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Biological control of leachate from municipal landfills

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Abstract

Landfilling is still a popular way for municipal solid waste (MSW) treatment. Leachate generated from landfills is becoming a great threat to the surroundings as it contains high concentrations of toxic substances. How to control leachate migration and to protect environmental pollution is now a concern for many environmentalists. In this work, eight effective microorganisms (EMs) were isolated from wastewater, sludge and soil samples by enrichment culturing techniques and used for leachate migration control in columns and pilot experiments. The preliminary experiments reveal that the EMs could remove 25% and 40% of chemical oxygen demand (COD) from leachate in fine sand and sabulous clay columns, respectively. An aquifer system was designed to simulate in-situ control for leachate migration with EMs. The EMs were injected into the simulated aquifer and formed a permeable biological barrier. The experimental results show that the barrier removed 95% of COD and approximately 100% inorganic nitrogen, that is, nitrate-N plus nitrite-N plus ammonia-N, from the migrating leachate. CO_2 production, redox potential and microbial number were monitored simultaneously in the aquifer during the experiment to assess the EMs' activities and the effect of the bio-barrier. The data indicate that the EMs isolated in this work had high activities and were effective for organic and nitrogenous contaminant removal throughout the experiment.

Keywords: Landfill; Leachate; Effective microorganisms; Bio-barrier; COD; Inorganic nitrogen

1. Introduction

Landfilling, which is still the most popular way for municipal solid waste (MSW) treatment in China as well as in other countries now, takes up lots of land and, worse, leads to serious pollution of its surroundings. Recent estimate indicate that 209 million tons of MSW are generated annually in the US and that approximately 61% of this waste is disposed by burial in sanitary landfills (USEPA, 1996). In 1997, 1060 million tons of industrial solid waste were generated in China and that only ca. 45% were properly disposed (China Environmental Protection Bureau, 1998), mostly by burial in landfills. The hazardous gases generated from landfills contaminate the surrounding air, and the liquid pollutants (leachate) generated when water passes through the refuse are a great threat to the surrounding soil, groundwater and even surface water. Leachate is considered to be the main pollutant to the environment in a landfill. In many cases, landfill leachates are highly contaminated and have much higher concentrations of organic matter and toxic substances such as metals, quite different from domestic wastewater (James et al., 1985). External treatment of landfill leachate may require physical, chemical and biological processes for the removal of high-strength organic and inorganic materials (Keenan et al., 1984). Knox (1985), Carley and Mavinic (1991), Hosomi (1991) and Monoharan et al.

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(1992) have investigated biological treatment of leachate with high ammonia concentration, the treatment being based on pump-out of leachate. So many landfills exist, especially in China, without leachate collection systems or with faulty liner systems, which have polluted the environment. How could we solve this problem?

Conventional treatment for contaminated groundwater has involved pumping groundwater to the land surface followed by treatment and redisposal (Blowes et al., 1997). These methods have proved to be expensive and, in many cases, ineffective at achieving the proposed level of cleanup (Mackay and Cherry, 1989). Pumping and treatment cannot effectively remediate contaminated soil, which may become a secondary contamination source to the groundwater flowing through it. Blowes et al. (1995) proposed the reactive barrier as an alternative to the pump-and-treat method. Aquifer material is excavated and replaced with reaction mixtures as horizontal treatment layers (Morrison and Spangler, 1992) or as vertical treatment walls (Bianchi-Mosquera et al., 1994; Gillham and O'Hannesin, 1994; Robertson and Cherry, 1995). These engineered barriers contain reactive solids that remove contaminants from solution by various in situ transformations (Blowes et al., 1995). As one of the innovative techniques for contaminated soil and groundwater remediation, bioremediation is used to stimulate the indigenous microflora to remove subsurface pollutants, which is attractive because it has the potential to: (1) permanently eliminate contaminants through biochemical transformation or mineralization; (2) avoid harsh chemical and physical treatments; (3) operate in situ; and (4) be cost effective (Kemenade et al., 1995; Ress et al., 1998). The addition of specialized microbial populations to degrade subsurface pollutants has been incorporated into many remedial projects (Christofi et al., 1998); however, their role in biodegradation could not be distinguished from that of the indigenous microflora (Kerr, 1987).

