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# Recycling of Remediated Soil for Effective Composting Of Diesel-Contaminated Soil

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Soil contaminated with diesel oil was remediated by the addition of remediated soil. Several mix ratios of contaminated soil to remediated soil were tested. Judging from TPH degradation rate and biochemical parameters, the optimum mix ratio (wet weight basis) was 1:1. In this mix ratio, the first order degradation rate constant of diesel oil based on TPH was 0.099/day. Degradation rate of TPH and total amount of  $CO_2$  evolved in this condition were two times larger than those of contaminated soil without adding remediated soil. The addition of remediated soil was a very effective treatment option to facilitate the degradation rate of diesel oil in contaminated soil.

### Introduction

Recycling of remediated soil has several advantages for effective composting of contaminated soil because the recycled soils usually have acclimated microorganisms. Acclimation of microorganisms on the target contaminants can significantly affect the degradation rate of contaminants in the composting process. Prior exposure of a microbial community to hydrocarbons may result in an increase of the hydrocarbon utilizing potential of the microbial community (Leahy and Colwell 1990). Also, acclimation of the microbial community to hydrocarbons such as diesel may increase the degradation rate of hydrocarbons associated with fuel oil contaminated environments.

Bioaugmentation (the addition of commercial inocula) is occasionally employed in the composting process, but its usefulness has yet to be proven and is currently an issue of controversy (USEPA 1996). Bioaugmentation may be inappropriate because introduced cultures may lack the microbial diversity that is an important factor in decomposing contaminant mixtures in natural systems. Pearce et al. (1995) indicated that differences in TPH (Total Petroleum Hydrocarbons) reduction capacity between commercial and indigenous microorganisms were insignificant. They concluded that microorganisms of indigenous microorganisms were efficient and was more cost effective compared to the commercial microorganisms for application at full scale. If possible, recycling a portion of previously composted (remediated) soil, that once contained the same contaminants, will ensure the presence of a large population of microbes capable of degrading the contaminant of concern (Peramaki and Blomker 1997; USEPA 1996). The best inoculum for remediation of hydrocarbons contamination is the contaminated soil (Cookson 1995). Minna Laine and Jørgensen (1996 1997) reported that addition of remediated soil was effective in degrading chlorophenols and PCP (Pentachlorophenol). Namkoong and Hwang (1997) reported that recycling of composted material was effective for nightsoil composting. Effect of remediated soil addition on the composting of the diesel-contaminated soil, however, has not still been reported. Also, appropriate mix ratio of contaminated soil to remediated soil has not been discussed. Therefore, this research was carried to find out the proper mix ratio of contaminated soil and remediated soil for effective composting of diesel-contaminated soil.

# Materials and Methods

#### Materials

Soil was air-dried and sieved to pass a 2mm sieve. Texture of the soil was classified as a typical sandy loam (the portion of sand, silt, and clay in soil was 59.5%, 23.0%, and 17.5%, respectively). Organic matter content of the soil was 2.6%. Field capacity of the soil was 26.1%, which was a typical value of sandy loam. CEC of the soil was 17.1 meq/100g dry soil. Remediated soil was obtained from the previous experiments (not included in this article). Organic matter content of the remediated soil was 10.4%. Field capacity and CEC of the remediated soil was 36.3% and 24.7 meq/100g dry soil, respectively.

#### Experimental Apparatus



Figure 1. Schematic diagram of experimental apparatus used for this research.

