

Composting and Phytoremediation Treatment of Petroleum Sludge

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Abstract

Composting and phytoremediation using elephant grass (*Pennisetum purpureum*) were compared in experiments aimed at investigating the potential of both techniques in the treatment of petroleum sludge. The compost consisted of poultry manure and sawdust. N-P-K fertilizer was used for biostimulation of indigenous microbes. It also served to enhance the growth of the elephant grass. The sludge was mixed with agricultural soil and both techniques were then utilized for treatment. The total hydrocarbon content (THC) of the sludge-soil mixture before treatment was 64,494 mg/kg. After an 84-day treatment period, the composting treatment recorded 47% reduction in THC, the phytoremediation treatment showed 69% THC reduction, while the combination of composting and phytoremediation had 29% THC reduction. Microbial numbers corroborated the THC reduction observed. The results of the study show considerable promise for the deployment of elephant grass in phytoremediation treatment of petroleum sludge.

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Composting and phytoremediation using elephant grass (*Pennisetum purpureum*) were compared in experiments aimed at investigating the potential of both techniques in the treatment of petroleum sludge. The compost consisted of poultry manure and sawdust. N-P-K fertilizer was used for biostimulation of indigenous microbes. It also served to enhance the growth of the elephant grass. The sludge was mixed with agricultural soil and both techniques were then utilized for treatment. The total hydrocarbon content (THC) of the sludge-soil mixture before treatment was 64,494 mg/kg. After an 84-day treatment period, the composting treatment recorded 47% reduction in THC, the phytoremediation treatment showed 69% THC reduction, while the combination of composting and phytoremediation had 29% THC reduction. Microbial numbers corroborated the THC reduction observed. The results of the study show considerable promise for the deployment of elephant grass in phytoremediation treatment of petroleum sludge.

Keywords: Biostimulation; composting; elephant grass; oily sludge; phytoremediation; total hydrocarbon content.

INTRODUCTION

Petroleum processing usually results in the generation of large amount of oily sludge. Oily sludge is a potentially dangerous waste product consisting of a mixture of petroleum hydrocarbons and other complex compounds with very high molecular weight, which makes it highly recalcitrant to degrade. Clean-up technologies and disposal in secure landfills are expensive, and appropriate landfill sites are becoming very rare. Hence, concerted efforts have been made to investigate cheaper and environmentally benign technologies.

A number of studies have demonstrated the potential of biological methods in the attenuation of total petroleum hydrocarbons (TPH) in oily sludge. The effectiveness of the combination of Composting and Bioaugmentation (the use of bacterial inoculums to degrade hydrocarbons) in TPH reduction has been documented. Composting has been shown to effect about 50 – 75% hydrocarbon removal (Milne et al, 1998; Ouyang et al., 2005; Kriipsalu et al., 2007; Zhang et al., 2007; Fountoulakis et al., 2009). Bioaugmentation has been credited with about 90% TPH reduction (Lazar et al., 1999; Giles et al., 2001; Mishra et al., 2001; Vasudevan and Rajaram, 2001; Ayotamuno et al., 2007). A combination of both techniques was used in studies documented above. Degradation rates varied with the initial hydrocarbon concentration, compost content, bacterial consortium used, and duration of the experiments. Biostimulation of indigenous microbes with commercial fertilizers have also recorded positive effects though at relatively lower efficiency (Machin-Ramirez et al., 2008).