Therefore, this research was aimed at isolating effective microorganisms (EMs) and injecting them into a simulated aquifer forming a reactive bio-barrier for in situ control of leachate migration and to distinguish the EMs' role in biodegradation from that of the indigenous microflora by comparison with the control. Some chemical indicators, such as CO_2 production, redox variation and microbial number, were monitored simultaneously during the experiment to assess the EMs' activities and the effect of the bio-barrier.

2. EM isolation and preliminary experiment

2.1. EM isolation

To remove the target contaminants from the leachate, the experimental EMs were isolated from wastewater, sludge and soil samples according to normal microbiological procedures (Zhuge and Wang, 1997). The culture media were: for bacteria isolation: (a) 0.5% glucose, 0.2% ammonium sulfate ($(NH_4)_2SO_4$), 0.1% sodium citrate, 0.02% magnesium sulfate ($MgSO_4 \cdot 7H_2O$), 0.4% dipotassium hydrogen phosphate (K_2HPO_4), 0.6% potassium dihydrogen phosphate (KH_2PO_4), the medium pH was adjusted to 7.0–7.2 by 0.1 N NaOH or 0.1 N HCl and autoclaved at 121°C for 20 min; (b) 1 g sodium nitrite ($NaNO_2$), 1 g sodium carbonate (Na_2CO_3), 0.5 g sodium chloride (NaCl), 0.5 g dipotassium hydrogen phosphate (K_2HPO_4), 0.5 g magnesium sulfate ($MgSO_4$), 0.4 g ferrous sulfate ($FeSO_4$), 1 l distilled water, the medium is autoclaved at 121°C for 20 min; (c) 10 g sodium thiosulfate ($Na_2S_2O_3 \cdot 5H_2O$), 4 g dipotassium hydrogen phosphate (K_2HPO_4), 4 g potassium dihydrogen phosphate (KH_2PO_4), 0.8 g magnesium sulfate ($MgSO_4$), 0.02 g iron, 0.4 g ammonium chloride (NH_4Cl), 1 l distilled water, the medium pH was adjusted to 7.0–7.3 by 0.1 N NaOH or 0.1 N HCl and autoclaved at 121°C for 30 min; (d) 0.5 g ammonium sulfate ($(NH_4)_2SO_4$), 0.2 g calcium chloride ($CaCl_2$), 0.5 g sodium nitrate ($NaNO_3$), 0.5 g dipotassium hydrogen phosphate (K_2HPO_4), 0.5 g magnesium sulfate ($MgSO_4 \cdot 7H_2O$), 10 g ammonium ferric citrate, 1 l distilled water, the medium pH was adjusted to 6–7 by 0.1 N NaOH or 0.1 N HCl and autoclaved at 121°C for 20 min; for yeast: (e) 10 g of bean sprouts with 100 ml water, the media was boiled for 30 min and then filtered and the filtrate was mixed with 5% sucrose (Du, 1992). The samples were inoculated into the culture media (a)–(e) for 5–7 days, then purified in the same culture media three times. Five bacterial and three yeast strains were isolated and identified according to Pelcza et al. (1977) and Lu (1993). They are: *Pseudomonas* sp., *Nitrobacter* sp., *Nitrococcus* sp., *Thiobacillus* sp., *Siderococcus* sp., *Pachysolen* sp., *Rhodotorula* sp. and *Coccidiascus* sp. These strains were mixed and acclimated for later use.

2.2. Preliminary experiment

The preliminary experiment was carried out to study the efficiency of the EMs for organic pollutant removal in columns filled with different media. The columns were made of plexiglass, 60 cm long and 10 cm in sectional diameter. Two columns were packed with fine sand and the other two were packed with sabulous clay (clay with 5–25% sand) to a height of 45 cm, respectively. One column of each medium was amended with EMs, and the other was blank for control. The columns were washed with 10% (v/v) HCl and then with deionized water to remove background effects of the medium before experiment. The EMs were injected into one fine sand column and one sabulous clay column, respectively, and acclimated for 10 days. The leachate used in the experiment was collected from a municipal landfill that was