Experimental apparatus used for this research consisted of a compost reactor, two  $CO_2$  removal traps, a humidifier, and a trap for collecting  $CO_2$  evolved through biodegradation as shown in Figure 1. Carbon dioxide was removed from the incoming air so that  $CO_2$  in the exiting air was attributed entirely to decomposition (Cook *et al.* 1994). Most of the  $CO_2$  in the

incoming air was removed by reaction with soda lime (Ba(OH)<sub>2</sub>). Any residual CO<sub>2</sub> that passed through the solid-phase soda lime column was efficiently removed by reacting with 25mL sodium hydroxide (4N NaOH) in a secondary CO<sub>2</sub> removal trap. Humidifier containing 25mL distilled water was used to prevent any aspirated alkali solution from entering the compost reactor, and to raise the moisture content of the incoming air to nearly 100% relative humidity at room temperature. Air tight glass vessel of 3 liters was used as reactor for this research. Carbon dioxide removed, humidified air was entered into the compost reactor from the bottom through perforated stainless steel plate. The perforated plate was covered with 5mm diameter glass beads. Volatile compounds from the reactor were collected using a 4 mm internal diameter and 7 cm long glass tube (SKC Cat. No. 226-01) containing 150 mg of charcoal. Charcoal tube was replaced to determine volatilization rate at each sampling interval. Evolved CO<sub>2</sub> was continuously trapped in a solution of 4N NaOH.

# **Experimental Conditions**

Target contaminant of this research was diesel oil, which was spiked at 10,000 mg/kg on a dry weight basis for all samples. Experimental apparatus was placed in an incubator in which temperature was maintained at 20°C in order to minimize effect of exterior temperature variation. Aeration rate of 100 mL/min was introduced into the reactor. Moisture content of sample to be treated was adjusted to 70% of field capacity. In this research, remediated soil was mixed with fresh contaminated soil in various mix ratios. The ratios of contaminated soil to remediated soil were 1:0.1, 1:0.5, and 1:1 as wet weight bases. Composting of remediated soil-only without adding contaminated soil

was also tested to find the degradation rate of diesel oil in remediated soil as a reference. Biocide control experiment (1:1 mix ratio) by the addition of  $HgCl_2$  of 2,000 mg/kg was carried out to differentiate the biological and chemical degradation.

#### Analysis

Sample for GC analysis was extracted for 2 hr at 200 rpm at the ratio of 1 to 5 of sample to methylene chloride. Average recovery efficiency of this procedure for TPH was 98.7%. The VOCs were extracted by placing the charcoal in 8 mL screw cap vial and adding 4 mL methylene chloride to the vial. A 1µL sample of the extract was injected into a gas chromatograph (Hewlett Packard Model 5890 Series II) equipped with an integrator (Hewlett Packard Model 3395), a flame ionization detector and HP-1 column. The initial temperature was kept at 50°C for 1 minute, increased at 25°C/min to a final temperature of 300°C, and maintained at that temperature for 1 minute in order to ensure that the column was clean. The injection port and detector temperatures were 250°C and 300°C, respectively. Hydrogen gas and air flow rate for the flame ionization detector was 33 mL/min and 330 mL/min, respectively. Nitrogen carrier gas was delivered at a rate of 28 mL/min. Carbon dioxide evolved by biological reaction was collected in 4N NaOH as proposed by Stotzky (1979). An excess of barium chloride (3N  $BaCl_2$ ) was added to precipitate the carbonate as  $BaCO_3$ . After adding a few drops of phenolphthalein indicator, the unneutralized alkali titrated with 1N HCl. Carbon dioxide-uncollected NaOH was titrated as blank. Dehydrogenase activity was used as a broad-spectrum indicator of microbial activity in soil. Dehydrogenase activity was measured spectrophotometrically using the reduction characteristics of TTC (2,3,5-triphenvltetrazolium chloride) to TPF (triphenylformazan). Dehydrogenase activity was expressed in micrograms of formazan per gram of sample (µgTPF/g sample).

# **Results and Discussion**

Volatilization-corrected TPH rapidly decreased with increasing amount of reme-

diated soil added (Figure 2). In the ratios of 1:0.5 and 1:1 of contaminated soil to remediated soil, significant decrease of TPH was observed until 15 days. Significantly slow decrease of TPH was monitored in 1:0.1 mix ratio experiment and soil-only experiment without adding remediated soil. This means that recycling (addition) of appropriate amount of remediated soil was effective in the degradation of TPH. In addition, remediated soil-only experiment also showed a rapid decrease of TPH. In biocide experiment, no significant decrease of TPH was observed, which means that decrease of TPH in other experiments was primarily due to biodegradation.

