

Degradation of diesel oil in soil using a food waste composting process

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Abstract We investigated the simultaneous degradation of diesel oil in soil and the organic matter in food waste by composting in 8 l reactors. Using a 0.5 l/min air flow rate, and 0.5–1% diesel oil concentrations together with 20% food waste, high composting temperatures (above 60°C) were attained due to the efficient degradation of the food waste. Petroleum hydrocarbons were degraded by 80% after only 15 days composting in the presence of food waste. In a 28 l reactor scale-up experiment using 1% oil, 20% food waste and 79% soil, removal efficiencies of petroleum hydrocarbons and food waste after 15 days were 79% and 77%, respectively.

Keywords Composting · Diesel oil-contaminated soil · Food waste · Petroleum hydrocarbons

Introduction

Incineration and landfill treatments of food waste or sewage sludge are not economical, primarily because these materials have high water and organic matter contents. The popularity of composting as an alternative treatment for organic wastes from the food industry or sludge from wastewater treatment plants is increasing because the organic matter content in these waste types is easily degraded by microbes. Composting of wastes has several advantages, including the hygienic safety of the end products after passing through the thermophilic phase, the suitability of the process for converting noxious substances into harmless matter, and the usability of the end product compost in the field as a soil conditioner or fertilizer (Kirchmann and Ewnetu 1998).

In another restoration process, bioremediation or biodegradation techniques have been used to restore soil polluted by oil (Venkateswaran and Harayama 1995; Jonge et al. 1997; Palittapongarnpim et al. 1998; Li et al. 2000; Sharma and Pant 2000; Seklemova et al. 2001; Gallego et al. 2001; Boopathy 2003). These methods are divided into two broad categories: bioaugmentation and biostimulation. Bioaugmentation involves seeding polluted sites with specific bacteria with oil-degrading abilities, while biostimulation involves adding nutrients or other growth-enhancing substrates to polluted soils with the aim of

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stimulating growth of indigenous biodegrading microbes (Lee and Merlin 1999).

If the composting process involves microorganisms that metabolize hazardous compounds, such as petroleum hydrocarbons, it is possible that composting could be used as a means of bioremediation (Valo and Salkinoja-Salonen 1986; Beaudin et al. 1996).

Factors that stimulate microbial activity include the presence of nutrients such as nitrogen (N), phosphorus (P) and oxygen (O₂), the availability of electron acceptors such as nitrate and sulfate, and the physical/chemical interaction. Generally, but depending on the source materials, the optimum nutrient conditions in a composting medium are generally reported as having a C/N ratio of 22 and an N/P ratio of 2–5 (Nakasaka et al. 1992; Michel et al. 1993). O₂ consumption and CO₂ production are also indicators of hydrocarbon degradation by microbes in compost (Head and Swannell 1999; Yerushalmi et al. 2003).

Research on the restoration of oil-contaminated soil using the composting process is increasing (Hogan et al. 1988; Beaudin et al. 1996, 1999; Jorgensen et al. 2000; Namkoon et al. 2002; Van Gestel et al. 2003). The removal rate of oil observed in situ is generally very low owing to a lack of nutrients. The addition of 45% horse manure as an organic substrate is reported to improve treatment of mineral oil and grease in biopile composting (Beaudin et al. 1996; Beaudin et al. 1999). On a wet weight basis using soil contaminated with diesel oil mixed with biowaste (vegetable, fruit, garden waste) at a ratio of 1:10, degradation ability was found to be greatest at a ratio of 1:0.5 diesel oil-contaminated soil to organic amendments (sewage sludge, compost) (Van Gestel et al. 2003; Namkoong et al. 2002).

As soil contaminated with petroleum hydrocarbon has a high C/N ratio and low organic matter content, food wastes will supply the proper nutrients and a diverse population of microorganisms for degradation, thus offering the possibilities of simultaneously purifying oil-contaminated soil and biodegradation of food waste.

In this study, we investigated the process of purifying soil polluted by petroleum hydrocarbons by first testing whether the composting reaction properly progresses in the presence of food wastes

as organic sources. We then conducted a quantitative analysis of the biodegradation of petroleum hydrocarbons in treatment soils.

