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Effect of Sewage Sludge Mix Ratio on the Biodegradation of Diesel-Oil in a Contaminated Soil Composting

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Abstract−The objective of this research was an evaluation of amendments for supplementing organic matter for composting of diesel-contaminated soil. The materials used for this study were soil and sewage sludge, which was added as amendments for supplementing organic matter. The ratios of contaminated soil to these materials were 1 : 0.1, 1 : 0.3, 1 : 0.5, and 1 : 1 as wet weight basis. Experiments with only additives were carried out as control, and biocide control experiments were also tested by the addition of $HgCl_2$. Degradation of diesel was affected by the sewage sludge mix ratio, and the most rapid degradation of TPH was observed in the mix ratios of 1:0.3 and 1:0.5 of contaminated soil to sewage sludge. However, excess addition of these materials did not necessarily facilitate degradation rate. Appropriate mix ratio for effective degradation was 1 : 0.5 as a wet weight, and 98.1% of TPH was degraded during the composting period. In biocide control experiments, 8.3% of TPH removal of diesel oil occurred, while 95.3% of TPH was removed in experiment without adding biocide. This indicated inactivation of microbial activity for degrading diesel oil was not completely occurring although significant suppress of microbial activity was observed. Carbon dioxide evolution rate and dehydrogenase activity were matched with the degradation of diesel oil well.

Key words: Soil, Amendments, Degradation, Sewage Sludge, Diesel Oil

INTRODUCTION

Soil and groundwater contamination due to the leakage of underground storage tanks (UST) has become a serious environmental problem. Most fuel oils consist of complex hydrocarbons and contain components hazardous to human health. Soil contaminated with fuel oils, therefore, must be treated in a proper manner. At present, more than 137,000 releases from USTs have been confirmed by USEPA [Fan and Tafuri, 1994]. In the united states alone, there are about 37,000 Superfund candidates, 80,000 sites covered under the Resource Conservation and Recovery Acts (RCRA), and 25,000 Department of Defense sites in need of remediation [USEPA, 1998]. In response to these situations, the 1984 reauthorization of the RCRA mandated that the USEPA regulate USTs used to store motor fuels and hazardous chemicals. However, soil and groundwater contamination by USTs was seldom quantified in Korea. The Soil Environment Preservation Act was enacted in 1996, but criteria for the fuel oil contaminated soils are not yet established in Korea. Therefore, research on remediation technology for the fuel oil contaminated soil is required for management and remediation.

Remediation technologies for contaminated soils can be divided into physical/chemical and biological technologies. Biological remediation (bioremediation) is emerging as the most cost-effective treatment for hydrocarbon polluted soils, especially when the contaminated is a medium-distillated fuel such as diesel-oil, jet fuel, and No. 2 heating oil; and remediation options also can divided into two categories such as *in-situ* and *ex-situ* depending on whether excavation of the soil is required or not [Wang and Bartha, 1994].

There are several ways of bioremediation for excavated soils, including landfarming, bioslurry treatment, and composting. Of these treatment options, composting has advantages over the other types of technologies because it involves relatively low capital cost and operating costs, simple operation and design, and relatively high treatment efficiency. Also, composting has general advantages that contaminants are not transferred to other environmental media and the cost is low relative to other treatment alternatives. On the other hand, a potential disadvantage of composting over other bioremediation techniques is the requirement of a large quantity of amendments. These amendments can significantly increase the volume of the compost [Freeman and Harris, 1995; USEPA, 1996]. Composting can also be integrated into a treatment train [USEPA, 1996]. Although composting of yard wastes, municipal wastewater sludge, and municipal soil waste has been performed for decades, composting of soils contaminated with hazardous materials is still an emerging technology. Composting has been demonstrated to be effective in biodegrading explosives and PAHs (polynuclear aromatic hydrocarbon) in soils during full-scale applications [Berg and Eggen, 1991; USEPA, 1996]. Park [1996] studied the applicability of composting for degrading phenolic compounds. Several researchers have demonstrated the possibility of composting of soils contaminated with petroleum hydrocarbons [Beaudin et al., 1996; Stegmann et al., 1991]. Thermal treatment of petroleum contaminated soil remediation by a fluidized bed desorber at higher temperature was carried out by Lee et al. [1999]. Hwang et al. [1998] and Oh et al. [1999] also studied thermal degradation of waste in which source

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materials and reusable oil are recovered. In contaminated soil composting, organic amendments including manure, yard wastes, and food-processing waster are often added for the contaminated soil composting to increase the amount of nutrients and readily degradable organic matter which are normally deficient in soil [Potter et al., 1997; USEPA, 1996]. The ratio of contaminated soil to organic amendments is important because a high ratio may retard or inhibit microbial activity [Thomas et al., 1992]. The major organic amendments used in other studies were manure and yard waste. Municipal sewage sludge has great potential as energy and nutrient source in contaminated soil composting because of abundant organic matter and inorganic nutrient. However, the effect of sewage sludge addition on the contaminated soil composting and proper mix ratio of sewage sludge was not available. Stegmann et al. [1991] studied the effect of compost addition on the composting of diesel contaminated soil. But proper mix ratio of compost can vary depending on the kinds of compost, compost age, and concentration of diesel oil. Therefore, appropriate mix ratio of contaminated soil to these organic amendments should be identified.

