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ORIGINAL ARTICLE

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Effect of leachate irrigation on methane oxidation in tropical landfill cover soil

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Abstract The effect of leachate irrigation on methanotrophic activity in sandy loam-based landfill cover soil with vegetation was investigated. Laboratory-scale experiments were conducted to investigate the methane oxidation reaction in cover soil with and without plants (tropical grass). The methane oxidation rate in soil columns was monitored during leachate application at different organic concentrations and using different irrigation patterns. The results showed that the growth of plants on the final cover layer of landfill was promoted when optimal supplement nutrients were provided through leachate irrigation. The vegetation also helped to promote methane oxidation in soil, whereas leachate application helped increase the methane oxidation rate in nonvegetated cover soil. Intermittent application of leachate (once every 4 days) improved the methane oxidation activity as compared to daily application. Nevertheless, the adverse effects of organic overloading on methane oxidation rate and plant growth were also observed.

Key words Cover soil · Landfill vegetation · Leachate irrigation · Methane oxidation · Tropical grass

Introduction

Landfill is one of the most common disposal methods for municipal solid wastes in developing countries. However, significant environmental impacts such as leachate and gas production could occur if the landfills are not designed and operated properly. Methane and carbon dioxide are the major principal gases generated and they are normally liberated directly to the atmosphere without any treatment, especially in small and medium-sized landfills. These gases are major contributors to the greenhouse effect in which methane has a global warming potential (GWP) about 23 times greater than that of carbon dioxide over a 100-year period.[1] Proper management of landfill gas is therefore one of the most important aspects of landfill operation. Although gas generation from landfills can be utilized as energy at large sites, it is not applicable in small to medium-sized landfills because the fluctuation of the gas production rate adversely affects economic infeasibility.

To control gas emissions from landfills, cover soil plays an important role in reducing the emission of methane gas. The recommended final cover may consist of multiple components, including a surface layer (vegetative support), protection layer, drainage layer, hydraulic barrier layer, foundation layer, and gas collection layer.[2] Nevertheless, it is not always possible to install all these components, especially in small landfills, due to the financial constraints in developing countries. For example, a clay layer 0.6–1.0 m in depth is commonly used for landfill closure in Thailand. The provision of a single-layered cover often allows substantial gas emission when the soil moisture level decreased to the shrinkage limit and cracking of the soil layer takes place.

The tropical climate provides unique environmental conditions that consequently influence gas production and emission from solid waste landfills. Typically, in such a climate, the extremely high precipitation rate during the monsoon is followed by a long dry period of up to 7–8 months with little rainfall. Previous observations of gas production in tropical landfills has shown that when the precipitation rate in the wet season is about four times that in the dry season, the gas production rate during the wet season could be higher by a factor of 10 than the dry season.[3] High rainfall intensity significantly affects gas emission from landfills by increasing the moisture content in solid wastes while reducing the gas permeability of the final cover soil. During the dry season, cracking of the final cover is commonly found due to the moisture content

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falling below the soil shrinkage limit. Thereby, the direct emission of methane gas to the atmosphere can take place during such a period.

In order to avoid direct methane emission to the atmosphere, the use of an appropriate final cover soil layer with effective moisture control is essential. Spraying or irrigation of water or landfill leachate on the final cover during the dry season could be used to maintain the soil water content above its shrinkage limit. Especially in tropical landfills, a reduction of leachate quantity produced during the rainy season could be an additional benefit if leachate irrigation is practiced during the dry season.[4]

Several reports have indicated the natural presence of methanotrophic bacteria, which can help convert methane to carbon dioxide, in the final cover soil.[5–7] Methane biodegradation via methanotrophic activities in topsoil has been suggested as a low-cost solution for controlling methane emission, especially from small landfills where methane gas utilization is not feasible. A number of researchers have investigated the optimum conditions for promoting biological methane oxidation in landfill final cover via laboratory-scale soil lysimeters. Some environmental conditions favoring biological methane oxidation, i.e., a highly porous soil texture,[8] appropriate soil depth,[8,9] and optimum temperature (25°–30°C)[7,9,10] and soil water content (10%–15%),[9,10] have been reported. However, significant decreases in methane oxidation rate (MOR) in long-term operation have also been observed by various researchers.[10,11] Nutrient imbalances affecting microbial activity[7,10] and excess production of methanotrophic extracellular polysaccharides (EPS)[10,11] created during unfavorable environmental conditions have been reported as the major causes for the deterioration of the methane oxidation reaction. Investigation of appropriate landfill cover design criteria and operating conditions is needed for implementing the methane oxidation concept in practice. In this regard, a field observation has suggested a positive effect of leachate irrigation on methane oxidation in landfill with vegetation.[12]

