization of the landfill leachates

Physicochemical parameters of the leachates from the active municipal landfill are presented in Table 1. Waste has been disposed in the site for more than 30 years and thus generated leachates had many typical attributes of mature leachates as alkaline pH, high content of ammonium nitrogen, and chlorides. However, BOD/COD ratio has ranged between 0.17 and 0.33, with an average of 0.21 indicating higher content of biodegradable organics typical for intermediate leachate. Also TOC/COD ratio being between 0.33 and 0.82 might classify the leachate among intermediate (0.3–0.5) or stabilized (>0.5) leachates [3]. The presence of biodegradable organics in the mature leachate is probably caused by a continuous disposal of municipal waste in the site. Some parameters (e.g. BOD₅, Cl⁻) showed variability that can be caused by seasonal weather changes, composition, and amount of currently disposed waste on the landfill [22].

3.2. Aerobic biodegradation

Landfill leachates can contain many hazardous compounds, which can act as inhibitors to degrading micro-organisms. Therefore, toxicity of leachates has to be assessed to select concentrations of leachates used in biodegradability tests which are harmless for heterotrophic micro-organisms of activated sludge. Furthermore, leachates must not inhibit nitrification, which is considered to be the key process in biological

Table 1

Physicochemical parameters of landfill leachates generated from investigated landfill (n = 4, mean \pm SD)

Parameter	
pH (/)	7.8 ± 0.6
$TOC (mg L^{-1})$	$1,165 \pm 195$
$COD (mg L^{-1})$	$2,274 \pm 691$
$BOD_5 (mg L^{-1})$	497 ± 348
BOD/COD (/)	0.21 ± 0.08
TOC/COD (/)	0.58 ± 0.25
$N-NH_4^+$ (mg L ⁻¹)	796 ± 297
$N-NO_{3}^{-}$ (mg L ⁻¹)	1.8 ± 1.4
$N-NO_{2}^{-}$ (mg L ⁻¹)	3.4 ± 2.9
$P-PO_4^{3-}P-PO_4^{3-} (mg L^{-1})$	5.3 ± 5.1
$\operatorname{Cl}^{-}(\operatorname{mg} \operatorname{L}^{-1})$	$1,150 \pm 453$

nitrogen removal [23]. Landfill leachates had no impact on heterotrophic micro-organisms in activated sludge (30 and 180 min $EC_{50} > 100\% v/v$). However, all leachates showed acute impact on nitrifying microorganisms (30 min $EC_{50} = 40 \pm 14\% \text{ v/v}$) that slightly increased with time of incubation (180 min $EC_{50} = 25$ $\pm 5\%$ v/v). Nitrifying micro-organisms are slowly growing micro-organisms and they are usually more sensitive than heterotrophic ones. Their increased sensitivity to mature leachates is given by their low tolerance to salinity and ammonium nitrogen [24,25] that were found in all leachate samples in high concentrations. Nevertheless, due to the low toxicity of the leachates to the heterotrophic micro-organisms, which are responsible for organic matter removal, biodegradation should be thus limited only by the presence of refractory compounds in landfill leachates.

Biodegradation of leachates under common natural conditions (ready biodegradability test) was compared to the reference compound, sodium acetate. Sodium acetate was always degraded more than 60% in the first five days of experiment and it reached $93 \pm 6\%$ degradation in 28 d confirming high activity of microorganisms and good performance of the test. In this test, biodegradability of leachates was up to $66 \pm 7\%$ and in the Zahn–Wellens test (inherent biodegradability test) the reduction of COD and DOC was enhanced to $78 \pm 2\%$ and $83 \pm 2\%$, respectively. Our results confirmed good biodegradability of leachates in two types of biodegradability tests (low- and high-cell density tests) and leachate was submitted to SBR for biological treatment.