Diesel oil of the major fuel oil is widely used as fuels for commercial trucks, railroad locomotives, and industrial engines. Diesel oil is also used as solvent to clean moving parts because the paraffins in diesel oil form a waxy coat that protects the metal parts from corrosion. Widespread use of diesel oil may result in numerous occurrences of diesel-contaminated soil, although information for diesel-contaminated soil is not currently available in Korea. Therefore, diesel oil was selected as representative petroleum hydrocarbon for this research. The main objective of this research is to evaluate the effects of organic amendments on the degradation of diesel oil, and to find the optimum mix ratio of sewage sludge with contaminated soil.

MATERIALS AND METHODS

1. Materials

Diesel oil was obtained from a gas station in Seoul. Average content of n-alkanes ranging from C10 to C20 was 23.5%. Normal alkanes up to 17 carbons in length are liquids, but alkanes with 18 or more carbons in length are actually solids at room temperature. The soil was collected from the top 15 cm of the soil surface in an open yard. The collected soil was air-dried and passed through a 2 mm sieve. The portion of sand, silt, and clay in the soil was 59.5%, 23%, and 17.5%, respectively. The texture of the soil was classified as a typical sandy loam according to USCS [Fuller and Warrick, 1985]. Table 1 shows the characteristics of soil, and sewage sludge used in this study. As shown, the soil contains organic matter of 2.6%. Sewage sludge contains a significantly larger amount of organic matter compared to the natural soil (45.2% and 43%, respectively). Field capacity is defined as water content after a certain period of

Table 1. Characteristics of soil and sewage sludge used for this study

Parameters		Soil	Sewage sludge
Moisture content (%)		13.8	66.0
Field capacity (%)		26.1	76.4
Volatile solids (%)		2.6	45.2
pH		6.3	7.8
Cation exchange capacity (meq/100 g dry matter)		17.1	34.3
Ultimate analysis	C	0.32	8.85
	H	0.28	2.42
	O[a]	1.92	32.70
	N	0.06	1.04
	S	0.02	0.19
	Ash	97.4	54.8
Heavy metals (mg/kg dry matter)	Pb	111.9	80.1
	Zn	408.7	211.6
	Cu	124.6	371.4
	Ni	113.2	44.0
	Cd	N.D.[b]	N.D.

[a]O=100-(C+H+N+S)-Ash
[b]N.D.=Not Detected.

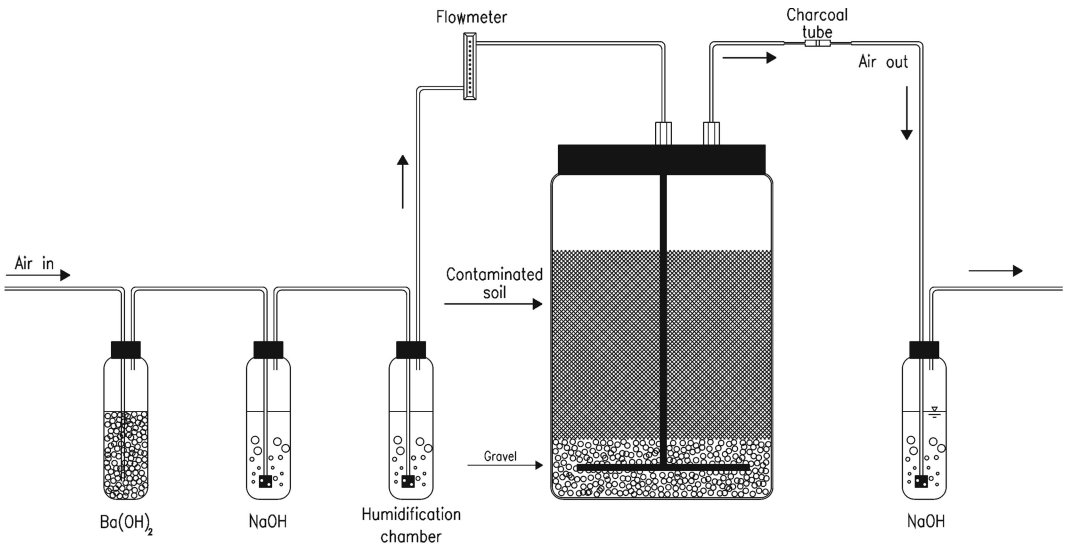

Fig. 1. A schematic diagram of experimental setup used for this research.

gravity drainage, and optimum moisture content for microorganisms has been estimated from the field capacity. The field capacity of the soil was 26.1%, and sewage sludge was 76.4%.