The initial TPH level in the mix ratio of 1:1 of contaminated soil to remediated soil was about 9,896 mg/kg. After 30 days,



Figure 2. Variation of volatilization corrected TPH and sum of n-alkanes during composting (● remediated soil only, ▼ 1:0.1, ◆ 1:1, ■ 1:0.5, ▲ biocide control, ◆ soil only).

and n-alkanes during 50 days of composting						
	Conditions	Removal	Volatilization	Degradation		
	Contaminated soil only (Control)	66.6	2.0	64.5		
TPH n-alkanes <sup>2</sup>	$1:0.1^{(1)}$	73.8	2.0	71.8		
	1:0.5	91.7	2.1	89.6		
	1:1	95.3	1.7	93.6		
	Remediated soil only	94.7	1.4	93.3		
	Contaminated soil only (Control)	75.3	1.4	73.9		
	1:0.1	88.3	0.3	87.7		
	<sup>2)</sup> 1:0.5	98.7	0.4	98.4		
	1:1	99.9	0.3	99.5		
	Remediated soil only	99.9	0.7	99.5		

TABLE 1. Percent removal, volatilization, and degradation of TPH and n-alkanes during 30 days of composting

The value implies the ratio of contaminated soil to remediated soil.
n-alkanes is the sum of individual n-alkane ranging from C10 to C20.

TABLE 2.
Estimation of degradation kinetic parameters based on
the first kinetic model

		k <sup>1)</sup>		Half-life (day)
Conditions	(1/day)	r <sup>2)</sup>		
Contaminated soil				
only (Control)	TPH	0.037	0.97	18.7
	n-alkanes	0.052	0.92	13.4
1:0.1	TPH	0.046	0.98	15.2
	n-alkanes	0.086	0.76	8.1
1:0.5	TPH	0.080	0.97	8.7
	n-alkanes	0.153	0.80	4.5
1:1	TPH	0.099	0.96	7.0
	n-alkanes	0.205	0.63	3.4
Remediated soil only	TPH	0.105	0.91	6.6
	n-alkanes	0.223	0.52	3.1

<sup>1)</sup> k = the first order kinetic constant. <sup>2)</sup> r = correlation coefficient.

TPH concentration decreased to 630 mg/kg, which corresponded to 93.6% degradation of TPH (Table 1). This is similar to the TPH degradation in remediated soil-only experiment (93.3%). Based on the first order model, degradation rate constants of TPH were 0.105/day and 0.099/day for remediated soil-only experiment and 1:1 mix ratio experiment. Degradation rate constants of TPH in these conditions was over two times larger than that of soil-only experiment (Table 2). For 1:0.1 and 1:0.5 mix ratio experiments, first order degradation rate constant of TPH was 0.046/day and 0.080/day, respectively. This indicated that the most active degradation of TPH occurred in these experimental conditions. In case of 1:0.5 mix ratio experiment, TPH decreased from initial concentration of 9,756 mg/kg to 1,008 mg/kg during 30 days of composting, which indicated that 89.6% of TPH was degraded.

Percent degradation of TPH in the mix ratio experiment of

1:0.1 was 71.8%. This is relatively low compared to 1:0.5 and 1:1 mix ratio experiments. That is to say, it is desirable to mix remediated soil over 0.5 part in contaminated soil of 1 part in order to obtain TPH degradation over 90%.

Minna Laine and Jørgensen (1997) reported that over 90% of the chlorophenols were removed during six months of composting using remediated soil as inocula. Also in a bench-scale experiment, an average of 60% mineralization of radiolabeled pentachlorophenol was obtained within 4 weeks (Minna Laine and Jørgensen 1996). Research on the effects of remediated soil on the degradation of diesel oil has not been reported. Appropriate mix ratio of contaminated soil to remediated soil also was not discussed in the literatures. In this situation, the appropriate mix ratio of contaminated soil to remediated soil that obtained from this research has a very important engineering significance even though further research in pilot or full scale is necessary for field application of this result.