Table 1
Composition of leachate used in the experiments

Elements	Concentrations (mg/l)
Ammonia nitrogen (NH_4^+-N)	600.0
Nitrite nitrogen (NO_2^--N)	257.5
Nitrate nitrogen (NO_3^--N)	45.0
COD	1438.3
Sulfate (SO_4^{2-})	1521.6
Calcium (Ca)	1788.6
Magnesium (Mg)	2166.0
Potassium (K)	282.0
Lead (Pb)	0.10
Copper (Cu)	0.36
Cadmium (Cd)	0.05

composed mainly of household waste (Ding, 1998). The leachate composition is shown in Table 1. During the experiment the leachate was infiltrated from the top of the columns and water samples were collected at the bottom. The experiment lasted for 25 days. Figs. 1(a) and (b) show effluent chemical oxygen demand (COD) of the water samples from fine sand and sabulous clay columns, respectively.

The control columns without EMs removed 10–40% of COD from the leachate through volatilization, sorption and abiotic transformation, while the columns amended with EMs removed more COD from infiltrated leachate than the control ones. The fine sand column amended with EMs removed 25% of COD more than the control column. The sabulous clay column removed 40% of COD more than the control one. The experimental results showed that the EMs isolated from wastewater, sludge and soil samples had high efficiency in organic pollutant removal. The abnormal peak values in Figs. 1(a) and (b) resulted from nutrient addition with EM injection.

3. Experimental principle

When a site is used for MSW disposal, a complex series of chemical and biological reactions begin with the burial of MSW (Barlaz et al., 1989; Ress et al., 1998). The landfill environment is a heterogeneous system in which different types of microorganisms coexist (Onay and Pohland, 1998). These microbial populations are capable of a variety of reactions, depending upon the prevailing environmental conditions and the organism–substrate specificity (Ragle et al., 1995). The main biological reactions in landfills are illustrated in Fig. 2 (Gu, 1993).

In addition to stimulating the indigenous microbial population to degrade organic compounds, another innovative technique is to add microorganisms with specific metabolic capabilities (Lee and Ward, 1985). Inoculation of bacteria into the subsurface for bioremediation has met with some success (Walton and Dobbs, 1980; Ohneck and Gardner, 1982; Quince and Gardner, 1982a,b; Sikes et al., 1984), though some issues concerned with this technique still remain, such as: adverse effects on human health, survival of the specialized microorganism in the environment, determination of set risk levels acceptable to the public and so on (Joyce, 1983). In our experiments, the EMs degrading target contaminants were selected by enrichment culturing; they were not genetically engineered organisms.

4. Materials and methods

4.1. Simulated landfill construction

To study the effect of EMs for in situ biological control of leachate migrating from a landfill, a three-dimensional aquifer designed to simulate the natural conditions underlying a landfill and to investigate potential leachate control was constructed with stainless steel of length 120 cm, height 60 cm, and width 50 cm. The whole system was composed of three subsystems: water-supply subsystem, treatment subsystem and monitor subsystem, as illustrated in Fig. 3.

The water-supply subsystem provided water for the simulated aquifer. Tap water was supplied at a rate of

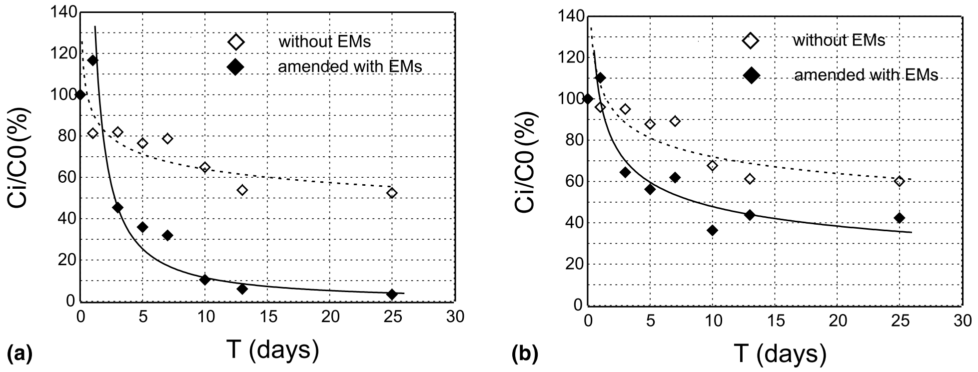

Fig. 1. Effluent COD in leachate from columns: (a) fine sand; (b) sabulous clay.

