Effect of Oxygen Diffusion on Total Petroleum Hydrocarbon in Petroleum Contaminated Soils

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Abstract
Crude oil pollution on soil is a major environmental issue in the Niger Delta region of Nigeria due to oil exploration and exploitation. Treatment of crude oil contaminated soils is a major challenge to researchers in this region when its penetration level is beyond agricultural soil (30cm). This study aimed at examining the effect of oxygen diffusion through petroleum contaminated soils on Total Petroleum Hydrocarbon (TPH) degradation at depth of 100cm and above in the presence of indigenous microorganism. An experimental investigation was carried out on the impacted soils by polluting three different soils namely, Sandy soil, Sandy loam and Clay soil with crude oil. The objective was to simulate condition of major crude oil spill. Oxygen was diffused through each of the soils. Analysis of TPH at two weeks interval for six weeks using Gas Chromatograph-Flame Ionization Detector(GC-FID) revealed that bioremediation occurred faster in the soils at the depth of 100cm and above with oxygen diffusion through the impacted soils. The study shows that, Sandy soil recorded 71% reduction in TPH concentration in the soil. Sandy loam had 75% reduction in TPH concentration and Clay soil had 85% reduction in TPH concentration at the end of the study period of six weeks.

Keywords: *Diffusion; Bioremediation; Total Petroleum Hydrocarbon*

Introduction

Soil pollution as an environmental problem is necessary to be eliminated from our environment to enhance the basic function of soil for plants and animal usage and benefit. Soil is contaminated with different inorganic and organic materials such as agricultural pesticides, petroleum, and heavy metals which are remediated by the appropriate method to reduce pollution level. The contamination of soil by crude oil and petroleum products has become a serious problem that represents a global concern for the potential consequences on ecosystem and human health (Onwurah et al., 2007). The amount of hazards imposed on the natural environment depends on the surface of the area contaminated by the petroleum products, their chemical composition, and the depth at which pollutants occur (Wolicka et al., 2009). The technologies used for soil remediation includes, mechanical, burying, evaporation, dispersion, and washing. These technologies are very expensive and can lead to incomplete decomposition of hydrocarbon contaminants (Das and Chandra, 2010). A suitable method can be selected based on the volume of pollutants, type of soil, and environmental conditions. Different methods such as adsorption, precipitation, complexation, consumption by plants, and microbial removal can efficiently reduce pollutants in the soil (Khorasani and Yaghmaei, 2012). Crude oil bioremediation in soil can be promoted by stimulation of the indigenous microbial population, by introducing nutrients and oxygen into the soil (bio-stimulation) or through inoculation of an enriched microbial consortium into soil (bio-

augmentation) (Bento et al., 2005). In oil pollution, nutrients are rapidly assimilated by soil microorganisms thus depleting the nutrient reserves (Rahman et al., 2002). Therefore, apart from the environmental problem caused by oil pollution, the agronomic and economic aspects are significant (Kuhn et al., 1998). The addition of inorganic or organic nitrogen-rich nutrients (bio-stimulation) is an effective approach to enhance the bioremediation process (Margesin and Schinner, 2001). Positive results of nitrogen amendment by application of nitrogenous fertilizer on microbial activity and petroleum hydrocarbon degradation have been widely demonstrated (Akinde and Obire, 2008; Agarry and Jimoda, 2013). Bioremediation involves the use of microorganisms and their biodegradation ability to remove pollutants (Ofoegbu et al., 2015). The byproducts of effective bio-remediation, water and carbon dioxide, are non-toxic and can be accommodated without harm to the environment and living organisms. This method is cheap compared to physical methods for decontaminating the environment, which are extraordinarily expensive (Sims et al., 1990). The microbial degradation of petroleum in the environment is limited primarily by abiotic factors, including temperature, nutrients, and oxygen (Brown et al., 1992). The biodegradation of petroleum and other hydrocarbons in the environment is a complex process, who's quantitative and qualitative aspects depend on the nature and amount of the hydrocarbons present, the ambient and seasonal environmental conditions, and the composition of the indigenous microbial community. The microbial degradation of petroleum in aquatic and soil environments is limited primarily by nutrient, (nitrogen and phosphorus) and oxygen availability (Ayotamuno, et al., 2006). The initial steps in the biodegradation of hydrocarbons by bacteria and fungi involve the oxidation of the substrate by oxygenases for which molecular oxygen is required (Sims et al., 1993). Aerobic conditions are, therefore necessary in this process of microbial oxidation of hydrocarbons in the environment. The availability of oxygen in soil with hydrocarbon is often limited, and it depends on the type of soil. Biodegradation of petroleum hydrocarbons at rapid rates requires molecular oxygen (Ayotamuno, et al., 2006). Therefore, for effective petroleum biodegradation, it is necessary to ensure an available supply of oxygen. Microorganism growing in the soil will perform effectively in biodegradation of hydrocarbon compounds by tilling of the soil to facilitate penetration of oxygen into the soil (Ayotamuno, et al., 2006). This is applicable and effective in bioremediation of agricultural soil within 30cm depth, but would not take care of the contaminants at the higher depths because of the difficulty in providing sufficient oxygen. This study presents the use of oxygen diffusion to effect the rapid bioremediation of petroleum contaminated soils at higher depth of 100cm and above in Niger Delta soils. Degree of degradation of hydrocarbon is obtained by calculation, using the equation below