For optimal biological growth in the wastewater treatment system, the ratio among carbon, nitrogen, and phosphorus (C:N:P) should be between 100:20:1 and 100:5:1 [26]. The C:N:P ratio of sewage is often ideal; however, landfill leachates usually contain excess of nitrogen and deficiency of phosphorous (Table 1). Another problem connected to the biological treatment of landfill leachate is bulking of sludge, which is caused by overgrowth of certain undesirable micro-organisms that do not settle readily. They can prevail in activated sludge during landfill leachate treatment because it contains high chloride contents [27]. Such problems can be limited by co-treatment of landfill leachate and sewage in SBR to fulfill requirements for optimal C:N:P ratio and to eliminate loss of sludge organisms in the effluent, which may eventually lead to a process failure. The result of lab-scale biological treatment of the synthetic sewage and the leachate in SBR reactors is presented in Fig. 1. Blank test with synthetic sewage was running well during the whole experiment. Average COD, DOC, and ammonium nitrogen reduction in the system without



Fig. 1. Removal efficiency of COD, DOC, and ammonium nitrogen during 19 cycles of laboratory SBR operation with synthetic sewage-blank test (A) and test with the leachate (B).

landfill leachate was 91 ± 9 , 74 ± 3 , and $81 \pm 13\%$, respectively (Fig. 1(A)).

Experiment with landfill leachate started with slow adaptation of sludge to landfill leachate because adaptation to low concentrations of leachate can fairly and quickly produce sludge with greatly enhanced ability to degrade organic matter in landfill leachate [28]. First four cycles with only synthetic sewage proceeded well (Fig. 1(B)) and COD and DOC removal was always above 80 and 70%, respectively. After addition of 10% v/v of leachate, the efficiency of the system started slowly to decrease and it was stabilized after the eighth cycle, where COD and DOC removal reached up to 57 and 70%, respectively. The treatment with 10% v/v of the leachate continued until the 11th cycle; afterward leachate concentration in the system was increased to 20% v/v. As a consequence, the treatment efficiency started to decrease and after addition of 40% v/v was significantly deteriorated: COD removal was reduced to 30% and DOC removal dropped to 59%. Reduction of treatment efficiency seems to be a consequence of low adaptation of sludge to leachate or perhaps daily addition of leachate could lead to accumulation of toxic components as well as hazardous degradation products that could be adsorbed to activated sludge, or entrapped in the system affecting the activity of the sludge. Similarly, Neczaj et al. [29] evaluated that 10% v/v of mature leachate from an old landfill used for co-treatment with sewage is a practical upper limit in the SBR process. Hence, biological treatment of leachates from old sites, notwithstanding their biodegradability, is limited by the compounds generated during long-term degradation processes within the landfill body.

Ammonium removal in the reactor with the landfill leachate seemed to be not affected by the increased concentration of the landfill leachate (Fig. 1(B)). However, additional batch experiments (without activated sludge) with air stripping of the leachate diluted by synthetic sewage (20% v/v of the leachate) showed that after 24 h 63% of ammonium nitrogen was stripped out. The reason is that ammoniacal nitrogen exists in landfill leachate as un-ionized NH₃ or as ammonium ion NH_4^+ and their ratio depends on pH. When the pH is higher than seven, the concentration of ammonium ion progressively decreases and the equilibrium shifts toward NH₃, which is easily stripped out during aeration [4]. Deficient nitrification was confirmed by measurement of nitrate concentration prior and after each cycle. In the test with landfill leachate concentration of nitrate nitrogen was, after treatment, comparable to results from the blank test with synthetic sewage (data not showed), and thus the main portion of the ammonium nitrogen was probably not removed by converting ammonium nitrogen to nitrate nitrogen via nitrification, but more probably stripped out during aeration [20]. Some authors (e.g. [30]) reported excellent removal of ammonium nitrogen after biological treatment of mature landfill leachate (COD = $2,560 \text{ mg L}^{-1}$, $BOD_5/COD = 0.07$) exceeding 99%. However, all mature leachates express high pH and thus it can be expected that similarly to our results the air stripping could possibly contribute to the final removal of ammonium nitrogen.