Materials and methods

Materials

Food wastes used in this study were obtained from a restaurant at the author's university (Seoul National University). These wastes contained 80% water and 95% organic matter (dry weight). Table 1 shows the water content (WC) and volatile organic matter content of the experimental materials. Microbial counts in the food waste ranged between 10⁷ and 10⁸ colony forming units (cfu)/g-wet basis (Joo et al. 2001).

In the first experiment, sawdust was used as a bulking agent. The soil used in this study was a common sandy soil (poor nutrient soil) obtained from a university play yard, and was sieved after drying using a 2 mm mesh sieve. We used a commercial diesel oil (LG Co., Ltd., Korea) for contaminating the soils.

Reactors

A common cylinder type composting reactor was used in this study. A small 8 l composting reactor (inner diameter 20 cm, height from the bottom to the cap 26 cm, working volume 6 l) was surrounded with insulators to minimize heat loss. To obtain optimum composting conditions, 4–6 reactors were used at the same time. In order to maintain aerobic conditions, air (not humidified) was supplied at 0.5 l/min through a compressor at

Table 1 The water and organic matter contents of the materials used in this study

	Sawdust (%)	Food waste ^a (%)	Soil (%)	Diesel oil (%)
Water content (%)	~8	80–81	12–14	0
Volatile solids (%) ^b	99.6	94–96	4–5	100

^a Food waste used in this study contained 32% grains, 51% vegetables, 15% meat, 15% fish and 2% others, with a C/N ratio of 11 (pH 3.5)

^b On dry weight basis

the bottom of the reactor. A 28 l reactor, with a basic structure similar to the small composting reactor, was used for a scale-up experiment.

Experimental procedure

Composting of the food waste/oil mixture using a bulking agent

To examine the influence of 2% oil concentration on the composting of food waste, 70% food waste was mixed with 30% (w/w) sawdust (bulking agent) and a WC of approximately 60% was maintained. Air flow rates were set at 0.5 l/min or 1 l/min under the same conditions. After that, the influence of concentration of diesel oil under same mixing ratio of food waste and sawdust was in detail examined. Concentrations of diesel oil were 0%, 0.5%, 1%, 3% and 6% (w/w—sawdust and food waste-wet basis).

Effect of food waste/diesel oil/soil mixing ratio

In this experiment, soil was used instead of sawdust as a bulking agent. Food waste was mixed at 0%, 2.5%, 5%, 7.5%, 10% and 20% (w/w) with soil, and no water supplementation was conducted. The concentration of diesel oil mixed with the food waste / soil mixture was fixed at 1% (w/w). At a mixing ratio of more than 25% food waste, composting did not proceed efficiently because the air supply was insufficient in the reactor as the mixture exhibited a mud-like consistency.

After the optimal mixing ratio of food waste and soil was determined, diesel oil concentrations were changed to 0.5%, 1%, 3%, 6% and 10% (w/w), and the temperature rise and degradation of petroleum hydrocarbon were examined. In these experiments, 0.15% (w/w) of a composting product, produced in our laboratory using rice bran and soybean curd residue (okara), was added to promote composting.

Scale-up experiment using a 28 l reactor

A 28 l reactor was used in a scale-up experiment using a mixing ratio of 20% food waste (wet basis), 1% diesel oil and 79% soil. Samples from the reactor were extracted every 2 days, and the

degradation of oil was measured by gas chromatography/flame ionization detector (GC/FID). The reduction of total mixture weight was monitored on a scale placed under the reactor.

Extraction and analysis of petroleum hydrocarbons

Although Soxhlet extraction is a popular method for extracting oil from soil, we used a shaking extraction in this study as it is more convenient and effective (Hwang et al. 2000). For the shaking extraction, 10 g of composting material was mixed with 5 g sodium sulfate anhydrate for dehydration. The samples were then mixed with 50 ml methylene chloride and shaken at 200 strokes per minute (spm) for 2 h. The methylene chloride layer was filtered with PTEF filter paper (pore size = 0.45 μm), and the filtrate was analyzed by GC/FID, on a HP-5 column. The injection temperature was 250°C and the detector temperature was 280°C. The oven temperature was programmed to increase from 40°C to 280°C at 12°C/min and then maintained at 280°C for 8 min. Nitrogen was used as the carrier gas and the make-up gas used was helium. A gas ratio of 1:10 (purge:total) was used. The standard curve was prepared by analyzing blanks containing 200 ppm, 50 ppm, 10 ppm and 2 ppm diesel range organics (DRO-diesel fuel 31064, Thames Restek UK Ltd., UK), which consists of C10, C12, C14, C16, C18, C20, C22, C24, C26 and C28 petroleum hydrocarbon components.

