The Journal of Microbiology, June 2003, p.169-173
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Vol. 41, No. 2

NOTE

Treatability Tests for the Bioremediation of Unsanitary Landfill Waste Soils

Sung-Chan Choi[2], Youn-Hee Lee[1], and Young-Sook Oh[1,*]

[1]*Department of Environmental Engineering and Biotechnology, Myongji University, Yongin 449-728, Korea*
[2]*Division of Life Sciences, Hallym University, Chunchon 200-702, Korea*

(Received February 26, 2003 / Accepted May 16, 2003)

A treatability investigation was conducted to determine if landfarming would be effective for the remediation of unsanitary landfill waste soils. Calculations based on biodegradable organic carbon contents and initial CO_2 evolution rates revealed that landfarming has a high potential for landfill site remediation and that the optimum strategy for bioremediation is site-specific.

***Key words*:** biodegradation, bioremediation, landfill, respirometry, treatability

In Korea, more than 1,000 uncontrolled or closed landfills pose potential hazards to soils, surface water and the groundwater environment (MoE, 2002a). Most landfills in Korea are unsanitary: uncontrolled, flat-topped mounds of waste, lacking pollution controls such as, top-caps, bottom liners, or leachate collecting systems. Over the last decade, considerable attention has been focused on the potential human health and environmental risks of unsanitary landfills. Today, not only must new waste containment facilities meet stringent government requirements, but many existing facilities also must be cleaned and closed, or retrofitted with pollution-reduction/prevention systems and monitored to ensure that current legal pollution requirements are met. Methods commonly used to clean up existing sites include, landfill mining and incineration, remediation, and the removal of unsanitary landfill waste to new sanitary landfill sites.

Biological methods, such as biopiling, landfarming and composting, or physicochemical methods, such as soil washing, chemical extraction and thermal treatment, could be used to remediate landfill waste (FRTR, 2002). Landfarming, in particular, has attracted considerable attention in Korea due to its low cost, simplicity, and its efficiency at degrading organic contaminants. Landfarming generally requires excavation and the placement of contaminated soils, sediments or sludges. The contaminated material is periodically turned, or tilled, to aerate the waste. Soil conditions, such as, moisture content, aeration, pH and other factors are often controlled to optimize the rate of contaminant degradation. However, careful considerations are required when controlling soil conditions. Therefore, a proper site-specific evaluation and a treatability investigation should be conducted to determine if the treatment is likely to be effective in a given situation.

The main purpose of this study was to evaluate the bioremediation of landfill waste soils by landfarming. An additional goal was identify a rapid optimal treatment strategy for landfarming by measuring CO_2 evolution as an index of the mineralization of organic compounds. Finally, the turnover time for the complete degradation of organic compounds was predicted to provide an estimate of the required treatment period.

Landfill soils were collected from closed, unsanitary, landfill sites in Gongju (GJ) and Chungyang (CY-1 & CY-2), Chungcheongnam-Do, Korea. The sites had been used for the disposal of typical municipal solid wastes, such as, food, paper, wood, etc. Representative core samples were collected at a depth of 5-6 m from each landfill site and integrated for analyses. The soils were dried for 72 h in a fume hood and then sieved through 4 mm and 2 mm screens. Soil pH, particle size distribution, water content, humic acid content and total organic carbon (in terms of weight loss on ignition (LOI) at 650°C) were determined using methods described by Carter (1993). The Hilgard cup method (Pramer and Schmidt, 1964) was used to determine the water-holding capacity (WHC) of soils, and the persulfate digestion methods (Clesceri *et al.*, 1998) were used to determine total nitrogen (T-N) and total phosphorus (T-P). To enumerate heterotrophic microorganisms, 1 g of each soil was added to 10 ml of sterilized saline solution, blended twice using a mixer at high speed for 30 s, and cooled on ice for 30 s between blendings.

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* To whom correspondence should be addressed.
(Tel) 82-31-330-6691; (Fax) 82-31-336-6336
(E-mail) ysoh@mju.ac.kr

Table 1. Selected properties of the tested soils

Soil properties	Soils		
	GJ	CY-1	CY-2
Sieve analysis			
Sand (%)	72.75	52.08	65.61
Silt + clay (%)	27.25	47.92	34.39
Physicochemical analysis			
pH	8.25	8.62	8.69
WHC (g-H_2O/g soil)	0.46±0.02	0.44±0.02	0.45±0.03
Moisture content (%, w/w)*	10.95±0.10	8.38±0.16	8.91±0.16
Weight loss on ignition (%, w/w)	7.59±0.08	5.76±0.05	10.25±0.20
Ash (%, w/w)	92.41±0.08	94.24±0.05	89.75±0.20
Humic acid (mg/g soil)	5.45	3.79	5.68
Heterotrophic bacteria (×10^8 cfu/g soil)	3.65	3.50	1.00
T-N (g-N/kg soil)	3.51	3.42	3.20
T-P (g-P/kg soil)	0.47	0.23	0.43

*Over 30% in original soil samples. The data presented were obtained after drying in a fume hood for 72 h.