Although phytoremediation has gained wide acceptance for petroleum-contaminated soils, there is a paucity of literature on successful deployments of the technology. Phytoremediation entails the use of plants and their associated microorganisms for the treatment of contaminated soils and sediments (Alkorta and Garbisu 2001; Reichenauer and Germida 2008). Hutchinson et al. (2001) investigated the effect of inorganic fertilizer on the phytoremediation of aged petroleum sludge with bermuda grass (*Cynodon dactylon*) and tall fescue (*Festuca arundinacea* Schreb.), TPH degradation resulted to 62% and 68% after one year for tall fescue and bermuda grass respectively. Ouyang et al. (2005) reported that the planting of Tall Fescue further decreased the total hydrocarbon content (THC) of oily sludge by 5 - 7% after it had been treated with bioaugmentation and composting. A number of plant species, including perennial ryegrass (*Lolium perenne* L.), crested wheat grass (*Agropyron cristatum* L.), common millet (*Panicum miliaceum* L.), alfalfa (*Medicago sativa* L.) and maize (*Zea mays* L.), have been effective in phytoremediation of oily-sludge-contaminated soil (Muratova et al., 2008). Rye was found to accelerate clean up most effectively, degrading contaminant fractions in the oily sludge by 52%.

In the present study, compost consisting of poultry manure and sawdust was compared with phytoremediation using elephant grass (*Pennisetum purpureum*) for the treatment of petroleum sludge. Poultry manure has been shown to be an effective organic amendment in attenuation of total hydrocarbon content (THC) in soils (Adesodun and Mbagwu, 2008). It has also been used with sawdust in composting for treatment of oily sludge (Zhang et al., 2007). Elephant grass is a common tropical grass with advantageous characteristics such as rapid growth, large biomass, strong resistance, and

effective stabilization to soils. The perennial rhizomatous grass is able to use solar energy, water and nutrients more efficiently compared to other plants (Heaton et al., 2004). It has been deployed for phytoremediation of crude oil-contaminated soils in which biostimulation of indigenous microbes were achieved with inorganic nutrients (Ayotamuno et al., 2006; Kogbara, 2008; Ayotamuno et al., 2009). To our knowledge, elephant grass has not been used for phytoremediation treatment of oily sludge, which has more recalcitrant polyaromatic hydrocarbons (PAH) than crude oil-contaminated soil. The grass is ubiquitous in the tropics, whose high temperature and precipitation encourages its growth. Hence, it is likely to provide a cost-effective method in the treatment of petroleum sludge especially as field deployment of phytoremediation is relatively new in the tropics.

The objective of this study was to evaluate the potential of elephant grass in the treatment of oily sludge. The study also sought to compare its effectiveness to that of composting and a combination of composting and phytoremediation.

MATERIALS AND METHOD

The petroleum sludge was obtained from the site of one of the Multinational Oil Companies at Ejammah-Ebubu, Rivers State, Nigeria. The sludge resulted from a crude oil spill due to equipment failure in the late 1960s. As biodegradation of the hydrocarbon content was the main objective of this study, there was no quantitative analysis of the sludge to determine the amounts of heavy metals present. The physical state of the sludge had previously been altered from slurry to damp solid through mechanical dewatering.

Experimental design

Plastic containers of 0.05 m³ capacity and 0.35 m depth were used as reactor vessels. Three treatments and a control were used in the experiments. Each treatment had three replicate reactors. The reactors served to provide controlled conditions for nutrient concentration, watering, tilling, and most importantly to prevent excessive run-off of the hydrocarbon contaminant. These were located at the teaching and research farm of the Rivers State University of Science and Technology, Port Harcourt, Nigeria. They were maintained under a transparent roof, thus exposed to a very good light transmission and solar radiation. They were shielded from the rain. The ambient conditions during the study period include mean daily minimum and maximum temperature of 23°C and 31.5°C respectively, and a mean monthly relative humidity of 85%.

As a pre-treatment operation, agricultural soil obtained from the experimental location was added to the oily sludge. Mixing of the soil with the sludge provided a source of microbes and nutrients and served as a growth medium for the plants. The mix ratio used was sludge:soil = 2:1. Thus, each reactor contained 28 kg of oily sludge and 14 kg of soil. The mixture was thoroughly mixed and allowed to settle for three days so that microbial activity could ensue before treatment applications. Composting and phytoremediation treatment commenced after the three-day period. Selected properties of the raw oily sludge, the agricultural soil used for pre-treatment, and the components of the compost used (poultry manure and saw dust) are shown in Table 1.