2. Experimental Apparatus and Conditions

Experimental apparatus used for this research consisted of a reactor, two CO_2 removal traps, a humidifier, and a tap for collector CO_2 evolved from biodegradation. Carbon dioxide was removed, by reaction with soda lime, from the incoming air so that CO_2 in the exiting air was attributed entirely to decomposition. An air-tight glass vessel of 2 liters was used. Volatile compounds from the reactor were collected by using a glass tube containing charcoal. Charcoal tube was replaced to determine volatilization rate at each sampling interval. Evolved CO_2 was continuously trapped in a solution of 4 N NaOH [Stotzky, 1979]. The experimental apparatus was placed in an incubator in which temperature was maintained at 20 in order to minimize effect of exterior temperature variation. A schematic diagram of experimental setup is shown in Fig. 1.

The aeration rate to meet the oxygen concentration of 5 to 15% by volume is in the range of 100 mL/min was chosen for this research. Fresh diesel oil was spiked at 10,000TPH (total petroleum hydrocarbon) mg/kg sample on dry basis. In order to minimize incomplete distribution of diesel oil in the sample, an appropriate amount of diesel oil was spiked to every sample of 100 g as a dry weight basis. Samples were mixed thoroughly after application of diesel oil and placed in the reactor. Moisture content of sample was controlled at 70% of field capacity based on previous studies [Frankenberg, 1992; Peramaki and Blomker, 1997]. Sewage sludge contains readily degradable organic matter as well as abundant nitrogen and phosphorus. Therefore, sewage sludge has a great potential to be used as energy and nutrient sources in bioremediation of contaminated soil. The ratios of contaminated soil to sewage sludge were 1 : 0.1. 1 : 0.3, 1 : 0.5, and 1 : 1 as wet weight basis. Also, a sludge-only experiment without mixing contaminated soil was carried out to find degradation rate of diesel oil in only sludge. In case of 1 : 0.5 mix ratio, a biocide control experiment by addition of $HgCl_2$ of 2,000 mg/kg also was performed to discriminate chemical and biological degradation.

3. Sampling and Analytical Methods

An incubation period of 30 days was selected, and sampling was carried out at day 0, 4, 9, 15, 22, and 30. After sampling, moisture contents of the sample were immediately measured to convert analytical data from wet weight basis to dry weight basis. Carbon dioxide collection trap was replaced at day 1, 2, 4, 6, 9, 15, 22, and 30 in order to monitor initial rapid degradation of diesel oil.

The soxhlet extraction and then GC analysis methods were used to analyze the TPH from the samples. The soxhlet extraction procedure was followed in accordance with the method modified by Namkoong [1988] except for the concentration step. Recovery efficiencies were in the range of 91 to 94% when the sample was extracted for 16 hours. In case of 4 hours and 8 hours extraction, recovery efficiencies was relatively lower than those of 16 hours, which was below 90%. An excess of Barium chloride (3 N) was used to precipitate the carbonate as $BaCO_3$ in determination of CO_2 evolved. After adding a few drops of phenolphthalein indicator the unneutralized alkali was titrated with 1 N HCl. The following formula was used to calculate the amount of CO_2 evolved. Carbon dioxide uncollected NaOH was titrated as blank.

$$\text{Milligrams } CO_2 = (B - V) \times N \times E$$

Where B is volume (mL) of acid to titrate the blank, V is volume (mL) of acid to titrate the CO_2 collected sample, N is Normality of acid, and E is equivalent weight, which is 22. The amount of CO_2 evolved in all experiments was corrected by subtracting CO_2 evolved by the degradation of organic matter in the only additives. Dehydrogenase activity was measured spectrophotometrically by using characteristic TTC (2,3,5-triphenyltetrazolium chloride) reduced to TPF (triphenylformazan). Dehydrogenase activity also was corrected by using the data of the only additives. A five-tube MPN technique was used to estimate the number of hydrocarbon utilizing microorganisms [Song and Bartha, 1990].

RESULTS AND DISCUSSION

1. Biodegradation

Effect of sewage sludge mix ratio on the degradation of diesel oil is shown in Fig. 2. Diesel concentration was expressed as TPH (total petroleum Hydrocarbons). Significant degradation of TPH was observed in the early stage of composting reaction (within 15 days) in all sewage sludge addition experiments. After significant degradation of TPH occurred in first 15 days, residual TPH was degraded slowly compared to the early stage. Degradation of diesel was affected by the sewage sludge mix ratio and the most rapid degradation of TPH was observed in the mix ratios of 1 : 0.3 and 1 : 0.5 of contaminated soil to sewage sludge. In the mix ratio of 1 : 1 contaminated soil to sewage sludge, degradation of TPH was relatively slow compared to mix ratios of 1 : 0.3 and 1 : 0.5 of contaminated soil to sewage sludge.