Although there have been several studies on methane oxidation in landfill cover, as mentioned above, most previous research has been done in temperate climate zones, which experience much less precipitation than tropical climate zones. This study initiated an effort to couple leachate management through irrigation practice with reduction of methane emission via biological methane oxidation in tropical landfills. The effect of leachate concentration and irrigation pattern on methane oxidation was investigated with experimentation specifically designed for developing Asian countries. Because leachate produced from fresh wastes in developing countries usually contains high concentrations of organic matter and salt, locally available salt-tolerant grasses were used as vegetation in a long-term experiment to study their effect on methane oxidation and their potential for use as landfill vegetation. The information obtained from this study will be useful to support further implementation of this concept at the field level using the methane oxidation mechanism to mitigate methane emission in tropical landfills.

Materials and methods

Experimental system

Acrylic soil columns 1.0 m high and 0.15 m in diameter were used to hold the simulated cover soil (Fig. 1). Sandy loam soil (79.6% sand, 5.8% silt, and 14.6% clay) was filled into the columns at an initial height of 0.60 m and a bulk density of 145 kg/m^3. The soil characteristics are shown in Table 1. All columns were purged with synthetic landfill gas containing 60% methane and 40% carbon dioxide through the multitube flow meter specific for inert gas (Cole Parmer) at a flow rate of 0.065 ml/s (at 25°–30°C, 1 atm), which is equivalent to a methane loading rate of 14 mol C/m$^3_{soil}$.day or approximately 100 g/m^2.day when cover soil of 0.6 m depth is used. This methane emission rate was set based on the upper boundary value of monitored data (94 g/m^2.day) at a full-scale solid waste landfill site in Thailand.[3,13]

The vegetation used in the study was chosen based on their salt tolerant characteristics. From preliminary pot experiments, two local grass species, i.e., *Sporobolus virginicus* and *Cynodon plectostachyus* were selected. The plants were acclimatized to leachate for 2 weeks in nursery pots fed with diluted leachate before being transferred into the soil columns. The initial height of plants was set at 0.10 m. Synthetic light was supplied through light bulbs installed at the top of columns (0.50 m above soil surface) for plant growth, providing a constant light intensity of about 35 000 lux.

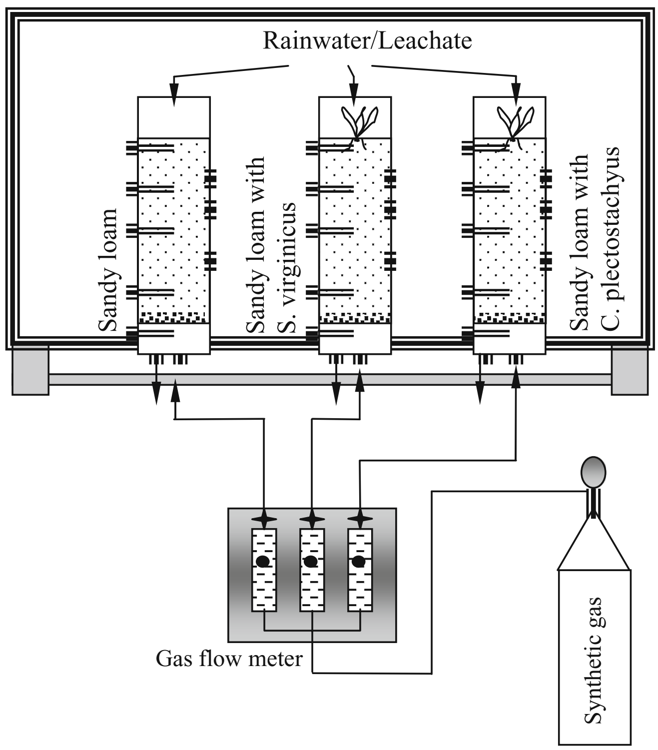


Fig. 1. Schematic of the experimental system. The plants used were *Sporobolus virginicus* and *Cynodon plectostachyus*