$$\% D = \frac{TPHi - TPHf}{TPHi} \times 100$$

Where D is degree of degradation, TPH_i is initial concentration of hydrocarbon and TPH_f is final concentration of hydrocarbon (Ofuegbu, et al, 2015)

Materials and Methods

Experimental Procedures

Soil Sampling

The Soil samples for this study were collected from Kolo creek in Bayelsa State, Oshie oil field, Ahoada west and Abua odua in Rivers state. It was collected from a depth of 20-30cm using a hand soil auger. The soil samples collected were bulked together and put in a well labelled Polyethylene bag and glass bottle, sealed with aluminum foil, especially for Total Petroleum Hydrocarbon (TPH) analysis in the Laboratory.

Oxygen Diffusion Rate Experiment

Fifty kilogram(50kg) of soil samples were weighed into nine bowel and contaminated with 5000mls (5litre) of Bonny light crude oil each , measured using 1000mls measuring cylinder five times by using standard pollution volume of 100mls of crude oil to 1kg of soil. This was done after collection of soil samples for determination of physiochemical properties. The objective was to simulate condition of major crude oil spill. Each of the mixture was properly mixed to ensure uniform concentration of the crude oil in the soil samples. It was left for three days to settle without any disturbance. The treatment of the soil commenced after three days by application and mixing of 340g of 27:13:13 NPK fertilizer with the soil samples B and C, while sample A had no fertilizer application because it served as control unit or sample for the process. Thereafter, the nine sample were transferred into nine separate Batch reactors labelled A, B, and C, of 1 meter depth and $4x10^{-2}m^3$ volume for each of the soil type, with application of 200mls of water every two day for 6 weeks to effect quick compartment and settlement of the soil samples. Soils in reactor C for the three type of soil had a PVC pipe which transferred oxygen gas from the gas cylinder into soils in reactor C. The PVC pipe had perforation of 4.2mm in diameter of four perforations per 10 cm intervals along the length of the Pipe. The purpose of the perforation was to allow the transfer of oxygen gas from the PVC pipe into the soil samples. A gas flow meter was fixed on the flow line into the reactors to measure the volume of oxygen transferred into the system. $14m^3$ of oxygen was delivered into soils in reactor C at an interval of 10minutes for 48hours and left to diffuse into the soil for a period of 42 days. Soil samples were collected from the reactors at intervals of 2, 4 and 6 weeks for Total Petroleum Hydrocarbon (TPH) and hydrocarbon utilizing bacteria analysis. This was carried out in three different soil samples, namely, Sandy Soil, Sand loamy soil and Clay soil. Total Petroleum Hydrocarbon of the soil was determined by using Gas Chromatograph-Flame Ionization Detector (GC-FID) Model, HP 5890 Series II, U.S.A., after extraction of hydrocarbon content (Risdon, et al. 2008). Standard deviation (SD), using the STDEV function in Microsoft® Excel 2013, simple percentages and ANOVA were used to analyze the data.