The blended soil solutions were spread on Nutrient Agar media (Difco, USA) after serial dilution.

Organic compounds detectable by gas chromatography (GC) were extracted from the soils using EPA method 3550 (US EPA, 1984). Subsequently, components in the extracts were quantitatively analyzed by gas chromatography (Hewlett-Packard Model 6890 plus equipped with a flame ionization detector). The operating conditions were: carrier gas (helium) flow rate, 5 ml/min; injector temp., 320°C, split ratio 20:1; oven temp., 100°C for 3 min increasing to 300°C at 15°C/min and held for 3 min, and detector temp., 340°C.

Biometer flasks (250 ml) were used to measure CO_2 evolution, as described by Oh *et al.* (2000). Soils were moistened with distilled water to 40% and 70% of their WHC, and 50 g of the moistened soil was added to each flask. Soil samples unaffected by landfill waste were collected near the landfill sites as controls, for the measurement of endogenous respiration. Four soil treatment conditions were tested: water content of 70% of WHC (T-1), water content of 40% of WHC (T-2), addition of nutrients (T-3) and the addition of a bulking agent (T-4). Slow-release fertilizer (SRF, Chobi Co., Korea) was added to the T-3 soils to produce a C:N:P ratio of 100:20:6. Horticultural hydroballs (Haeran Co., Korea) were added to the T-4 soils at a weight ratio of 1:4. Moisture contents in T-3 and T-4 were adjusted to 40% WHC. Each soil type-treatment combination was prepared in triplicate and all flasks were incubated at 20°C in the dark. Evolved CO_2 was trapped in 10 ml of 0.1 M KOH located in the side-arm of the biometer flask. The KOH solution was taken from the biometer flask at pre-determined intervals, and the amount of CO_2 evolved was determined as described by Sharabi and Bartha (1993).

Properties of the tested soils are shown in Table 1. After 110 days of treatment, most soil properties, such as pH and counts of heterotrophic bacteria, were unaltered (data not shown). Although the humic acid fraction of the soil organic matter has been considered resistant to biodegradation, significant decreases in humic acid contents were observed in GJ (21.7%) and CY-1 (12.9%) soils. Other studies have also shown microbial degradation of the humic acids formed in landfills (Filip *et al.*, 2000; Filip and Kubát, 2001). According to these other studies, recently formed humic acids are more easily decomposed than older humic acids. Little decomposition of humic acids (0.5%) was observed in soil from CY-2, a site that had been closed for 10 years. However, a significant proportion of humic acids decomposed in soils from recently closed landfills.

The levels of GC-detectable organics in soils decreased significantly when the moisture content was adjusted to 40% of WHC (Fig. 1). The low level of GC-detectable organic compounds in the CY-2 soil was possibly due to the exhaustion of easily degradable organic compounds and the stabilization of organic compounds into humic-like substances (Filip and Küster, 1979). The decrease in the levels of both humic acid and GC-detectable organic compounds indicates an active biodegradation in the waste soils. The effects of nutrients and bulking agents were further investigated by respirometric analysis using biometer flasks.