Table 1. Characteristics of soil and leachate used

Soil characteristics		Leachate characteristics	
pH (soil:H_2O = 1:2)	8.35	pH	5.7–6
EC (soil:H_2O = 1:2, dS/m)	0.03	EC (dS/m)	8.0–8.3
Moisture content (%)	5.2	BOD (mg/l)	13 000–16 500
Organic matter (%)	0.092	COD (mg/l)	16 000–19 000
Available P (mg/kg)	26	TKN (mg/l)	400–800
Available K (mg/kg)	104	NH_3-N (mg/l)	300–500
NH_3-N (mg/kg)	5.628	NO_3-N (mg/l)	3–8
NO_3-N (mg/kg)	0.072	Orthophosphate (mg/l)	60–80
CEC ($cmol_c$/kg)	4.2	Chloride (mg/l)	700–1500

EC, electrical conductivity; BOD, biochemical oxygen demand; COD, chemical oxygen demand; TKN, total kjeldahl nitrogen; CEC, cation exchange capacity

Two soil columns without plants were prepared as control experiments; one column was irrigated with rainwater whereas the other was irrigated with leachate. The chemical characteristics of the applied leachate are shown in Table 1. Tropical grasses (*S. virginicus* and *C. plectostachyus*) were planted into the other four columns, i.e., two columns each. They were operated under the same conditions as the control experiment. Rainwater or leachate was applied to the soil columns on a daily basis to maintain soil moisture content at 10%–15%. Leachate was diluted with rainwater to obtain a final chemical oxygen demand (COD) concentration of 1880 mg/l (i.e., 10% of the original concentration) in the first experiment (days 0–154). The COD concentration of applied leachate was then increased to 3760 mg/l (20% concentration) and 9400 mg/l (50% concentration) at day 155 and day 225, respectively. It is noted that these experimental conditions were set based on different dilution rates of raw leachate from landfill in Thailand with rainwater.

Subsequent experiments were conducted to determine the effect of leachate irrigation pattern under the same hydraulic loading rate (1030 mm/year) and organic loading rate (3.88 kgCOD/m^2.year). Three soil columns were examined by applying the optimal leachate COD of 3760 mg/l (20% concentration, from previous experiment) at an irrigation rate of 50 ml per day (50 ml/d), 100 ml every two days (100 ml/2d), and 200 ml every four days (200 ml/4d). Other soil columns were applied with leachate at concentrations of 40%, 80%, and 100% at irrigation rates of 50 ml every 2 days (50 ml/2d), 50 ml every 4 days (50 ml/4d), and 165 ml every 7 days (165 ml/7d), respectively. During leachate irrigation, rainwater was applied to the columns at application rates of 50 ml/d for the columns exposed to 40% and 80% concentrations and 27 ml/d for the 100% concentration. These operations yielded equal average hydraulic and organic loading rates in all columns. The moisture content of soil at 0.05–0.15, 0.25–0.35, and 0.45–0.55 m depth in a separate soil column irrigated with rainwater was also monitored by soil moisture sensors. The details of the experimental column are shown in Fig. 2.

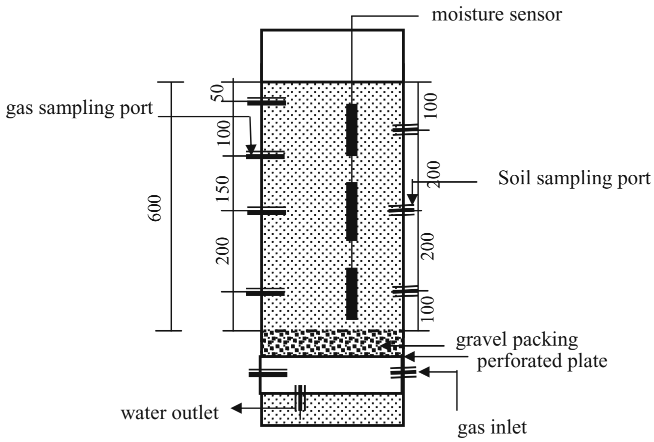


Fig. 2. Detail of experimental column. All dimensions are in mm

Monitoring parameters and sample analysis

The chemical characteristics of leachate used in this study are shown in Table 1. Analysis was carried out using the procedures described in the standard method for the examination of water and wastewater.[14] Soil properties were analyzed according to the methods described by Anderson and Ingram.[15] Soil moisture content was monitored online by moisture sensors (ECHO model EC-10, Decagon). Gas samples were collected from the sampling ports by a gas-tight syringe and the compositions were analyzed by gas chromatography (GC6890, Agilent; TCD, carrier gas: helium 1.08 ml/s; packing material: CRT1, Alltech, in double stainless steel columns). The methane oxidation rate (MOR) was used to determine the methanotrophic activity using the following equation:

$$\text{MOR (mol/m}^3\text{.day)} = Q\,[(CH_4)_{in} - (CH_4)_{out}]/V \quad (1)$$

where Q is the gas flow rate (m^3/day), $(CH_4)_{in}$ is the inflow methane concentration (mol/m^3), $(CH_4)_{out}$ is the outflow methane concentration (mol/m^3), and V is the volume of soil (m^3).