The maximum daily volume of the leachate generated from the landfill site is 56 m^3 . A nearby local municipal wastewater treatment plant (PE = 4,000) treats approximately 1,020 m³ of municipal wastewater per day [31]. If maximal daily amount of the landfill leachate is transported and co-treated with municipal sewage, the highest concentration in the system is 6%. Based on our results, such addition of landfill leachate will not affect the efficiency of the wastewater treatment plant regarding to organic matter removal. Due to the neutral pH of municipal wastewater (typically ~7), un-ionized NH_3 from leachates is expected to partially turn into ammonium ion NH_4^+ that is less toxic and stable in water medium [32], where it is easily available for group of microorganisms that are concerned in nitrification. Hence, co-treatment of landfill leachates with municipal sewage could be an option for reduction of lechate's organic and inorganic pollution.

3.3. Anaerobic biodegradation

The next step of the study was to evaluate the possibility of anaerobic biological treatment of the leachates resulting in useful biogas production. First of all, the toxicity of the leachates to degrading microorganisms in anaerobic activated sludge was evaluated (Fig. 2).

After seven days of the experiment, carbon dioxide produced was removed by fixation with 6 M NaOH. As a result, pressure decreased. It is assumed that remaining pressure in the closed bottle arose from methane production during methanogenesis. Anaerobic degradation of glucoses in the reference system proceeded well and production of biogas reached 80% of the theoretically calculated value. This confirmed validity of the test and activity of the micro-organisms. Degradation of glucose in presence of leachate was not affected by low leachate concentrations: 16 and 32% v/v of the leachate (Fig. 2). According to our measurements, it was calculated that in the headspace of the system with glucose there is 38% of CO₂, while the rest (62%) are probably methane and other gasses. Such ratio is also typical for biogas generated in landfill (65-70% of methane with remaining 30–35% of CO₂) [33]. Similar results were obtained for glucose in presence of 16 and 32% v/v of the leachate. Higher concentrations of the leachate (72% v/v) had significant impact on anaerobic biodegradation of



Fig. 2. Toxicity of the leachate to micro-organisms of anaerobic activated sludge.



Fig. 3. Anaerobic degradation of glucose (reference compound) and the leachate.

glucose resulting in 75% inhibition of methane production in comparison to corresponding control. It means that anaerobic bacteria were able to convert glucose to CO_2 during acetogenesis (in the headspace it was 75% of CO_2), but methanogenesis was probably inhibited resulting in generation of only 25% of other gasses. Although not determined, it is expected that volatile fatty acids, hydrogen, and NH₃ appeared in the biogas [34] and were not transformed to methane due to high concentrations of ammonium nitrogen in landfill leachate which acts as inhibitor of methanogenesis during anaerobic digestion process [35].

A second set of experiments under anaerobic conditions was setup to determine biodegradability of landfill leachates. Glucose was degraded well again under experimental anaerobic conditions (76%). A higher non-toxic concentration of the leachate (32% v/v) was introduced into the biodegradability tests, which proceeded for 10 d (Fig. 3). During the monitoring period there was no production of biogas observed and it was concluded that micro-organisms are unable to degrade organic matter from the leachate. In spite of higher BOD₅ which represents biodegradable organic matter, present organics are biodegradable only in aerobic conditions, while anaerobically degradable matter was already digested within the body of landfill, and leachates from old landfills cannot be anaerobically treated.

4. Conclusion

Regarding many advantages, biological treatment of landfill leachate is often employed. However, the results of our study showed that in spite of high biodegradability displayed in ready and inherent biodegradability tests, the treatment of leachate in biological treatment plant can be accomplished up to 10% v/v of leachate in municipal sewage. In case of anaerobic treatment of landfill leachate, no biogas production was recorded. It has been concluded that conventional biological treatment is suitable for treatment of leachates only under aerobic condition and co-treated with municipal sewage.

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