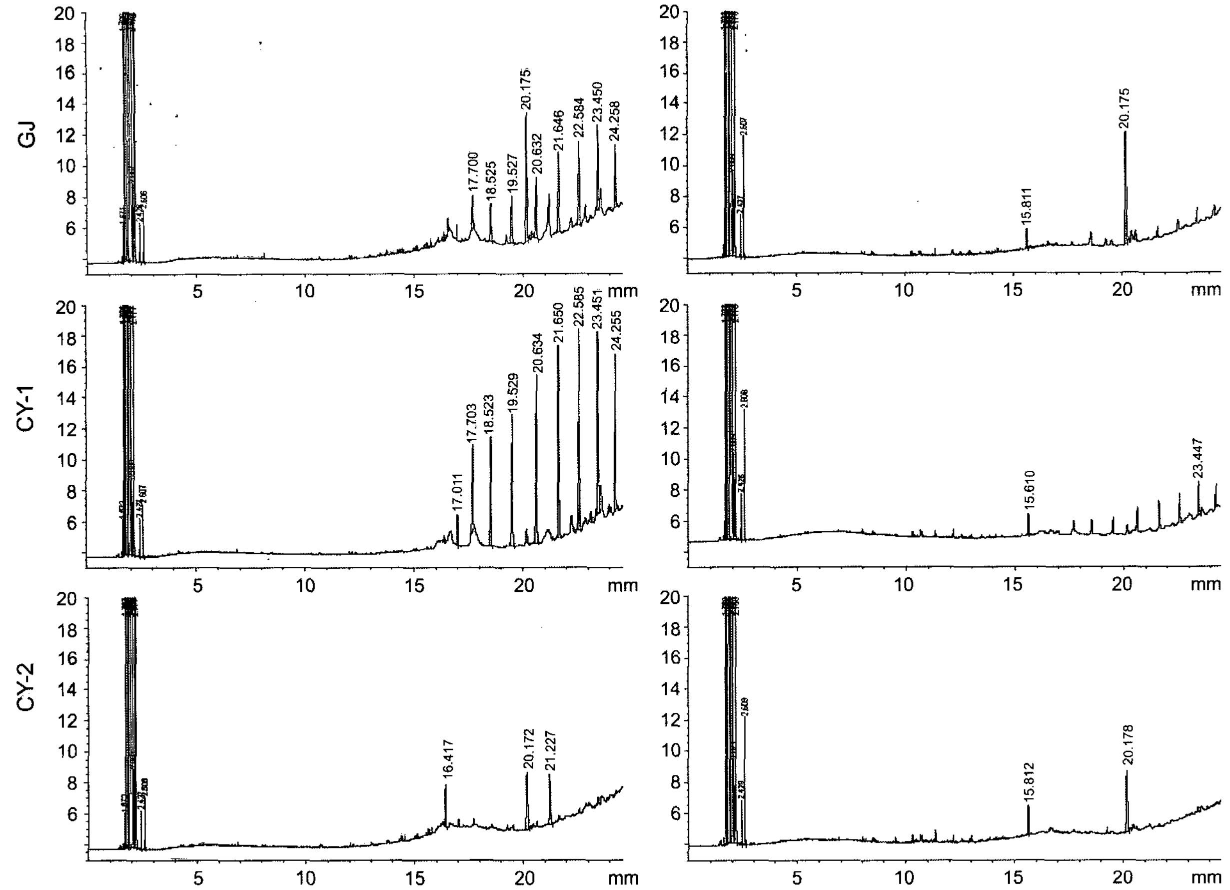

Fig. 1. Gas chromatograms of organic compounds extracted from landfill waste soils (GJ, Gongju; CY, Chungyang). Moisture levels of waste soils were adjusted with distilled water to 40% of WHC (left panel, before treatment; right panel, after 110 days of treatment). Control soils unaffected by landfill waste exhibited no significant GC peaks.

Table 2. Net CO_2 evolution and initial CO_2 evolution rate in landfill waste soils, by treatments, calculated from the results of respirometric analysis

Treatments	Net CO_2 evolution[a] (g CO_2/kg soil)			Initial CO_2 evolution rate[b] (g CO_2/kg soil/day)		
	GJ	CY-1	CY-2	GJ	CY-1	CY-2
T-1[c]	4.63	2.81	1.55	0.21	0.14	0.079
T-2[d]	5.88	3.51	1.80	0.30	0.21	0.085
T-3[e]	5.89	4.48	2.27	0.31	0.25	0.090
T-4[f]	6.36	3.74	1.83	0.34	0.23	0.083

[a] Net CO_2 evolution=total CO_2 evolved - CO_2 evolved from control soil
[b] Data obtained from 0-30 day incubation
[c] Moisture content adjusted to 70% of WHC
[d] Moisture content adjusted to 40% of WHC
[e] T-2 plus slow-release fertilizer (C:N:P=100:20:6)
[f] T-2 plus hydroballs as a bulking agent (1:4 wt ratio)