In commercial diesel oil, other compounds than the 10 petroleum hydrocarbons mentioned above were present. Some of these compounds were impossible to detect or quantify with the HP-5 column used in this study. We assumed, however, that the removal patterns of other petroleum compounds would be similar to the 10 hydrocarbons tested in this study, and so petroleum hydrocarbon concentrations were calculated on the basis of these alone.

Analysis of other factors

Samples were taken from three points in the reactors (upper, middle, bottom). WC was measured as the difference in weight before and

after drying at 105°C, and volatile solids (VS) were measured similarly after burning at 550°C. The WC and VS reduction ratios were calculated using these values. The temperature inside the reactor was automatically recorded by a temperature sensor. Carbon dioxide and oxygen concentrations in the exhaust were periodically measured by a Gas Data LMSx Multi-gas Analyzer (Gas Data Ltd., Whitely Coventry, UK).

Results

Composting of food waste and oil with sawdust as a bulking agent

Composting of food waste and 2% oil mixed with sawdust as a bulking agent was conducted for 13 days at air flow rates of 0.5 and 1 l/min. Figure 1A shows the temperature change during the composting process. The composting pattern of 2% oil at 0.5 l/min air flow rate was similar to the pattern seen for the control (0% oil) at air flow rates of 0.5 and 1 l/min, indicating that food waste degradation was not influenced by the presence of 2% oil at this air flow rate. However, at an air flow rate of 1 l/min, the temperature declined rapidly after 6 days, indicating that composting had diminished. On the basis of these results, 2% oil and an air flow rate 0.5 l/min were judged to be the most appropriate conditions for composting.

When oil concentrations were varied in the range 0–6% at an air flow rate of 0.5 l/min, the temperature rose above 60°C in the 0%, 0.5% and 1% diesel oil (Fig. 1B). At 3% and 6% oil, however, the temperature did not rise above 55°C.

Composting at different mixing ratios of food wastes

Oil concentration was fixed at 1% in variable ratio mixtures 0–20% food waste in soil, and composting was carried out without sawdust as a bulking agent. At less than 10% food waste, the temperature rise was relatively low (Fig. 2), due mainly to the low organic matter content present in the mixture. However, when the ratio of food wastes was increased to 20%, the temperature

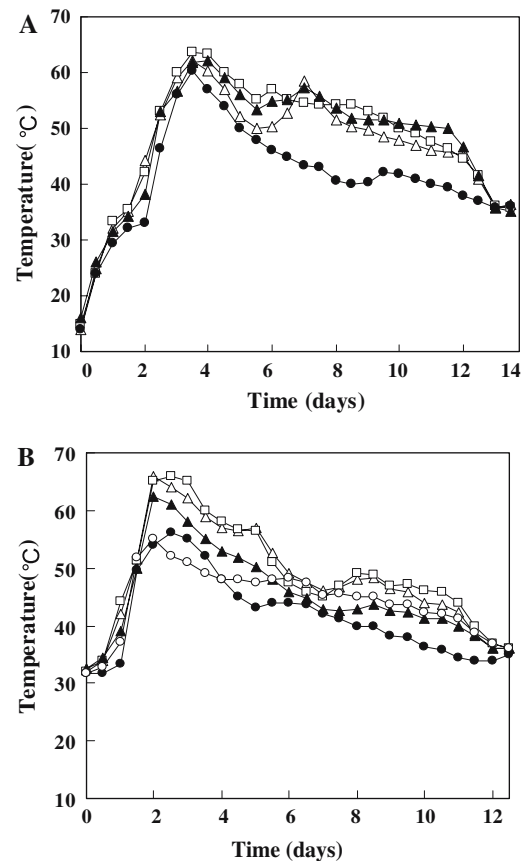


Fig. 1 Temperature changes during composting in 8 l reactors using (A) different air flow rates and in the presence of diesel oil and (B) different concentrations of oil with an air flow rate of 0.5 l/min. Oil was added to food waste with sawdust as a bulking agent and mixing ratio of food waste was the same. Symbols: A: (□) 0% oil (w/w), 0.5 l/min air flow rate, (△) 0% oil (w/w), 1 l/min air flow rate, (▲) 2% oil (w/w), 0.5 l/min air flow rate and (●) 2% oil (w/w), 1 l/min air flow rate, B: (□) control (0% oil, w/w), (△) 0.5% oil, (▲) 1% oil, (●) 3% oil and (○) 6% oil

reached approximately 55°C. At mixtures with greater than 25% food waste, the temperature rise was again relatively low, due mainly to the insufficient supply of air through the food waste mixture as it became wetter and mud-like in consistency (data not shown).