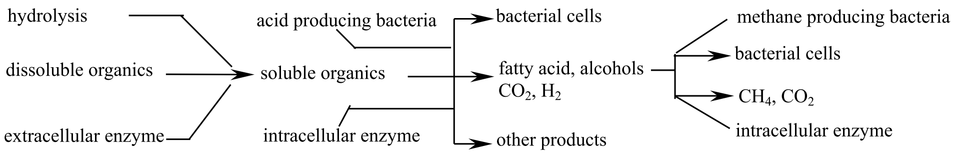

Fig. 2. Pathways of organic pollutants degradation.

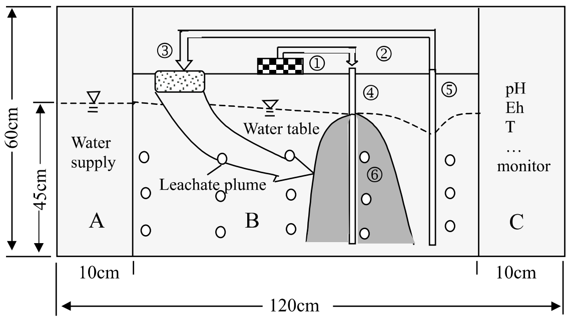

Fig. 3. Configuration of simulated system for in situ biological control of leachate. A. Water-supply subsystem, water was supplied by Mariotte bottle; B. Treatment subsysem; C. Monitor subsystem for operation conditions monitoring; 1 – EMs and their nutrient injection; 2 – recirculated water withdrawal; 3 – simulated landfill; 4 – injection well; 5 – withdrawal well; 6 – biological barrier; ○ – sampling site.

5 l/d through the use of a Mariolite bottle to keep a steady groundwater table. The leachate is the same as that used in the preliminary experiment shown in Table 1. The treatment subsystem was packed with fine sand used in the preliminary experiment and some wells constructed for leachate injection, EM injection, nutrient injection and water withdrawal. Fifteen sampling holes arranged in five columns were located along the leachate flow path at the front of the simulated aquifer with the columns 20 cm apart. The monitor subsystem consisted of a water level meter and other meters for monitoring operation conditions, such as temperature, pH value, oxidation–reduction potential (ORP), etc. The groundwater table was measured with a piezometer. A pHS-2 meter (Shanghai Instrument Company, China) was connected to the aquifer for monitoring temperature, pH and ORP on-line.

4.2. Experimental procedure and analytical methods

To distinguish the EMs causing biodegradation from the indigenous microflora, the experiment was divided into two stages: Stage A without EM injection, that is, no biological barrier existed in the aquifer, leachate was infiltrated through unsaturated zone to groundwater at the upstream and flowed in its natural condition downstream; and Stage B with EM injection, due to which a biological barrier formed in the aquifer. Leachate was also infiltrated upstream, but flowed through the bio-barrier. Through comparing the results of the two stages, the efficiency of the bio-barrier could be determined. Before each stage, the aquifer was washed with 10% (v/v) HCl and then distilled water to remove background effects of the medium. In Stage A, the leachate was injected with 0.2% $HgCl_2$ to sterilize the medium (i.e., remove indigenous microorganisms). Each stage lasted for 30 days. Water and sand samples were collected in intervals according to the experimental design. The samples were analyzed for COD, ammonium nitrogen, nitrite, nitrate, chloride, sulfate, carbon dioxide, and microbial number. COD was analyzed by oxidation by potassium bichromate. Microbial number was calculated with the most possible number (MPN). Anions were analyzed with an ion chromatograph. All other analyses were conducted according to standard methods (APHA, 1985).

5. Results and discussion

5.1. COD removal

COD was the main concern in the experiments. Figs. 4(a) and (b) illustrate COD variation upstream and downstream of the bio-barrier during Stages A and B described above.