Table 2 shows total TPH removal, volatilization and degradation in this research. Volatilization-corrected TPH degradation was

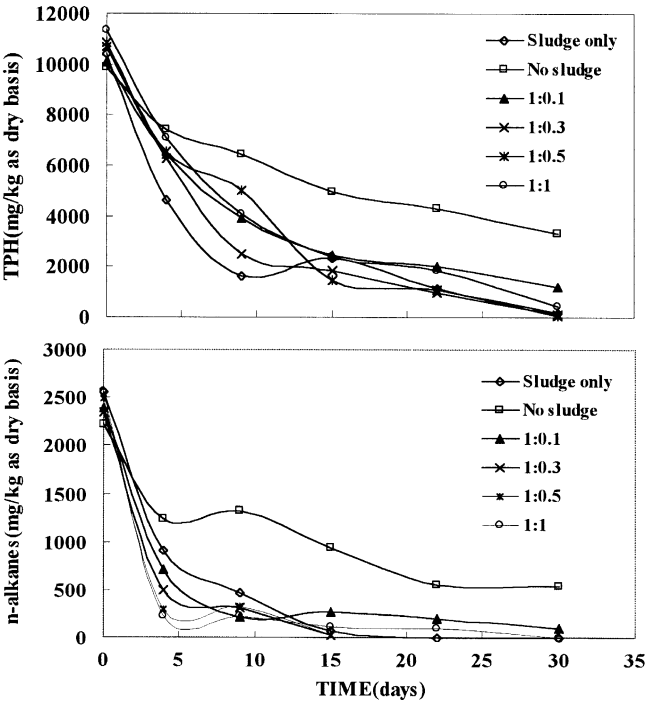

Fig. 2. Variation of volatilization corrected TPH and sum of normal alkanes in the sewage sludge addition study.

Table 2. Total removal, volatilization, and degradation in the sewage sludge addition study

Mix ratio		Total removal[a]	Degradation[a]
Sludge only	TPH	98.2	95.8
No sludge		66.6	64.5
1 : 0.1		88.1	86.0
1 : 0.3		99.0	98.1
1 : 0.5		99.5	98.1
1 : 1		95.9	94.6
Sludge only	Normal alkanes	99.8	99.1
No sludge		75.3	73.9
1 : 0.1		95.7	93.9
1 : 0.3		100.0[b]	99.9
1 : 0.5		100.0[b]	99.9
1 : 1		99.7	99.7

[a]These values were calculated from the result of the 30 days experimental period.
[b]These values were calculated from the result of the 22 days experimental period.

the greatest when sewage sludge was mixed in the ratios of 0.3 part and 0.5 part to 1 part contaminated soil on a weight basis. The initial TPH levels in the mix ratios of 1 : 0.3 and 1 : 0.5 of contaminated soil to sewage sludge were 10,685 mg/kg and 10,839 mg/kg, respectively. After 30 days, TPH concentration decreased to 203 mg/kg and 209 mg/kg, respectively. These present a 98.1% degradation of TPH during the composting period. The first-order degradation rates indicated that most active degradation of TPH occurred in the ratios of 1 : 0.3 and 1 : 0.5 of contaminated soil to sludge (k=0.120/day for 1 : 0.3 mix ratio, k=0.124/day for 1 : 0.5 mix ratio). This is about three times larger than that of no sludge experiment where sludge was not added (k=0.037/day). This also means that the addition of sewage sludge was very effective for the degradation of diesel oil contaminated oil. Demque et al. [1997] reported that the first order removal rate of diesel in land treatment of sand contaminated with the concentration of 10,000 mg/kg was 0.099/day. The first order removal rate of diesel oil in 1 : 0.3 and 1 : 0.5 mix ratio experiment (k=0.136/day for 1 : 0.3 mix ratio, k=0.145 for 1 : 0.5 mix ratio) was significantly larger compared to the result of Demque et al. [1997]. This means that the addition of sewage sludge was very effective for the degradation of diesel oil in contaminated oil.

In the mix ratio of 1 : 1 in which the most amount of sewage sludge was contained, TPH showed degradation from an initial concentration of 11,326 mg/kg to 615 mg/kg at the end of the 30-day composting period. This indicated that 94.6% of TPH was degraded during 30 days. In 1 : 1 mix ratio experiment, degradation rate was 0.096/day, which is relatively small compared to 1 : 0.5 and 1 : 0.3 mix ratio experiments. These results implied that degradation of diesel was enhanced by addition of sewage sludge. However, degradation rate of TPH was not necessarily enhanced as the amount of sewage sludge added increased, which indicates a proper mix ratio of contaminated soil to sewage sludge could exist. In case of 1 : 0,1 mix ratio, TPH concentration exhibited a value of 1,410 mg/kg after 30 days, which indicates a degradation of about 86.0%. In this experiment, the first order degradation rate was 0.072/day, which was significantly large compared to no sludge experiment.

Thomas et al. [1992] suggested that addition of organic amendments could increase degradation rate of target contaminants, but might inhibit degradation rate depending on the amount of organic amendments added. The carbon source must not represent a preferential carbon source which presents degradation of the target contaminants [LaGrega et al., 1994]. In other words, when the added carbon source was preferentially degraded over target compounds, microbial activity for degrading contaminants may have been inhibited [Cookson, 1995]. In this research, it is considered that sewage sludge added as carbon source had not been used as a competing energy source, but has increased degradation rate of the diesel oil. This fact was induced from high degradation rates of sewage sludge addition experiments compared to the no sludge study.