At the end of the experiments, soil samples taken from columns at depths of 0 (soil surface), 0.05, 0.15, 0.30, and 0.60 m were analyzed for extracellular polysaccharide (EPS) determination in terms of D-glucose by using the total and labile polysaccharide analysis of soils method.[16]

Results and discussion

Effect of leachate concentrations on MOR

MOR was determined in six soil columns operated with leachate irrigation at different concentrations. In the column with no plants that was irrigated with rainwater, MOR was found to be highest at 10–12 mol/m^3.day during the first 50 days and gradually declined afterwards to less than 3 mol/m^3.day after 220 days of operation (Fig. 3a). As mentioned earlier, MOR reduction had been similarly observed in other laboratory-scale experiments.[9,10] In the soil column irrigated with leachate, MOR was maintained above 8 mol/m^3.day when low concentrations of leachate (10% and 20%) was applied during the first 220 days of operation, but this figure fell to below 5 mol/m^3.day when 50% concentration of leachate was applied after 220 days. A decrease in total MOR caused by oxygen deficiency through the deeper soil layers as a result of cumulative slime, or extracellular polysaccharides (EPS), excreted from methanotrophic bacteria especially at high methane oxidation levels or high oxygen levels near the soil surface has been reported.[10] EPS production at 10–12 mg/g soil was reported as a significant level for limiting oxygen transfer into the soil and subsequently reducing MOR.[10] In this study, the EPS production at different depths of the soil column was determined at the end of the experiment, as shown in Table 2. It was found that the EPS content at a depth of 0–0.05 m in the column irrigated with rainwater was 9.3–11.4 mg/g soil, which is higher than that of the soil irrigated with leachate (7.2–9.5 mg/g soil). These results can be explained by the fact that the provision of leachate at optimal concentration could promote the utilization of oxygen by heterotrophic bacteria near the soil surface and yield appropriate oxygen levels for methane oxidizing bacteria.[17] Besides, additional nutrients supplied through leachate irrigation could prolong the high-MOR period of the system. It was found that soils irrigated with leachate had higher average nutrient contents, especially for nitrate and phosphate, which are strong determining factors for methane oxidation.[12,18] Nitrate was believed to be essential for methanotrophic bacteria, whereas high levels of ammonium and nitrite, in turn, are thought to inhibit methane oxidation.[8] However, when leachate with a high organic concentration (50%) was applied, a sharp decrease in MOR to 2–3 mol/m^3.day was observed. This was a consequence of reduced oxygen availability to methanotrophic bacteria from heterotrophic degradation of overloaded organic substances and impeded oxygen transfer because of clogged soil pores.[19] It was noted that other soil characteristics did not affect methane oxidation in this study and they appeared to be in the normal range for MOR in both columns, i.e., a pH of 7.16–8.18, an electrical conductiv-

Table 2. Soil extracellular polysaccharides (EPS, mg/g soil) in columns with rainwater and leachate irrigation

Lysimeter condition	Depth from soil surface (m)				
	0	0.05	0.15	0.30	0.60
Rainwater	9.28	11.40	5.27	6.74	7.56
Rainwater with *S. virginicus*	9.20	11.52	9.24	4.42	4.01
Rainwater with *C. plectostachyus*	9.56	5.93	6.74	8.38	5.27
Leachate	7.19	9.48	9.03	6.70	5.07
Leachate with *S. virginicus*	10.50	8.01	4.66	3.35	4.01
Leachate with *C. plectostachyus*	14.18	9.44	4.05	4.62	3.97