Decrease of n-alkanes concentration occurred rapidly compared to TPH (Figure 2, Table 1). In the mix ratio of 1:1, n-alkanes were degraded from the initial concentration of 2,390 mg/kg to 13 mg/kg at 30 days. This result is similar with the result of remediated

soil-only experiment (Table 1). Degradation of n-alkanes for all experiments was higher compared to the TPH degradation. This preferential degradation of n-alkanes is in line with the literatures (Chaineau et al. 1996; De Jonge et al. 1997; Frankenberger 1992; Thomas et al. 1992). The first order degradation rate also indicates preferential degradation of n-alkanes (Table 2). In 1:1 mix ratio experiment and remediated soilonly experiment, for example, degradation rates based on the first order kinetics were 0.223/day and 0.205/day, which were about two times larger than those of TPH (Table 2).

Notable decrease of individual nalkanes and isoprenoids was observed during the first 4 days for the experiments of high mix ratio of remediated soil (Figure 3). Almost all the individual n-alkanes and isoprenoids were degraded after 30 days for 1:1 mix ratio experiment and remediated soil-only experiment. In case of 1:0.1 mix ratio



Figure 3. Variation of n-alkanes and isoprenoids (pristance, phytane) during composting of 30 days (Pr: pristane, Ph: phytane).

experiment, however, n-alkanes remained in the concentration of 8 to 51 mg/kg. Pristane and phytane also remained to the concentration of 51 mg/kg and 38 mg/kg.

It is generally accepted that degradation rate of individual n-alkane compounds decreases with increasing of molecular weight. Degradation rate based on the first order kinetic model, however, indicated that no significant difference among the different molecular weight individual n-alkanes was observed in remediated soil addition experiments (data was not presented). It may be due to the effect of acclimation of microorganisms on the n-alkanes. More research is needed

to confirm this result.

Cumulative amount of  $CO_2$ evolved had not shown significant difference among experiments until 4 days, but after 9 days rapid increase of cumulative amount of CO<sub>2</sub> evolved was observed in the mix ratio 1:1 experiment (Figure 4). Total amount of CO<sub>2</sub> evolved was 7,288 mg/kg for 1:1 mix ratio experiment, which was the greatest among the experiments. This means indirectly that degradation of TPH was the most active in 1:1 mix ratio experiment. The remediated soilonly experiment showed a rather lower  $CO_2$  evolution (5,789 mg/kg) than that of 1:1 mix ratio experiment. The difference in total amount of  $CO_2$ 



Figure 4. Variation of cumulative amount of CO<sub>2</sub> evolved and dehydrogenase activity during composting (● remediated soil only, ▼ 1:0.1, ◆ 1:1, ■ 1:0.5, ▲ biocide control, ◆ soil only).

evolved between 1:1 mix ratio experiment and remediated soil-only experiment was observed, although the first order degradation rate of TPH and percent degradation were almost the same. In 1:0.1 mix ratio experiment, total amount of  $CO_2$  evolved was 4,697 mg/kg, which represented the lowest among the remediated soil experiments. This value is about two times greater than that of soil-only experiment (without adding remediated soil). Dehydrogenase activity matched well with cumulative amount of  $CO_2$  evolved (Figure 4). These results indicate that microorganisms in remediated soil are metabolically active and contribute to degradation process when added to contaminated soil.

### Conclusions

The addition of remediated soil was considered as an effective treatment options for diesel oil degradation in contaminated soil. Appropriate mix ratio (wet weight basis) was 1 part remediated soil to 1 part of contaminated soil, judging from TPH degradation rate constant and biochemical parameters. In this mix ratio, the first order degradation rate constant of diesel oil was 0.099/day and 94% of TPH was degraded within 30 days. Degradation rate of TPH and total amount of  $CO_2$  evolved in this condition were two times larger than those of soil only experiment without adding remediated soil.

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