Normal alkanes were degraded more rapidly than TPH and total degradation also was significantly higher than TPH as shown in Fig. 2. At the same condition, removal of normal alkanes was in the range of 95.7 to 100%, and degradation was in the range of 93.9 to 99.9%. Significant differences in degradation of normal alkanes were not observed in various mix ratio experiments except the 1 : 0.1 mix ratio study. Although percent degradation in the mix ratio of 1 : 0.1 of contaminated soil to sewage sludge was the lower among the sewage sludge addition experiments (93.9%), it was still greater that that of no sludge experiment (73.9%). These results indicated

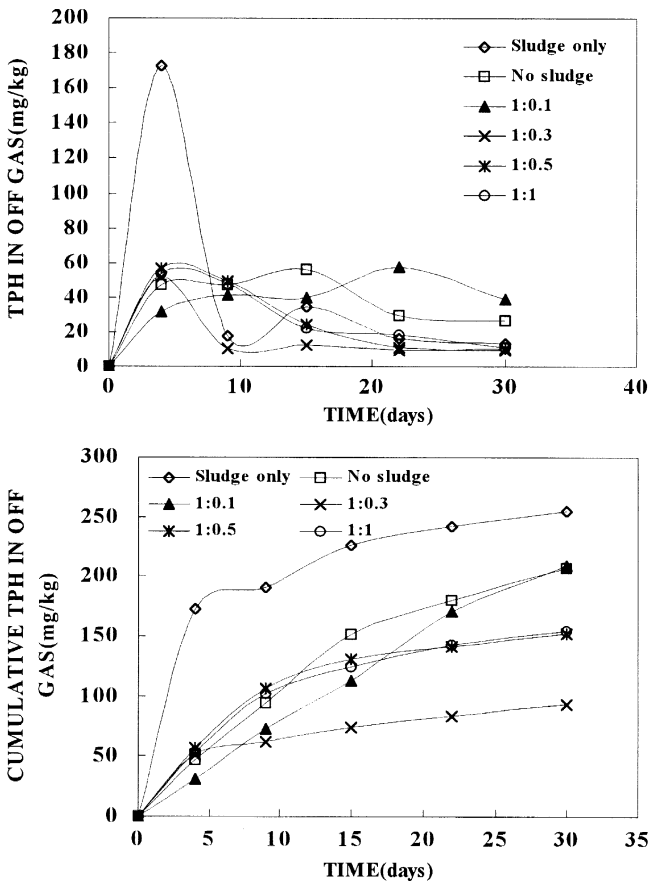

Fig. 3. Variation of TPH and cumulative TPH in off-gas in sewage sludge addition study.

that normal alkanes were preferentially degraded compared to other components in TPH [De Jonge et al., 1997]. Normal alkanes in the mix ratios of 1 : 0.3 and 1 : 0.5 of contaminated soil to sewage sludge, especially, were completely degraded in composting period of 22 days. This degradation pattern in these mix ratios was in line with the results of TPH degradation. The first order degradation rate also indicated preferential degradation of n-alkanes. In the mix ratio of 1 : 0.5, degradation rate of n-alkanes was 0.252/day, which was about two times larger than that of TPH. In addition, degradation rate was 0.183/day and 0.188/day for sludge only experiment and 1 : 1 mix ratio experiment, respectively.

2. Volatilization of Diesel Oil

Fig. 3 illustrates a variation of TPH and cumulative TPH in off-gas. Volatilization tended to increase rapidly in the early stage of composting. For the 1 : 0.3 mix ratio experiment, TPH of about 1.5 mg/kg was volatilized until 4 days and then after volatilization loss pattern of TPH decreased sharply to about 10 mg/kg. Similar volatilization loss pattern was observed in the mix ratios of 1 : 0.5 and 1 : 1 of contaminated soil to sewage sludge. In these experiments, volatilization of TPH was observed in the concentrations of about 50 mg/kg until 9 days, and then decreased dramatically. This is an inverse proportion to the result of TPH removal and degradation, which indicates active degradation activity in the early stage of composting may result in initial high volatilization loss. In the mix ratio of 1 : 0.1 of contaminated soil to sewage sludge and no sludge experiment without adding sewage sludge, volatilization loss continued to the concentration of 30 to 50 mg/kg until 30 days. These results show that appropriate sewage sludge addition in contaminated soil will increase degradation rate of TPH as well as decrease volatilization loss of TPH.