For each ratio of food waste tested, the oil components were measured by gas chromatography after composting for 10 days and the petroleum hydrocarbon elimination ratios were calculated (Fig. 3). Compared to a control (A), higher food waste content corresponded to lower

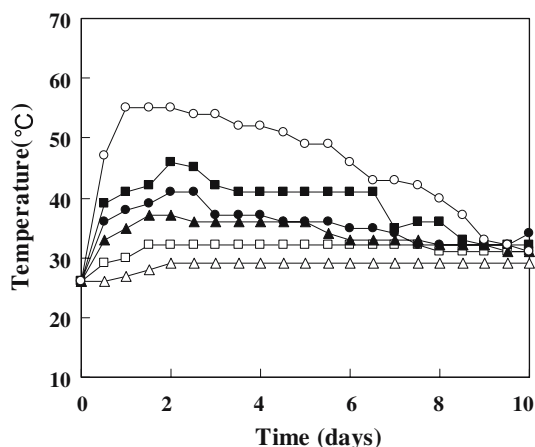


Fig. 2 Temperature change during composting with different ratios of food wastes and soil at a fixed concentration of 1% oil. Symbols: (\square) control (0% food waste, 99% soil), (\triangle) 2.5% food waste, 96.5% soil, (\blacktriangle) 5% food waste, 94% soil, (\bullet) 7.5% food waste, 91.5% oil, (\blacksquare) 10% food waste, 89% soil and (\circ) 20% food waste, 79% soil (w/w)

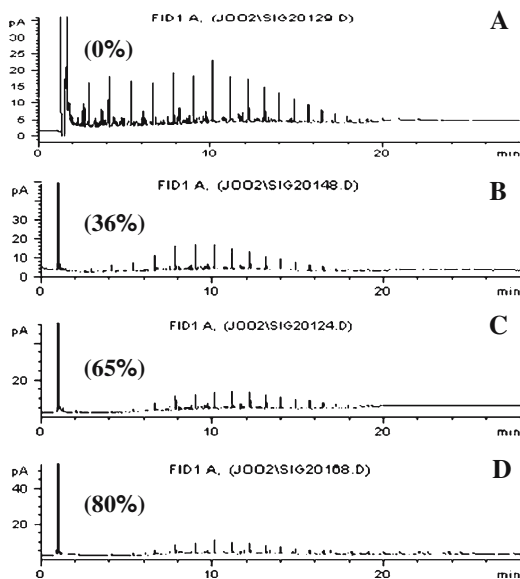


Fig. 3 GC chromatogram of remaining petroleum hydrocarbon components in soil after 10 days composting with different ratios of food wastes and soil as shown in Fig. 2: **A**, 0%; **B**, 5%; **C**, 10% and **D**, 20% of food waste (w/w). The retention times (min) of each component were as follows: C10 (4.1), C12 (6.6), C14 (9.0), C16 (11.1), C18 (13.1), C20 (14.8), C22 (16.4), C24 (17.9), C26 (19.3), C28 (20.9). The figures in parentheses are the removed percentages of petroleum hydrocarbons against 0% food waste (**A**)

peak heights. The peaks of long-chain petroleum hydrocarbons, particularly C14–C20 hydrocarbons, were relatively higher than those of short-chain hydrocarbons. The similar results were shown by Jonge et al. (1997). With an increase in food waste, the degradation of petroleum hydrocarbons increased, reaching 80% degradation of them in a 20% food waste mixing ratio.