The following is a description of the method used for each treatment.

Reactor O: Control

This reactor contained the sludge-soil mixture to which no treatment was applied.

Reactor A: Composting

Poultry manure and saw dust as bulking agent was composted in bins and allowed to cure for 28 days by which time the stink has subsided. Thereafter the compost was added to the soil-sludge mixture and mixed thoroughly. The sludge:soil:compost ratio was 4:2:1, implying that 7 kg of compost was added to each replicate of reactor A. The mixture was tilled five times a week. It also received 2 liters of water three times a week corresponding to the findings of Kogbara (2008) which showed the effectiveness of the levels utilized.

Reactor B: Phytoremediation

200 g of 20-10-10 NPK fertilizer was mixed with the sludge-soil mixture to facilitate plant growth. Thereafter, five stands of elephant grass were planted on the sludge-soil system. The same quantity of fertilizer was applied after three, six and nine weeks of treatment. Watering volumes were the same as in the composting treatment, but there was no tilling due to the presence of the plants.

Reactor C: Composting and Phytoremediation

Compost was added to the sludge-soil mixture at the same rate as the composting treatment, and the combinations were mixed. Thereafter, 200 g of 20-10-10 NPK fertilizer was added to the sludge-soil-compost mixture, and five stands of elephant

grass grown in the reactors. Watering and subsequent fertilizer application was the same as in the phytoremediation treatment.

Sampling

Samples were collected from the reactors at set sampling periods by auguring different random spots and bulking them together. Samples for THC measurements were placed in glass bottles and sealed with aluminium foil. The samples were immediately transferred to the laboratory for analysis.

Analytical methods

The THC was the main parameter used for comparing the treatments. It was determined using PRESTIGE-21 IR Spectrophotometer by measuring light absorbance at wavelengths of 3704 to 3333nm according to ASTM D 3921 (1996). Other parameters were determined using methods adapted from Page et al (1982) and APHA (1998) standards. Particle size distribution was carried out using the hydrometer method. pH was determined using an EIL model 7020 pH meter by dipping the electrode into a 1:5 soil:water suspension that has been stirred and allowed to equilibrate for about 1 hour. The oven drying method was used for moisture content determination. Total Organic carbon was determined by the Walkley-Black combustion method, while total nitrogen was determined by the Kjeldahl method. Bacterial counts were determined using plate count agar (Oxoid Ltd.).

Statistical significance of THC data was conducted using the multi sample median test. The objective was to test whether the three different treatments had the same median THC at 42 and 84 days.

RESULTS AND DISCUSSION

The sludge-soil mixture was weakly acidic due to the weak acidity of the oily sludge and soil used. The pH of both materials was approximately 5 (Tables 1 and 2). The pH interval of the sludge-soil mixture varied over time with the different treatments. The pH range in the control reactors (untreated oily sludge-soil mixture) during the study period was 4.70 – 5.48. While the pH ranges in the other reactors were 4.86 – 5.88 for the composting treatment, 5.70 – 6.50 for the phytoremediation treatment and 4.80 – 5.76 in the composting and phytoremediation treatment. Moreover, the pH intervals recorded in the reactors with phytoremediation treatment were in the 5.2 – 6.8 range, which is the recommended pH for growth of the grass in this study (USDA-NRCS 2009).

The oily sludge contained a low moisture content of approximately 5%. Mixing with soil did not cause any appreciable increase in the parameter (Tables 1 and 2). As a result, the moisture content of the control was approximately 5% throughout the duration of the experiment. However, the other reactors showed increased moisture contents due to watering. The moisture content ranged from 10 – 16% in the composting treatment, 12 – 18% in the phytoremediation treatment and 9 – 15% in the composting and phytoremediation treatment. It is interesting to note that the average moisture content in reactor B (phytoremediation treatment characterized by transpiration) at 42 and 84 days of treatment was about 2 to 3% higher than that in compost treated reactors, A and B (Tables 3 and 4). Composts have been known to improve moisture retention (Singer et al., 2006). Even so, elephant grass has minimal

water use compared to other plants; hence, it has high tolerance to drought and restricted water conditions.