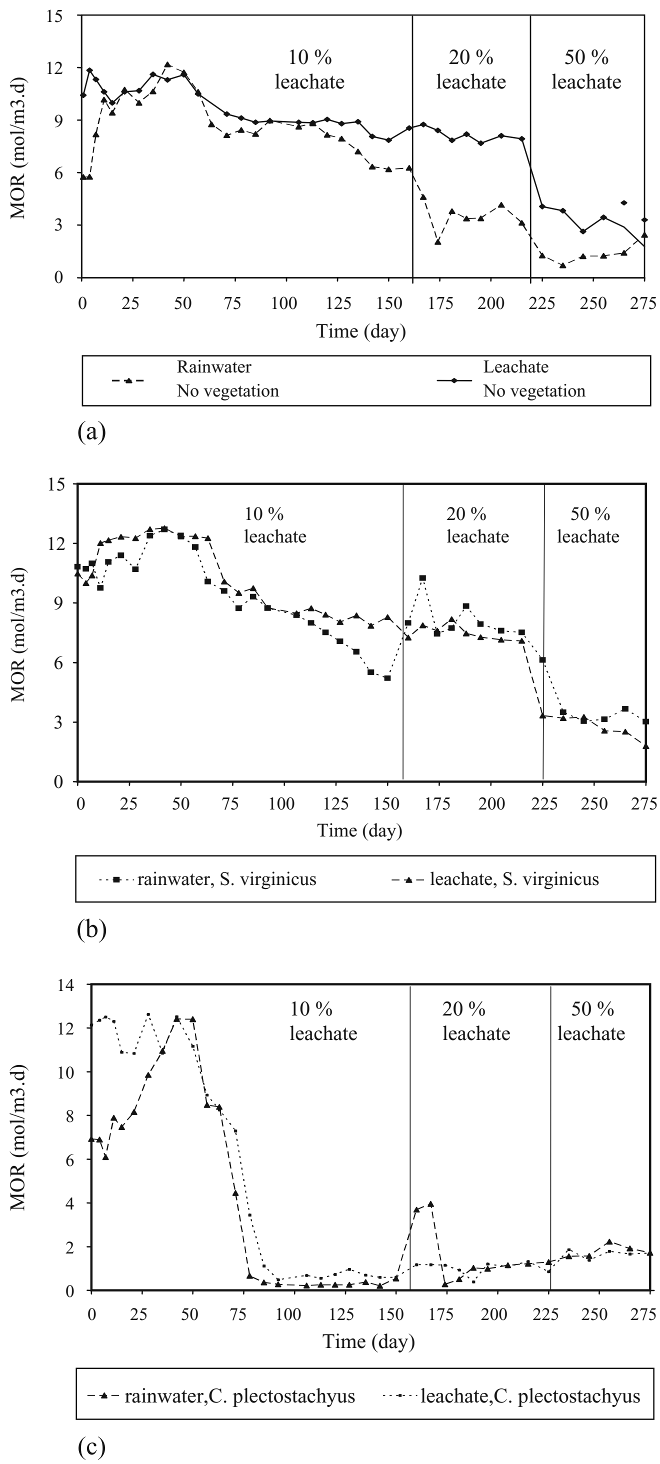


Fig. 3. Variation of methane oxidation rate (*MOR*) in the cover soil. **a** No vegetation, **b** with *S. virginicus*, **c** with *C. Plectostachyus*

ity (EC) of 0.02–0.03 dS/m, an organic matter content of 0.14%–0.22%, NH_4^+ of 4.02–4.70 mg/g soil, and NO_3^- of 0.09–0.78 mg/g soil.

Effect of vegetation on methane oxidation rate

Figures 3b and 3c illustrate MOR during rainwater and leachate irrigation in vegetated soil columns. For columns with *C. Plectostachyus*, the MOR for rainwater and leachate irrigation had similar trends as the nonvegetated column during the first 50 days, after which the MOR in the vegetated columns rapidly dropped to less than 2 mol/m^3.day. Leachate application increased competition for oxygen between plants and soil bacteria. The presence of plant roots can yield either positive or negative effect on soil microorganisms. Roots can induce oxygen diffusion from the atmosphere to the deeper soil through their penetration into the soil, or via oxygen transfer from leaves to the root system. On the other hand, plant roots also consume oxygen during respiration. The respiration rate of plant roots is usually higher than that of soils by three orders of magnitude.[20] In the present study, the adventitious roots of stolons in the case of *C. plectostachyus* commonly found near the soil surface might influence the oxygen availability for methane oxidation. Its root penetration at the soil surface could also stimulate higher EPS accumulation (9.4–14.2 mg/g soil), as shown in Table 2, due to methanotrophic activities at high oxygen levels.