To examine the effect of petroleum hydrocarbon volatilization by aeration, a control test was carried out by comparing the initial petroleum hydrocarbon concentrations (1% diesel oil) with samples composted for 10 days (**A** in Fig. 3). The concentration of petroleum hydrocarbons in the samples after 10 days was about 4% of the initial concentration (6% in C10–C16, 2% C18–C28) lower than that in the initial samples. In 3% diesel oil concentration, the sample after 10 days showed 3% lower than in the initial samples. It is probable that the decrease was caused by aeration for 10 days, or the biodegradation by indigenous microorganisms present in the original soils. However, we regarded this 3–4% loss minor because more than 80% removal of the initial oil was obtained in 20% food waste (**D** in Fig. 3).

Decomposition of different oil concentrations in soil at 20% food waste

In mixtures of 20% food waste (WC: 80%, volatile solid: 95% of dry weight) and 80% soil, oil was added to produce 0.5%, 1%, 3%, 6% and 10% oil concentrations (wet weight basis) and composted at an air flow rate of 0.5 l/min. As shown in Fig. 2, the maximum temperature among these treatments was approximately 55°C, which was not sufficient for efficient composting. Therefore, 0.15% (w/w) of the compost (microbial agent), produced in our laboratory using rice bran and soybean curd residue, was added as a seed to raise the activity of the composting reaction and consequently the decomposition of petroleum hydrocarbons. Temperature changes in the composting mixtures are shown in Fig. 4. A desirable composting environment was attained at concentrations of 0%, 0.5%, and 1% oil, in which maximum temperatures reached 65°C. Temperature increases were relatively low at concentrations of 3%, 6%, and 10% oil.

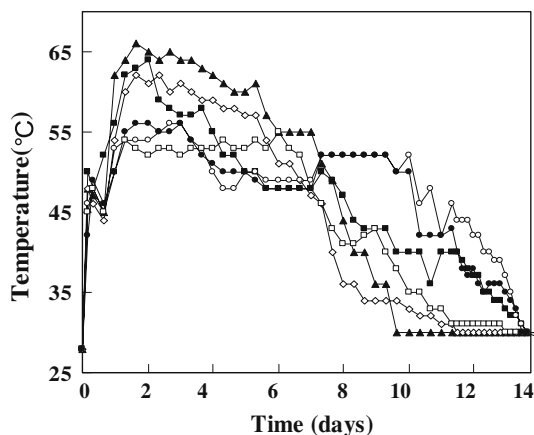


Fig. 4 Temperature changes in composting with 20% food waste and different oil concentrations: Symbols: (■) control (0% oil), (▲) 0.5% oil, (□) 1% oil, (○) 3% oil, (□) 6% oil and (●) 10% oil (w/w)

The changes in oil components, as measured by GC, at 0.5% and 1% oil concentrations in mixtures of food waste (20%) and soil (80%) at days 0 and 14 are shown in Table 2. Reduction efficiencies of C10–C28 components were 93% and 80% at 0.5% and 1% oil concentrations, respectively.

28 l reactor composting

A 28 l reactor was used in order to compare the decomposition of oil on a large scale with results

Table 2 Initial (day 0) and final (day 14) soil petroleum hydrocarbon concentrations at 0.5% and 1% diesel oil (w/w) concentrations (unit: mg/kg)

Hydrocarbons	0.5% diesel oil		1% diesel oil	
	0 day	14 day	0 day	14 day
C10	34	1	109	0
C12	44	3	127	16
C14	57	4	133	30
C16	72	5	132	31
C18	64	4	105	24
C20	37	3	71	21
C22	27	2	40	10
C24	17	0	20	5
C26	9	5	15	10
C28	3	0	33	7
Total	364	27	785	154
Total reduction efficiency of hydrocarbons	93%		80%	

obtained from the smaller reactors. The air flow rate in the 28 l reactor was fixed at 0.5 l/min, the same as that used in the 8 l reactors.

As shown in Fig. 5A, from 1.5 to 6 days, composting progressed actively and the temperature increased to 55–66°C. At the highest temperatures attained, the production of carbon dioxide was highest and oxygen content was close to 0%. Composting activity then decreased slowly until day 10. After day 10, no composting occurred as indicated by a fall in temperature to below 30°C, stabilization of the oxygen concentration, and a decrease in carbon dioxide concentration.

The changes in the sum of C10–C28 petroleum hydrocarbons over 15 days of composting are shown in Fig. 5A. During this period, petroleum hydrocarbons decreased by 79% from 865 to 176 ppm. The temperature after 10 days was below 30°C, but the degradation of petroleum hydrocarbons still progressed. Although maximum temperatures in the composting experiments were different, composting of 20% food waste mixed with 1% oil resulted in approximately 80% degradation of oil (Fig. 5A).