Cumulative volatilization loss shows more distinctly the difference of the amount of TPH volatilized depending on sewage sludge mix ratios as shown in Fig. 3. In the mix ratio of 1 : 0.3, cumulative volatilization loss was increased dramatically until 4 days and changed little thereafter. Mix ratio experiments of 1 : 0.5 and 1 : 1 showed a rapid increase until 9 days and changed little thereafter. Continuous increase of volatilization loss in mix ratio 1 : 0.1 and no sludge experiments was observed until the end of the composting period. Consequently, total cumulative amount lost by volatilization was equal to 2.1% of initial TPH concentration, which in-

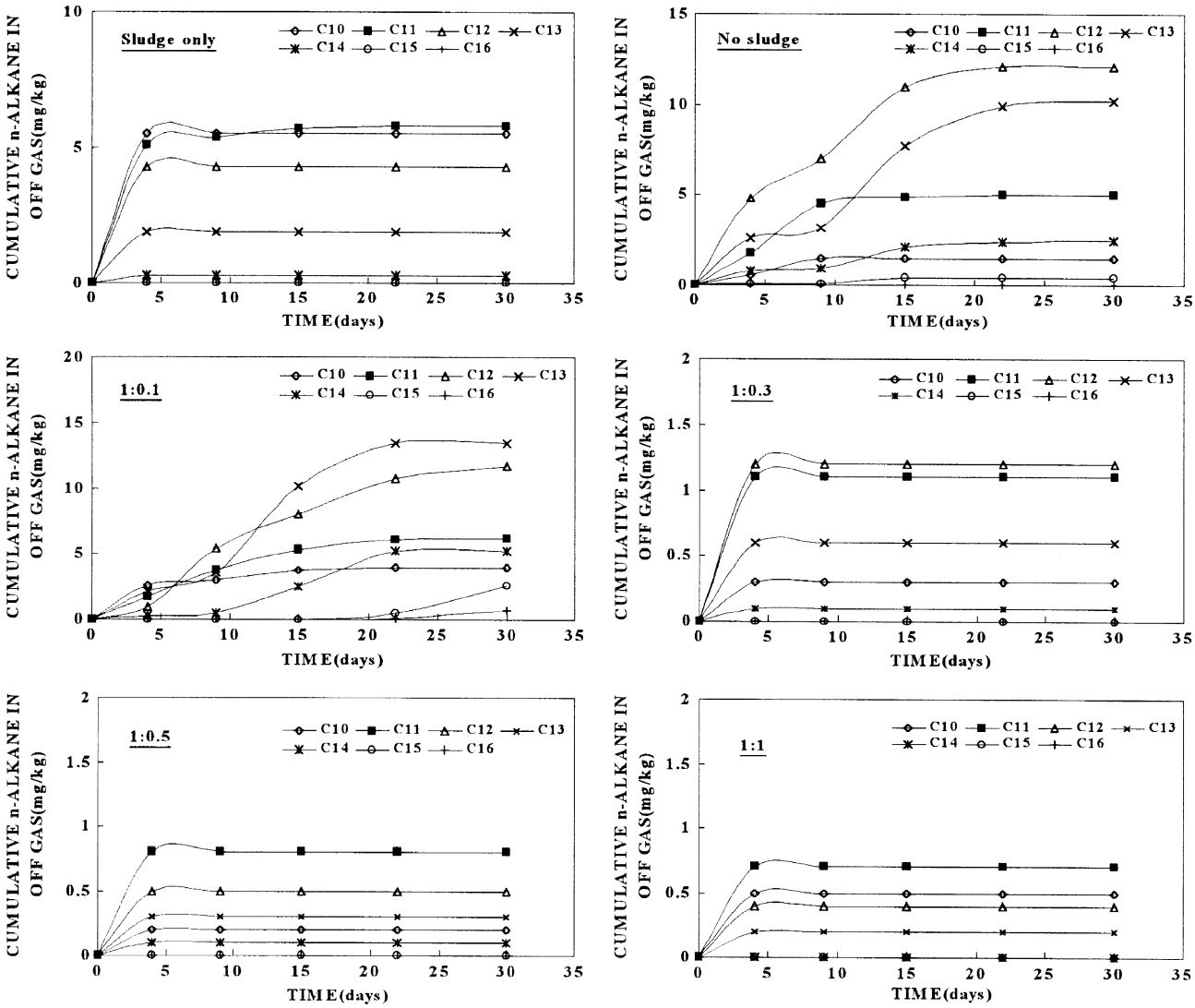

Fig. 4. Variation of cumulative normal alkanes in off-gas in sewage sludge addition study.

dicated the greatest among the sewage sludge addition experiments. Total volatilization losses in all sewage sludge addition experiments were in the range of 0.9 to 2.4%, which indicated contribution of volatilization on removal of diesel oil in contaminated soil was much less significant than that of degradation. Pitrowski et al. [1992] suggested that volatile compounds comprise approximately 30% of diesel oil by weight and so volatilization may have accounted for a loss of 30% of the TPH contamination from the soil.