For soil columns with *S. virginicus*, on the other hand, MOR gradually declined from 10–12 mol/m^3.day to 5 and 8 mol/m^3.day after 150 days of rainwater and leachate irrigation, respectively. The higher MOR observed for columns with *S. virginicus* compared with nonvegetated columns or those vegetated with *C. Plectostachyus* could be associated with the different root systems. The longer roots of *S. virginicus* could possibly provide more available oxygen in the deeper soil via its roots as compared to the nonvegetated cases for both rainwater and leachate irrigation (Fig. 4). These results show that plantation with *S. virginicus* or the use of 10%–20% leachate irrigation onto cover soil yield higher MOR. However, *S. virginicus* could not help reduce EPS content at the soil surface in all cases. Leachate irrigation did not significantly affect MOR in vegetated soil except for higher leachate concentrations (50%), where MOR was reduced to only 3 mol/m^3.day. This might be due to limitation of oxygen caused by heterotrophic organisms.[10] The adverse effects of high organic loading on MOR and plant growth was observed in all cases. As the leachate concentration was increased to 50% after day 225, MOR in all columns dropped to 1–4 mol/ m^3.day. The plants were also damaged from the overloading of organic matter, as observed by the reduction of the number of leaves and the appearance of yellow leaves. From the experimental results, it can be seen that methane oxidation was successfully sustained for more than 200 days in nonvegetated soil with 20% leachate irrigation and in landfill cover soil with *S. virginicus* plantation. Most methane oxidation took place at a depth of 0.05–0.15 m in the nonvegetated case (Fig. 4a), but the active zone was found to be deeper (0.05–0.3 m.) in cover soil with *S. virginicus* (Fig. 4b). No plants could survive the high organic loading condition; however, some methanotrophic activity in the cover soil was still detected.

Effect of leachate irrigation pattern on methane oxidation rate

Subsequent experiments were conducted by irrigating with 20% leachate on cover soil with *S. virginicus.* Table 3 shows MOR for different leachate irrigation patterns. During the experiments, the MOR gradually increased from 4 to 10 mol/m^3.day within the first 50 days. As the experiment extended beyond 50 days, MOR in cover soil undergoing 200 ml/4d of leachate application continued to gradually increase and finally reached a plateau of 12 mol/m^3.day, which was maintained until the end of experimental period. For the 50 ml/d and 100 ml/2d irrigation patterns, MOR reached a maximum value of 10.6 mol/m^3.day and finally declined to 9 mol/m^3.day. The average MOR was found to be 8.50, 9.10, and 10.05 mol/m^3.day for 50 ml/d, 100 ml/2d, and 200 ml/4d irrigation, respectively. From the experimen-

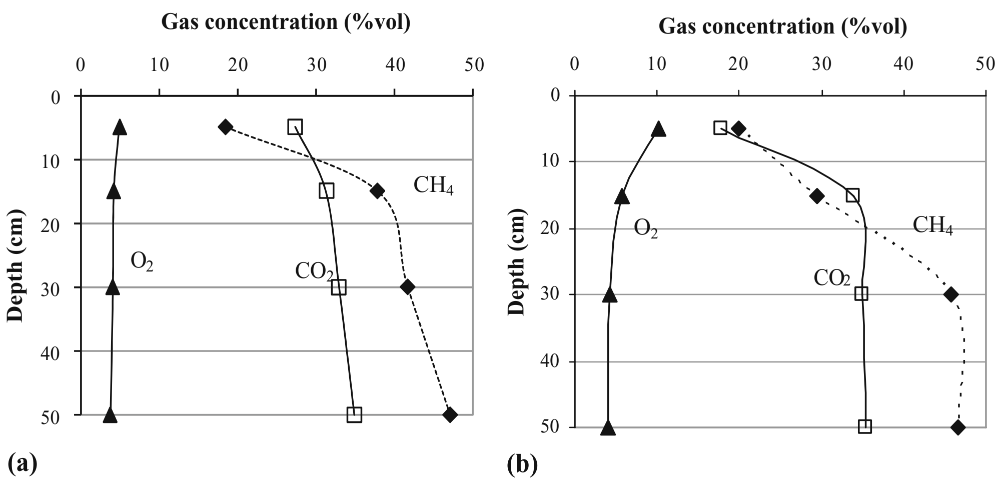


Fig. 4. Gas concentration profile in experimental column with leachate irrigation. **a** No vegetation, **b** with *S. virginicus*

tal results, it was found that intermittent application of leachate was preferable to continuous application for sustaining methane oxidation. This could be because oxygen for methane oxidation could be easily transferred into the soil during the resting period. Determination of the oxygen content of soil gas also confirmed that a higher oxygen concentration was available for methane oxidation for intermittent leachate application (Fig. 5a). Another benefit of a longer resting period between leachate irrigation is the conversion of ammonia to nitrate through nitrification processes. Moreover, the intermittent application of leachate at 200 ml/4d could maintain the soil moisture content closer to its optimum value for methane oxidation (15%) along the entire depth of the soil column, as shown in Fig. 6. Considering EPS production, 20% leachate application could reduce EPS accumulation, especially near the soil surface, to the same extent as the previous experiment. A similar benefit of leachate irrigation on methane oxidation under controlled application rates is also reported in other studies.[12,20]