Figure 5B presents the change in weights of organic matter, volatile solids and WC in the 28 l reactor over the composting period. The weight of organic matter is the net weight of organic matter in the reactor. Reduction of organic matter weight was significant from 1.5 to 6 days, when composting microorganisms were most active. The difference in WC between initial and final composting stages was about 15%, indicating that water could not escape via evaporation due to the tightly sealed lid. Volatile solids decreased about 40% through composting. In this composting reaction, reduction weight was mainly due to the change in organic matter and WCs.

The component material weight changes over 15 days composting using the 28 l reactor are shown in Table 3. The total net reduction of weight (organic matter in food waste, oil and water) was 2,210 g (27,500–25,290 g). Water (WC) and volatile solid (VS) contents were reduced from 22% to 19% and from 9.4% to 6.2%, respectively. 80% of water (4,815 g) and 61% of organic matter (1,568 g) remained in the reactor. Of the total net weight reduction of 2,210, 1,208 g

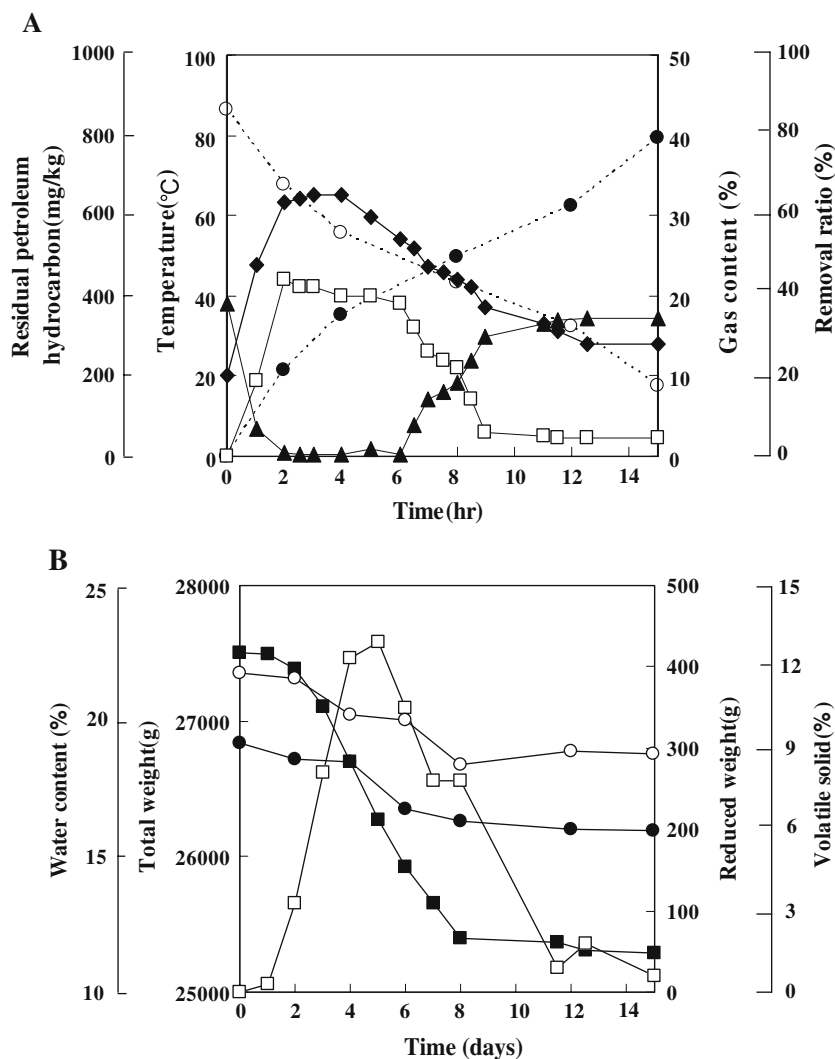


Fig. 5 (A) Changes in temperature (□), concentrations of carbon dioxide (□) and oxygen (▲) in the effluent gases, and changes in petroleum hydrocarbon concentration (○) and removal ratio of petroleum hydrocarbon (●) during

composting with 20% food waste, 79% soil and 1% diesel oil in a 28 l reactor. (B) changes in total reactor weight (■), reduced weight (□), weight of volatile solids (●) and water content (○) during composting in a 28 l reactor