Fig. 4 indicates that normal alkanes in volatilized TPH mainly consisted of components in the range of C10 (normal decane) to C16 (normal hexadecane). Especially, normal alkanes ranging from C10 to C14 were significantly larger than C15 and C16 in volatilization loss. This is caused by the difference of vapor pressure. De Jonge et al. [1997] reported normal alkanes ranging from C16 to C20 were not lost by volatilization. In the mix ratio experiment of 1 : 0.1 of contaminated soil to sewage sludge, as its molecular weight increased, volatilization of individual normal alkanes decreased. Besides, in other mix ratio experiments including 1 : 0.3, 1 : 0.5 and 1 : 1, normal alkanes were lost by volatilization until 4 days, and changed little thereafter.

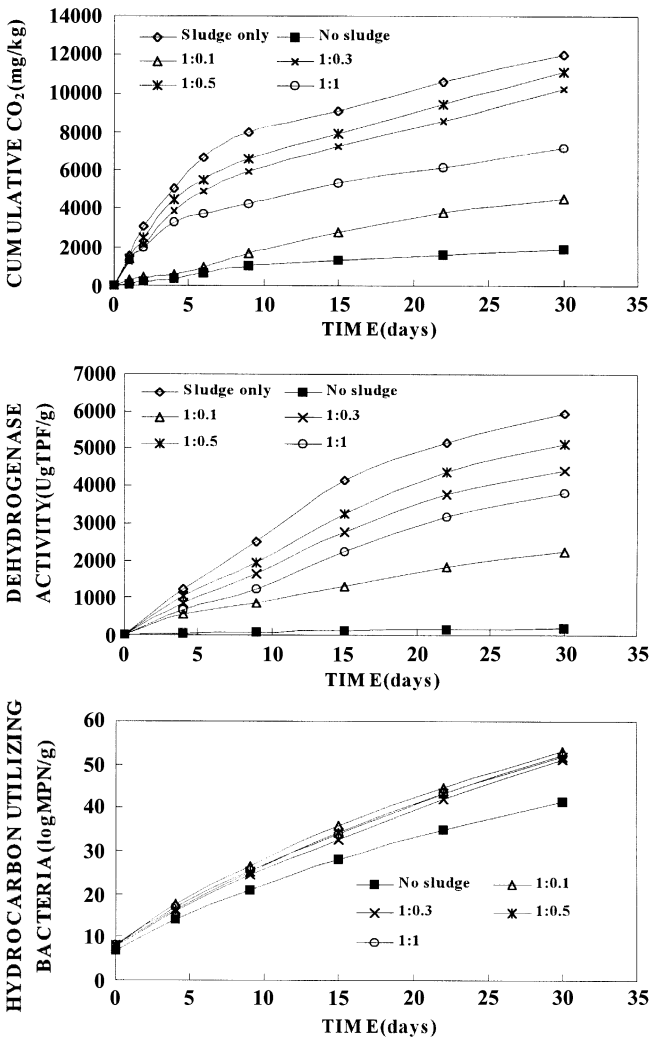

Fig. 5. Variation of cumulative CO_2 evolved, dehydrogenase activity, and the number of hydrocarbon utilizing bacteria in sewage sludge addition study.

3. Biochemical Parameters

Carbon dioxide generation rate showed a similar trend with TPH degradation. In other words, as TPH concentration decreased, the amount of CO_2 evolved increased. In general, CO_2 evolution rate has been used as an index representing microbial activity because it is a by-product from organic compound degradation. In sewage sludge addition, except 1 : 0.1 mix ratio experiment, CO_2 evolution increased dramatically until 4 days which was initial stage of composting due to active degradation of TPH at the same period. The cumulative amount of carbon dioxide evolved shows more distinctly the effect of depending on the addition of sewage sludge as presented in Fig. 5. Total amount of CO_2 evolved was 11,241 mg/kg and 11,271 mg/kg in the mix ratios of 1 : 0.3 and 1 : 0.5, respectively. Which degradation of TPH was the greatest among the sewage sludge addition experiments? In case of 1 : 1 mix ratio experiment, total amount of CO_2 evolved was 7,194 mg/kg and this value was lower than that of 1 : 0.3 and 1 : 0.5 mix ratio experiments. This result agreed with the difference of TPH degradation under the same conditions. Also this indicated microbial activity for degrading TPH in 1 : 1 mix ratio study was lower than that of 1 : 0.3 and 1 : 0.5 mix ratio experiments. In case of 1 : 0.1 mix ratio study, CO_2 evolution rate in the early stage of composting was not discriminated with control experiment without adding sewage sludge, which indicated degradation of TPH was more slow than that of other mix ratio studies. Although total CO_2 evolved in the 1 : 0.1 mix ratio study was the lowest among the sewage sludge addition study, it was about two times greater than that of no sludge addition. This means that addition of organic amendment including sewage sludge facilitated degradation of diesel oil. Considering TPH degradation and total amount of CO_2 evolved, it is desirable to mix sewage sludge in the ratios of 0.3 part or 0.5 part to 1 part contaminated soil.