When higher concentrations of leachate (40%, 80%, and 100%) were applied to the cover soil, lower MOR was observed in all cases. In the case of 40% leachate, MOR initially increased from 4 mol/m^3.day to a maximum value of 6.5–8.7 mol/m^3.day within the first 50 days and gradually declined thereafter. An average MOR of 6.4 mol/m^3.day was obtained for 40% leachate, which is slightly higher than for the two higher concentration leachates (80% and 100%). These results suggest that the organic concentration of applied leachate needs to be carefully controlled to sustain methane oxidation activities. The insufficient oxygen availability for methanotrophic bacteria under irrigation with high-concentration leachate was the major cause of methane oxidation decline. As shown in Fig. 5b, oxygen levels in soil exposed to higher leachate concentrations were found to be lower than those for lower concentrations. The MOR during the application of higher concentrations of leachate was only about 50% of that observed for 20% leachate. Therefore, a maximum concentration of irrigated leachate onto the cover soil with vegetation of about 4000 mgCOD/l is recommended as a result of this study. As shown in Table 4, EPS production became higher as the leachate concentration increased. The unfavorable conditions created by excessive organic loading from leachate could yield a higher production of EPS[21] and thus result in a rapid decline of MOR.

Table 3. Methane oxidation rate (MOR) in columns with different leachate irrigation patterns

Leachate concentration (%)	Leachate concentration (mg/l)	Irrigation pattern[a]	MOR (mol/m^3.day) Range	MOR (mol/m^3.day) Average (SD)[b]
20	3760	50 ml/d	4.3–10.2	8.5 (1.59)
		100 ml/2d	4.4–10.6	9.10 (1.88)
		200 ml/4d	3.9–12.2	10.05 (2.72)
40	7520	50 ml/2d	4.8–7.8	6.40 (1.31)
80	15040	50 ml/4d	4.4–6.5	5.17 (0.58)
100	18800	165 ml/7d	4.2–8.7	5.62 (1.69)

[a] Hydraulic loading rate was 1030 mm/year in all columns
[b] Determined over 165 days during steady period

Table 4. Soil EPS (mg/g soil) in columns with different leachate irrigation patterns

Leachate concentration (%)	Irrigation pattern	Depth from soil surface (m) 0	0.05	0.15	0.30	0.60
20	50 ml/d	7.13	7.97	6.76	7.02	5.68
20	100 ml/2d	7.88	5.99	9.43	8.15	12.35
20	200 ml/4d	5.05	7.96	5.17	9.17	8.10
40	50 ml/2d	10.46	9.18	9.02`	8.21	10.11
80	50 ml/4d	11.87	9.24	8.87	10.14	11.33
100	165 ml/7d	12.74	18.26	10.05	11.97	12.16