of water was evaporated as aqueous vapor by oxidation heat. 2,582 g of initial total weight was sum of organics in soil and in two materials (food waste and diesel oil). 1,272 g of them was originally the existed organics in soil and they were not needed to degrade. 1,310 g was organics in food waste and diesel oil, and thus 1,014 g of them was degraded. Of the removed organic matter, 217 g of the initial diesel oil (275 g) and 797 g of the initial organic matter in the food waste ($5,500 \text{ g} \times 0.198 \text{ (solid)} \times 0.95 \text{ (VS)} = 1,035 \text{ g}$)

were degraded in the composting reaction. Reduction ratios of added diesel oil and food waste were 79% and 77%, respectively.

Discussion

When composting using 8 l reactors, the maximum temperature (and hence most efficient composting) was achieved at a mixing ratio of 20% food waste. At 25% food waste or more,

Table 3 Change in organic material contents over 15 days of decomposition of a mixture of diesel oil and food waste in a 28 l reactor

		Total	Food waste	Diesel oil
Initial	(g)	2,582	1,035	275
	(%)	100	100 (40)	100 (11)
Final	(g)	1,568 ^a	238	58
	(%)	61	23 (15)	21 (4)
Reduction	(g)	1,014	797	217
	(%)	39	77 (79)	79 (21)

All data were calculated by organics weights on the basis of removal ratio of diesel oil, water content, volatile solid and the measured weight in Fig. 5. Total is sum of organic weight of food waste, diesel oil and soil. Percentages in parentheses in food waste and diesel oil are ratios against total weights

^a sum of organics in food waste (238 g), in diesel oil (58 g) and in soil (1,272 g)

composting was not effective because excessive WC hindered aeration. When our composting stimulation product was added, temperatures rose by a further 10°C. Higher temperature rises in the composting process reflected more efficient degradation of organic matter, and the reduction ratio of diesel oil was dependent on the amount of food waste present in the mixture.

Temperature rises in 0.5% and 1% oil treatments were higher than in the control. Above 3% oil, however, temperatures did not rise as much. At less than 1% oil, sufficient removal of the petroleum hydrocarbon was confirmed. We assume that composting of more than 3% diesel oil is not effective due to the inhibition of oil to thermophilic microbes. To treat soil contaminated with more than 3% diesel oil, dilution of the surrounding soils will be required to reduce oil concentrations to degradable levels. However, most petroleum hydrocarbon biodegradation studies have used less than 3% oil in test media, and levels above this in the field are not common (Beaudin et al. 1999; Sharma and Pant 2000; Marquez-Rocha et al. 2001; Mohn et al. 2001; Seklemova et al. 2001; Namkoong et al. 2002; Thomassin-Lacroix et al. 2002; Juteau et al. 2003; Van Gestel et al. 2003).

When using a 28 l reactor for composting in a scale-up experiment, diesel oil reduction ratios were lower compared with those achieved in the

small reactor. This problem was due primarily to the accumulation of water vapor, and could be easily overcome through modification of the lid structure and the use of a vacuum pump.

Beaudin et al. (1999) achieved 68% degradation of mineral oil and grease (MOG, 17,000 ppm) using composting over 29 days. Van Gestel et al. (2003) observed an 80% degradation of diesel oil (16,000 mg/kg) in composting of bio-waste for 40 days, rising to 84% degradation after 85 days. In an on-site bioaugmentation experiment, Thomassin-Lacroix et al. (2002) biodegraded 70% of diesel fuel (2,400 mg/kg initial TPH concentration) over 65 days.

Our study had several advantages over previous studies. First, no bulking agent was used as soil acted as a bulking agent. Secondly, high diesel oil reduction ratios of 80% and 77% of food waste were observed in the relatively short period of 15 days. In this study, the efficient removal of diesel oil was obtained by decomposing oil and food waste simultaneously.

In conclusion, oil-contaminated soils should be treated on-site by biopile and windrow pile composting and subsequent excavation of the contaminated soil by a scraper (<http://www.frtr.gov>; Mohn et al. 2001; Thomassin-Lacroix et al. 2002; Juteau et al. 2003). If soil is mixed with food waste on-site, the removal rate of petroleum hydrocarbons will be considerably improved.

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