Dehydrogenase activity was matched with cumulative CO_2 amount well as shown in Fig. 5. Dramatic increase of dehydrogenase activity was monitored until 9 days in the studies which mixed in the ratios of 1 : 0.3, 1 : 0.5 and 1 : 1 contaminated soil to sewage sludge. Thereafter, dehydrogenase activity decreased because of the exhaustion of TPH due to the degradation. In case of 1 : 0.1 mix ratio study, slight increase of dehydrogenase activity was observed after 9 days, but the pattern of increase was relatively less remarkable compared to other mix ratio studies. No sludge study had little change in dehydrogenase activity during the composting period. Dehydrogenase activity in 1 : 0.5 mix ratio study increased to about 1,280 μgTPF/g, which was the most notable increase. In other words, microbial activity for degradation TPH could be regarded as the higher among the sewage sludge addition study because dehydrogenase activity due to the degradation of organic matter in sewage sludge was measured and corrected by the mix ratio of contaminated soil to sewage sludge. This is the same pattern with cumulative amount of CO_2 evolved. Dehydrogenase activity measurement could represent TPH degradation and CO_2 evolution rate well. Dehydrogenase activity indicated addition of sewage sludge had rapidly increased microbial activity in soil/sludge mixtures. This means that organisms in sewage sludge are metabolically active and may contribute to degradation process of diesel oil. Martens et al. [1992] reported the activity of soil enzymes may be inhibited by addition of certain organic amendments of such sewage sludge since it contains signifi-

cant amount of metals. Initial number of hydrocarbon utilizing bacteria existed about 10^8 MPN/g in sewage sludge mix studies, while it existed about 10^7 MPN/g in control study. Fan and Tafuri [1994] suggested effective degradation of the target contaminants can occur if number of hydrocarbon bacteria is in the range of 10^8-10^6 CFU/g. In this research, the number of hydrocarbon utilizing bacteria met this condition. Irregular variation of the number of hydrocarbon utilizing bacteria was observed in all sewage sludge addition experiments, although slight increase was observed.

4. Biocide Control

Degradation of diesel oil did not nearly occur in the biocide controls, while significant degradation diesel oil was observed in the experiments without biocide. This indicated microbial activity for degrading diesel oil was severely suppressed by the addition of biocide. Removal of TPH in the biocide control was 8.3%, while 99.5% of TPH was removed in experiments without adding biocide at the same condition. In biocide controls, dehydrogenase activity increased slightly and small amount of CO_2 was also evolved. This indicated differentiation of biological and chemical/physical degradation was not feasible, although contribution of biodegradation on diesel oil reduction was relatively large. Incoming air and incomplete inactivation of microorganisms might have affected the increase of values of biochemical parameters such as CO_2 and dehydrogenase activity. However, it can be stated that removal of diesel oil was mainly attributed to the biodegradation judging from significant difference of diesel oil removal between biotic and abiotic experiments.

According to Smith [1987], saturated aliphatic hydrocarbons have a low activity in chemical oxidation. Margesin and Schinner [1997] reported that 16% to 23% of diesel oil added to five alpine soils was lost by abiotic processes. Abiotic losses in this research were significantly low compared to the result of Margesin and Schinner [1997]. This indicates composting can be a very effective treatment option for diesel-contaminated soil.

In the experiments without adding biocide, degradation of TPH was 98.6%, while volatilization loss was 1.4%. Volatilization loss was insignificant when compared to degradation, which indicated volatilization loss of diesel was not the main removal mechanism in composting diesel oil-contaminated soil. Because the moisture content was adjusted to 70% of the field capacity, leaching by free liquid also was not occurring Therefore, volatilization might be a main abiotic removal mechanism in these experimental conditions. Percent degradation of n-alkanes was relatively high compared to TPH.

In case of biocide control experiments, degradation contributed from about 69.3% of TPH removal. This was relatively large compared to volatilization loss, which indicated inactivation of microbial population by biocide was not completely occurring though significant suppression of microbial activity was observed. It was found from the fact that dehydrogenase activity slightly increased during composting period.

CONCLUSION

The degradation of diesel oil in contaminated soil was significantly enhanced by the addition of sewage sludge. But excess addition of sludge had not necessarily facilitated degradation of diesel oil. It was desirable to mix contaminated soil to sewage sludge in the ratios of 1 : 0.3 and 1 : 0.5 as wet weight bases judging from degradation of TPH (98.1%) and biochemical parameters. The most effective mix ratio, however, was 1 : 0.3 when considering the decrease of contaminated soil to be treated due to the addition of sludge.

Volatilization losses of diesel oil were in the range of 0.9 to 2.4%, which indicated that volatilization was not a significant removal mechanism in composting of diesel oil. Individual n-alkanes ranging from C10 to C16 were lost by volatilization. Especially, normal alkanes ranging from C10 to C14 were significantly volatilized to C15 to C16. Biochemical parameters such as carbon dioxide and dehydrogenase activity were desirable to monitor the degradation of diesel oil because these parameters were well matched with the diesel degradation.

Removal of TPH was 8.3% in biocide control, and biocide control experiment, degradation contributed to about 70% of TPH removal. This was a relatively large value compared to volatilization loss, which indicated inactivation of microbial population by biocide was not completely occurring though significant suppression of microbial activity was observed.

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