Upstream of the bio-barrier, COD in water increased with leachate injection during Stages A and B. As there were no EMs upstream in the two stages, COD changed accordingly. Downstream of the bio-barrier, COD variations were different during two stages. During Stage A, COD increased like that upstream because there was no bio-barrier at this stage, but 60% COD was removed. During Stage B, COD decreased to a very low concentration that was only 5% of the infiltrated COD concentration, which meant that the bio-barrier removed 95% of COD from the leachate passing through it. Compared with Stage A, it was indicated that the EMs removed 35% COD from the leachate, while 60% COD was removed through abiotic processes. The simulated aquifer amended with EMs

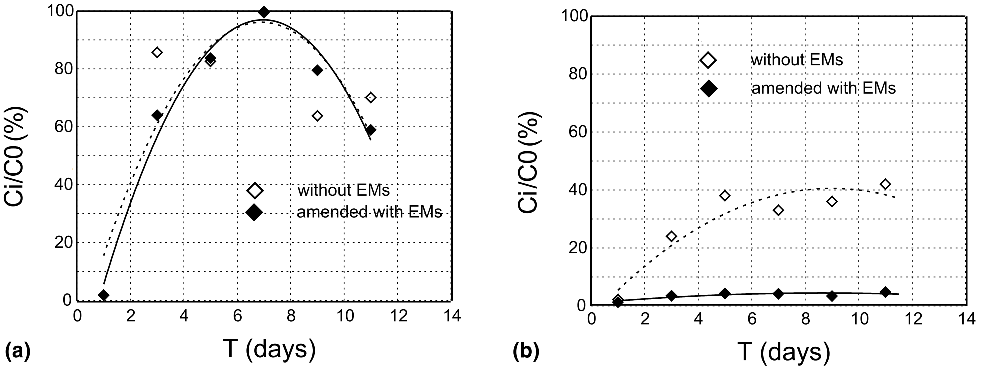

Fig. 4. Variation of COD in the simulated aquifer: (a) upstream of bio-barrier; (b) downstream of bio-barrier.

can remove more COD than the columns do because of the scale effect.

5.2. Inorganic nitrogen removal

High concentrations of inorganic nitrogen is also hazardous to groundwater and soil surrounding landfills. Inorganic nitrogen, within the context of this paper, refers to the sum of nitrate-N, nitrite-N and ammonia-N. Inorganic nitrogen removal was studied with variations of nitrate-N, nitrite-N and ammonia-N concentrations in the water samples. Figs. 5(a) and (b) reveal inorganic nitrogen variation upstream and downstream of the bio-barrier during Stages A and B.

Inorganic nitrogen variations were the same as those of COD. Upstream of the bio-barrier, inorganic nitrogen increased with time during the two stages because of infiltration of the leachate, while downstream of the bio-barrier, inorganic nitrogen decreased sharply during two stages, which indicated that the fine sand packed in the aquifer had high efficiency of inorganic nitrogen removal because of physical and chemical processes. The biological reactions could not be distinguished easily from other processes for almost 100% of inorganic nitrogen removal during Stage A (without EMs) and Stage B (with EMs). NH_4^+-N was converted into NO_3^--N, which was revealed by variations of NO_3^--N, NO_2^--N and NH_4^+-N concentrations. During an experimental interval, NH_4^+-N changed from 127.00 mg/l to 12.50 mg/l, NO_3^--N changed from 65.00 mg/l to 111.00 mg/l, and NO_2^--N changed from 50.00 mg/l to 30.00 mg/l, respectively, and the sum decreased from 242 mg/l to 153.5 mg/l, which meant that NH_4^+-N was converted into NO_3^--N, and inorganic nitrogen was also converted into other nitrogen forms, such as biomass or adsorbed by the aquifer medium.

5.3. Assessment of biodegradation

Organic and nitrogenous compounds removal from the migrating leachate may occur for a variety of reasons. Although some researchers, e.g. Sturman et al. (1995), considered that volatilization, off-site migration, sorption onto soil particles and abiotic transformation make biodegradation a difficult process to prove in situ,