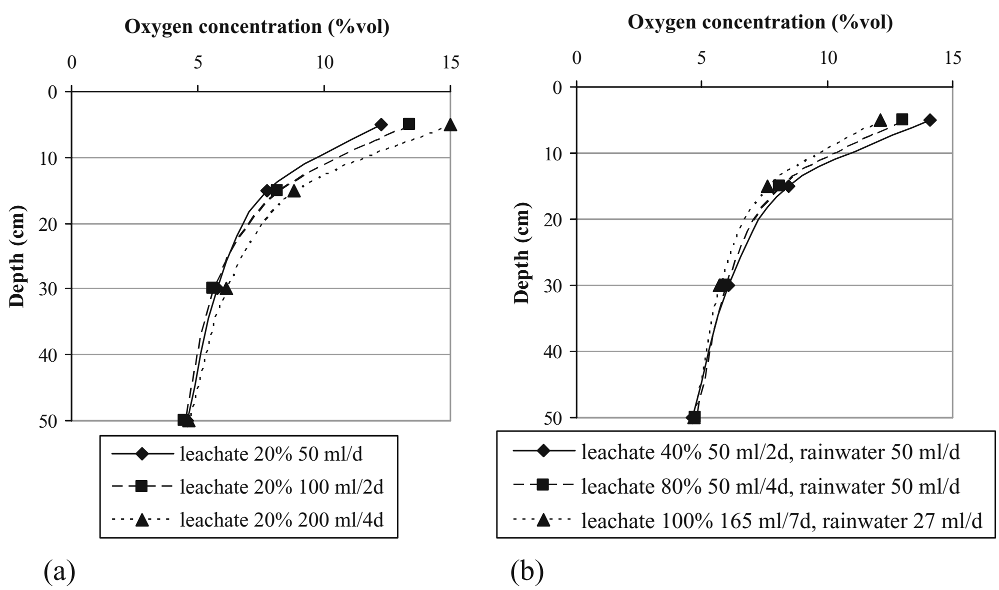
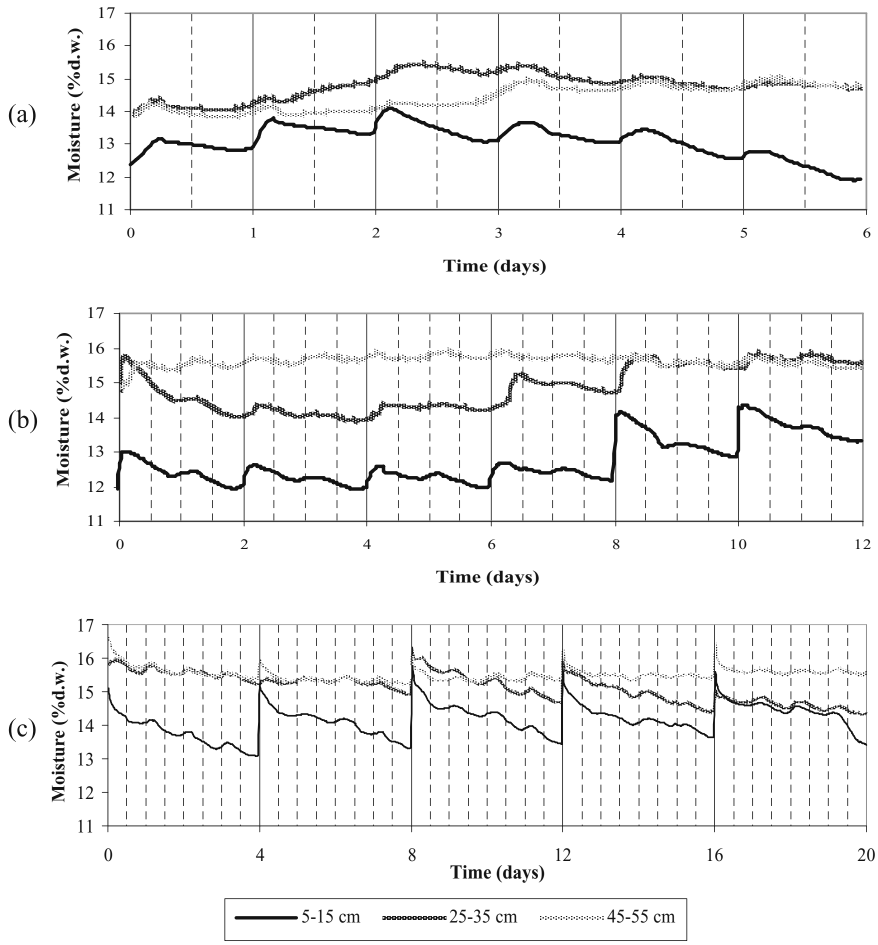


Fig. 5. Oxygen concentration in the soil column for different leachate irrigation patterns: **a** 20% concentration at 50 ml/d, at 100 ml/2d, and at 200 ml/4d; **b** 40% concentration at 50 ml/2d, 80% concentration at 50 ml/4d, and 100% concentration at 165 ml/7d

Fig. 6. Moisture profile at different depths in soil column for different leachate irrigation patterns: **a** 50 ml/d, **b** 100 ml/2d, and **c** 200 ml/4d



Conclusions

From the experimental results obtained, the following conclusions can be drawn:

1. Irrigation of landfill cover soil with leachate at an appropriate organic (COD) concentration of 4000 mg/l at a hydraulic loading rate of 1030 mm/year promotes methane oxidation in landfill cover soil. The presence of tropical grass *S. virginicus* helped enhance methane oxidation by facilitating oxygen penetration into soil through its root system.
2. When the cover soil was irrigated using leachate with organic (COD) levels above 4000 mg/l, the vegetation was severely damaged and the methane oxidation rate dropped significantly due to the shortage of oxygen available for methane oxidation.
3. The leachate irrigation pattern also affected the methane oxidation rate in cover soil with vegetation. Intermittent application of leachate promoted methane oxidation by allowing oxygen transfer into the soil during the resting period. The application of leachate at higher concentrations, although maintaining equal hydraulic and organic loading rates, resulted in a lower methane oxidation rate.

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