Petroleum sludge usually has a very high carbon-nitrogen ratio. The sludge used in this study had a C:N ratio of 4730:1 (Table 1). This affects bacterial growth and the utilization of carbon sources. However, in the course of pre-treatment of the sludge with soil and subsequent treatment with the remediation techniques, the total nitrogen level in the treatment reactors increased due to the application of the nitrogenous fertilizer. Increment in nitrogen level and bacterial utilization of the hydrocarbons led to an enormous decline in the parameter over time in the treatment reactors - down to 112:1 in reactor B (phytoremediation option) at 84 days, while that of the untreated sludge-soil mixture was at 3100:1 (Table 4).

Mixing of the oily sludge with soil reduced the THC of the sludge from 98,032 mg/kg to 64,494 mg/kg; hence, percentage THC reduction is calculated with reference to the latter figure. Samples collected after 42 days of treatment showed that there was attenuation in THC in all reactors including the untreated soil-sludge mixture (5% reduction). The composting option (reactor A) had 38% reduction. The phytoremediation option (reactor B) had 52% while the combination of composting and phytoremediation (reactor C) had 22% THC reduction (Table 3). At this time, plants grown in reactors B and C had some traces of leaf burn and retarded growth indicating their response to toxic conditions in the hydrocarbon-rich sludge-soil mixture. However, after 84 days of treatment, the leaf burn had subsided coupled with improved growth of the plants, and the phytoremediation option recorded the highest THC attenuation (down to 20,101 mg/kg, that is 69%). On the other hand, the composting

option and the combination of composting and phytoremediation had 47% and 29% THC reduction respectively (Table 4). The THC attenuation in the control was 12%; this was due to mixing of the oily sludge with soil, which facilitated the introduction of hydrocarbon degrading bacteria resulting to small amount of hydrocarbon removal. The multi sample median test indicated that differences between THC values obtained in the different treatments at 42 and 84 days were statistically significant at the 0.05 level (Table 5).

These results demonstrate the potential of elephant grass to facilitate the attenuation of petroleum hydrocarbons in oily sludge. Previous studies have shown that the plant root zone has significantly larger numbers of microorganisms, than soils that do not have plants growing in them. This appears to enhance the biodegradation of organic pollutants. The increased microbial numbers are primarily due to the presence of plant exudates and sloughed tissue which serve as sources of energy, carbon, nitrogen, or growth factors (Lee and Banks, 1993; Banks et al., 2003). This was corroborated by the results of the total heterotrophic bacterial (THB) counts as microbial numbers increased in all reactors, but the phytoremediation treatment had the largest microbial population at both sampling times (see Tables 3 and 4). Furthermore, the THB counts followed the same trend as the THC reduction and microbial numbers were of the decreasing order: phytoremediation > composting > composting and phytoremediation > control. Thus, this trend gave evidence to microbial degradation of the hydrocarbons.

It is interesting to note that the combination of composting and phytoremediation was less effective than the use of either technique although plant growth was similar in reactor B (phytoremediation) and reactor C (composting and phytoremediation). The

mechanism responsible for this is poorly understood. It was expected that the combination of both techniques would be more effective in the attenuation of petroleum hydrocarbons. This is because composting and phytoremediation are associated with rich microbial diversity, hence the combined effect of both techniques was projected to yield a better result than the use of one technique. There is insufficient literature on the combined simultaneous use of both techniques in treatment of hydrocarbon-contaminated soils or sludge. Accelerated dissipation of polycyclic aromatic hydrocarbons was reported to occur in plant rhizosphere after the addition of compost (Mahro et al., 1994). Vouillamoz and Milke (2001) reported that compost helped in phytoremediation of diesel-contaminated soils, while Palmroth et al. (2002) noted that the addition of compost did not significantly enhance the removal of diesel fuel from contaminated soil because the appropriate types and amounts of hydrocarbon-degrading microbial populations were already present in the soil.