Investigations on the aerobic biodegradability of organic compounds have indicated that CO_2 evolution data can be used as a tool to evaluate treatability (Venosa *et al.*, 1992; Sharabi and Bartha, 1993; Oh *et al.*, 2000; Pagga *et al.*, 2001; Kahng, 2002). Therefore, CO_2 evolution measurements were used in this study to evaluate treatment efficiencies during the mineralization of organic compounds. The cumulative amounts of CO_2 produced by the treated soils over 110 days of incubation were significantly higher than those from the uncontaminated control soil (Table 2). Initial CO_2 evolution rates, calculated by linear regression analysis of the initial 30 days of incubation, showed the effects of treatments. High moisture content (T-1), a common property of landfill soil, was inhibitory to the biodegradation of organic compounds. The net amount of CO_2 produced by the CY-2 soil was low, as compared to the amounts from GJ and CY-1 soils, due to the lack of easily degradable organic compounds and the low bioavailability of organic compounds in CY-2 soil. Nutrient additions (T-3) generally caused an increase in the total amount of CO_2 produced and on the initial CO_2 evolution rate, as compared to T-2 soils. These results indicate that nutrient availability is an important factor limiting biodegradation, especially in CY-1 soil, which displayed the highest increase in CO_2 evolution than the other soils.

Bulking agents are materials of low density that, when added to soil, lower the soils bulk density, increase porosity, may increase oxygen diffusion, and may help to form water-stable aggregates. To enhance the degradation of organic compounds in composting or landfarming systems, bulking agents, such as wood chips, straw, and loam, are often added to increase aeration and microbial activity (Hillel, 1980; Lo *et al.*, 1993; Printz *et al.*, 1995; Chang and Weaver, 1998). In the present study, mixing soils with horticultural hydroballs resulted in increased CO_2 evolution rates and net CO_2 evolution, especially in GJ soil (Compare T-1 and T-4 in Table 2). The positive effect of aeration on biodegradation was also confirmed by an experiment in which fresh air was forced into the biometer flasks (100 ml air/min) for 10 min at 6 to 7 day intervals to exchange the air in the headspace of the flasks with fresh air. This direct aeration resulted in almost a doubling of the CO_2 evolution rate in GJ soil. However, only a marginal effect was observed in other soils (data not shown). As predicted by the high CO_2 evolution rates, oxygen limitation was a very important factor in the biodegradation of GJ soil, in which oxygen consumption rate was highest.

The results obtained were used to predict turnover time, i.e., the period required for the complete degradation of organic compounds (Table 3). Weight loss on ignition (LOI) was calculated for total organic compounds in the soils. Biodegradable organic carbon (BOC) content was calculated based on the previously reported conversion factors. Fifty-five percent of total organic carbon in municipal solid wastes is comprised of BOC (Miguez, 2000), and 28.9% of LOI is accounted for by total organic carbon (MoE, 2002b). The degradation rate of organic carbon was calculated based on the CO_2 evolution data using a respiratory quotient of 0.6 (Sharabi and Bartha, 1993; Mϕller *et al.*, 1996). For soil moisture contents of 70% of WHC (T-1), BOC turnover times were calculated to be 126, 145 and 453 days for GJ, CY-1 and CY-2 soils, respectively. Optimized treatment conditions, including the adjustment of moisture content and nutrient or bulking agent additions, effectively shortened the turnover times of BOC in GJ and CY soils (Table 3). A treatment period of more than one year was deemed necessary for cleaning CY-2 waste soil.

Landfill reclamation is a relatively new approach and has been used to expand the capacities of municipal solid waste landfills and to avoid the high costs of acquiring additional land for waste disposal. The results of the current study suggest that landfarming could be an effective countermeasure for the bioremediation and stabilization of landfill waste soils, such as GJ and CY-1 waste soils. In addition, although the effectiveness of the treatment strategies was site-specific, the treatments used in this study could play a key role in the biostabilization of unsanitary landfill sites.

Table 3. Various degradation parameters obtained from landfill waste soils[a]

Parameters	Soil[b]		
	GJ	CY-1	CY-2
Biodegradable organic carbon[c] (g-C/kg soil)	12.1	9.2	16.3
Degradation rate[d] (g-C/kg soil/day)	0.155	0.114	0.041
Turnover time[e] (days)	78	81	398

[a] Moisture contents were adjusted to 40% of WHC.

[b] A bulking agent or inorganic nutrient was added to GJ and CY soils, respectively.

[c] Assuming BOC comprises 55% of TOC, and 28.9% of LOI corresponds to TOC.

[d] Calculated based on the CO_2 evolution data using a respiratory quotient of 0.6

[e] Time required for the complete degradation of organic carbon

This work was supported by 2001 Research Fund of Myongji University.

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