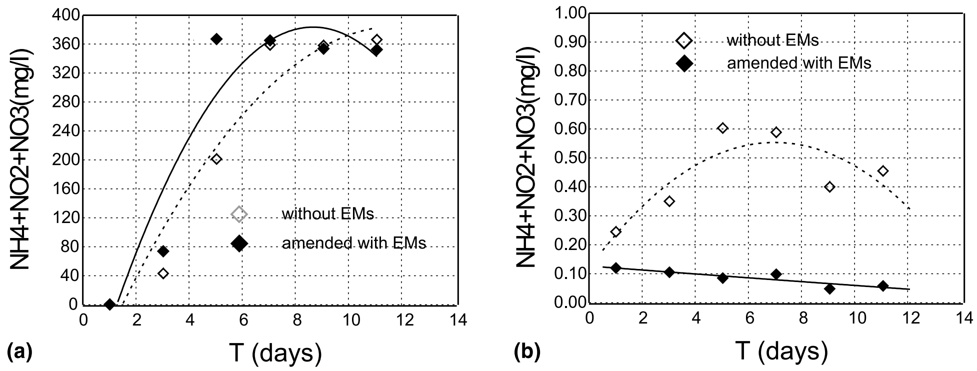

Fig. 5. Variation of inorganic nitrogen in the simulated aquifer: (a) upstream of bio-barrier; (b) downstream of bio-barrier.

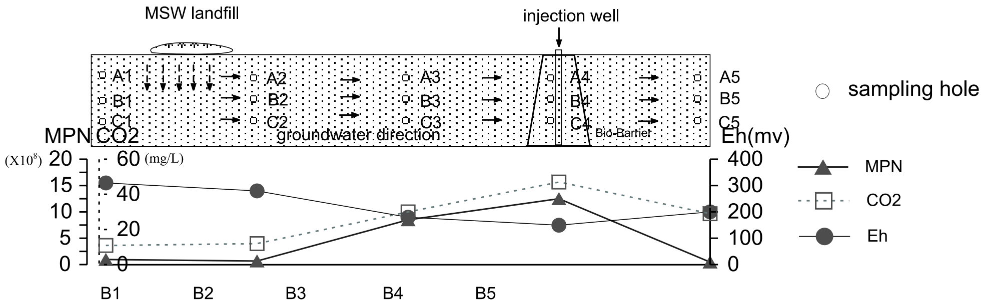

Fig. 6. Variation of CO_2 production, EM population and ORP in the simulated aquifer.

in this research, it is indicated that 35% COD removal results from biodegradation. The highly controlled conditions of the microcosm allow substantiation of the biodegradation claims, but the addition of unmeasurable contaminant sinks in field-scale systems have led researchers to the use of a variety of site characteristics to corroborate contaminant disappearance data. Many researchers have relied on contaminant disappearance and concurrent hydrocarbon-degrader population increases to prove biodegradation. Recent research has sought to determine in situ biodegradation rates through correlation with O_2 removal and CO_2 production in bioventing applications (Hinchee et al., 1991) or through observed changes in groundwater chemistry (Rifai and Bedient, 1994; Wiedemeier et al., 1994; Braddock and McCarthy, 1996; Chapelle et al., 1996; Amirbahman et al., 1998). During this experiment, besides the controls which were used to distinguish biodegradation from other contaminant-removing processes, a battery of methods, e.g., CO_2 production, groundwater ORP and MPN techniques, were taken together to offer compelling evidence that EMs are responsible for the observed contaminant disappearance. The CO_2 production, groundwater ORP and microbial MPN variations along leachate migration path are shown in Fig. 6. Where CO_2 production and MPN are high in the aquifer, the ORP is low. Also, the three indicators show good correlation with each other, which indicates that biodegradation rate in the bio-barrier is highest in the simulated aquifer.

6. Summary and conclusions

Eight EM strains with high efficiency of organic pollutant removal were isolated from wastewater, sludge and soil. Preliminary experiments revealed that the EM mixture removed 25% and 40% COD in fine sand and sabulous clay columns, respectively. A biological system for leachate control was designed and operated with in situ removal of organic pollutants and inorganic nitrogen by EMs in a simulated aquifer. EMs were injected into the aquifer with nutrient and formed a permeable bio-barrier downstream of the leachate migration path. The bio-barrier had more effective microorganisms than indigenous populations. The experimental results showed that the bio-barrier removed 95% of the COD and almost 100% of the inorganic nitrogen. Several indicators, CO_2 production, groundwater ORP and MPN techniques, were taken together to corroborate the effectiveness of the isolated EMs and the bio-barrier. It must be emphasized, however, that the demonstration of leachate control in a disturbed sand aquifer under laboratory conditions does not necessarily adequately predict the efficacy of leachate control under field conditions. Generally, in situ leachate control is dependent on soil properties and the soil microbial activities as well as on the characteristics of the landfill leachate. Anyhow, the in situ bio-barrier, compared with other remediation methods, is promising for leachate migration control.

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