Furthermore, Kriipsalu et al. (2007) reported the possibility for organic amendments to release petroleum hydrocarbons to oily sludge during composting since their organic content is degraded along with target organic contaminants in the sludge. In this study, the THC of the organic amendments used in the compost was not determined hence their contribution to the final hydrocarbon content could not be accounted. These materials have relatively high TOC content ($30.74 \pm 2.06\%$ and $53.25 \pm 1.57\%$ for poultry manure and sawdust respectively) compared to the soil TOC content (0.34 ± 0.03). Additional carbon sources easier to be degraded than some petroleum hydrocarbons are used by microorganisms first. This preferential path is likely to occur in reactors A and C but not in B. Microbial transformation of easily degradable organic matter into water and carbon dioxide, resulting in relative higher concentration of

organics not easily degradable in the matrix may have led to lower contaminant attenuation in the compost treated reactors. A similar observation was reported by Kriipsalu et al. (2007).

In addition to the above, a number of factors may be responsible for the levels of contaminant reduction observed in the different treatments. It is obvious that since the reactors were exposed to solar radiation, certain amounts of contaminant loss are due to abiotic processes such as sorption and volatilization. However, the study sought to evaluate the potential of elephant grass in phytoremediation treatment and to compare the different treatments employed. Hence, the contribution of abiotic processes to contaminant loss was not determined. A possible superiority of the composting treatment over the combination of composting and phytoremediation might be the fact that shadows created by plants might reduce the contribution of abiotic processes to the removal of hydrocarbons from the soil-sludge mixture. Furthermore, when mixing oily sludge with soil and - in some treatments - with manure and sawdust, the heterogeneity of each solid material plus uneven mixing generate spots where PAH concentration is much higher than others. This could also have some effects on the results obtained. Overall, a combination of factors may be responsible for the lesser performance of the compost treated reactors compared to the phytoremediation treatment. It may also be due to the suitability and amount of the compost used for the plant in question. Further research is required in this direction.

CONCLUSION

This study has shown the potential of elephant grass to facilitate the attenuation of total hydrocarbon content of petroleum sludge. Its deployment in phytoremediation would provide a cost effective technology for the treatment of oily sludge. The study compared composting with poultry manure and sawdust to phytoremediation for treatment of petroleum sludge. Although, there are uncertainties associated with the outcome of the experiments, the available data indicates that phytoremediation had a better performance. The combination of both treatment techniques proved less effective compared to the individual techniques. Some likely reasons for the trend of contaminant reduction observed have been highlighted. Further investigations are required to improve our understanding of the exact mechanisms responsible. Further work may continue along the lines of combining bioaugmentation with phytoremediation or the subsequent use of phytoremediation after deployment of bioaugmentation in order to maximize the benefits of phytoremediation for treatment of petroleum sludge.

ABBREVIATIONS: C:N ratio-carbon:nitrogen ratio, CFU/ml-colony forming unit per millilitre, THC-total hydrocarbon content (mg/kg), THB-total heterotrophic bacteria, TOC-total organic carbon, Total N-total nitrogen.

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Table 1. Selected properties of base materials used in the experiments

Parameter	Raw Oily Sludge	Soil	Poultry manure	Sawdust
Sand (%)	-	11.3 ± 0.2	-	-
Silt (%)	-	41.6 ± 0.6	-	-
Clay (%)	-	47.1 ± 0.5	-	-
Texture	-	Silty clay	-	-
pH	5.40 ± 0.08	4.73 ± 0.20	8.14 ± 0.09	7.30 ± 0.20
Moisture (%)	5.2 ± 1.9	10.4 ± 1.2	48.2 ± 4.1	32.0 ± 3.8
TOC (%)	9.46 ± 0.02	0.34 ± 0.03	30.74 ± 2.06	53.25 ± 1.57
Total N (%)	0.002 ± 0.0004	0.110 ± 0.0060	2.560 ± 0.0500	0.130 ± 0.0400
C:N ratio	4730:1	3:1	12:1	410:1
THC (mg/kg)	98,032 ± 456	19 ± 1.5	Not analyzed	Not analyzed
THB (x10 ⁶ CFU/ml)	Not analyzed	5.8 ± 0.02	Not analyzed	Not analyzed

Results represent mean ± standard deviation of three replicates

Table 2. Selected properties of the Oily Sludge-Soil mixture 3 days after mixing before commencement of treatment applications

Parameter	Value
pH	5.50 ± 0.23
Moisture (%)	5.1 ± 1.20
TOC (%)	7.21 ± 0.04
Total N (%)	0.003 ± 0.0004
C:N ratio	2403:1
THC (mg/kg)	64,494 ± 456
THB (x10 ⁶ CFU/ml)	6.4 ± 0.06

Results represent mean ± standard deviation of the four reactors

Table 3. Selected properties at 42 days of treatment

Parameter	Reactor O	Reactor A	Reactor B	Reactor C
pH	4.90 ± 0.24	5.20 ± 0.30	5.80 ± 0.09	5.00 ± 0.20
Moisture (%)	5.2 ± 1.20	12.0 ± 1.8	14.3 ± 2.0	11.0 ± 2.0
TOC (%)	6.76 ± 0.03	4.52 ± 0.04	3.14 ± 0.07	5.49 ± 1.57
Total N (%)	0.002 ± 0.0009	0.008 ± 0.0004	0.009 ± 0.0010	0.011 ± 0.0400
C:N ratio	3380:1	565:1	349:1	499:1
THC (mg/kg)	60,957 ± 300	40,203 ± 250	31,204 ± 280	50,622 ± 150
THC reduction (%)	5	38	52	22
THB (x10 ⁶ CFU/ml)	10.71 ± 0.08	14.72 ± 0.12	21.99 ± 0.10	11.19 ± 0.05

Results represent mean ± standard deviation of three replicates

Table 4. Selected properties at 84 days of treatment

Parameter	Reactor O	Reactor A	Reactor B	Reactor C
pH	5.00 ± 0.50	5.80 ± 0.10	6.20 ± 0.30	5.40 ± 0.36
Moisture (%)	5.5 ± 2.0	14.0 ± 2.3	16.0 ± 1.8	13.8 ± 1.6
TOC (%)	6.20 ± 0.05	4.15 ± 0.03	2.23 ± 0.07	4.80 ± 0.02
Total N (%)	0.002 ± 0.0006	0.025 ± 0.0030	0.020 ± 0.0050	0.028 ± 0.0040
C:N ratio	3100:1	166:1	112:1	171:1
THC (mg/kg)	56,574 ± 180	34,364 ± 200	20,101 ± 150	45,775 ± 250
THC reduction (%)	12	47	69	29
THB (x10 ⁶ CFU/ml)	13.27 ± 0.06	23.64 ± 0.04	32.48 ± 0.03	21.23 ± 0.04

Results represent mean ± standard deviation of three replicates

Table 5. Multi sample median test for differences between THC values

Time (days)	Median THC (mg/kg)				Chi-Square	DF	Prob. > Chi-Square
	Comp.	Phytorem.	Comp. + Phytorem.	All			
42	40,205	31,212	50,638	40,205	6.3	2	0.044*
84	34,376	20,111	45,819	34,376	6.3	2	0.044*

Ho: all treatments have the same median Ha: at least two treatments have different medians DF: Degrees of Freedom

Comp.: Composting Phytorem.: Phytoremediation Comp. + Phytorem.: Composting and Phytoremediation

Prob.: Probability *Significant at $p < 0.05$