Results and Discussion

Table 1: Total Petroleum Hydrocarbon (TPH) concentration and percentage degradation for Sandy soil

REACTOR	Two Weeks (PPM)	Percentage Degradation (%)	Four weeks (PPM)	Percentage Degradation (%)	Six Weeks (PPM)	Percentage Degradation (%)
A	1124.89	1	1089.42	4.2	1056.74	7
B	976.43	14	679.49	40	536.34	53
C	930.34	18	416.75	63	325.00	71

× TPH concentration three day after contamination with crude oil =1136.79 PPM

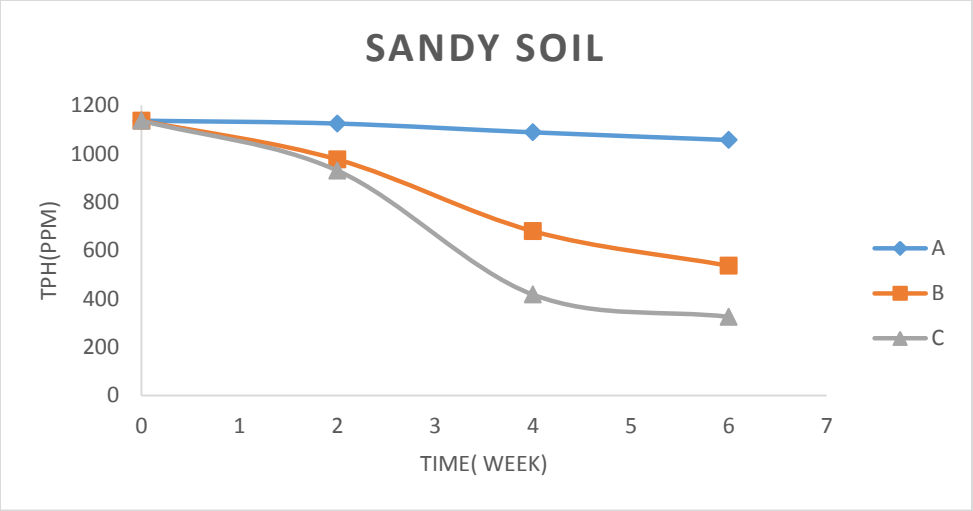

Figure 1: Graph of TPH concentration against Time for sandy soil

Table 2: Total Petroleum Hydrocarbon (TPH) concentration and percentage degradation for Sandy loam soil

REACTOR	Two weeks (PPM)	Percentage Degradation (%)	Four weeks (PPM)	Percentage Degradation (%)	Six weeks (PPM)	Percentage Degradation (%)
A	1367.45	0.4	1249.56	9	1134.74	17.4
B	1098.83	20	889.52	35	619.73	55
C	985.38	28	558.15	59	348.58	75

× TPH concentration three day after contamination with crude oil =1373.59 PPM

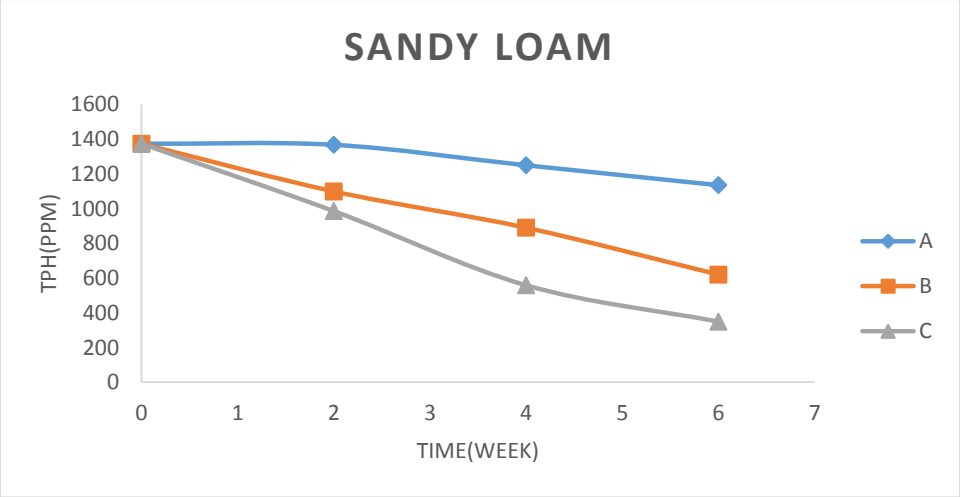

Figure 2: Graph of TPH concentration against Time for sandy loam soil

Table 3: Total Petroleum Hydrocarbon (TPH) concentration and percentage degradation for Clay soil

REACTOR	2 (Weeks)	Percentage Degradation	4 (weeks)	Percentage Degradation	6 (weeks)	Percentage Degradation
	(PPM)	(%)	(PPM)	(%)	(PPM)	(%)
A	1438.45	0.6	1428.92	1.2	1415.96	2.1
B	1227.72	15	642.72	56	488.20	66
C	1135.34	22	747.84	48	219.20	85

× TPH concentration three day after contamination with crude oil =1446.75 PPM

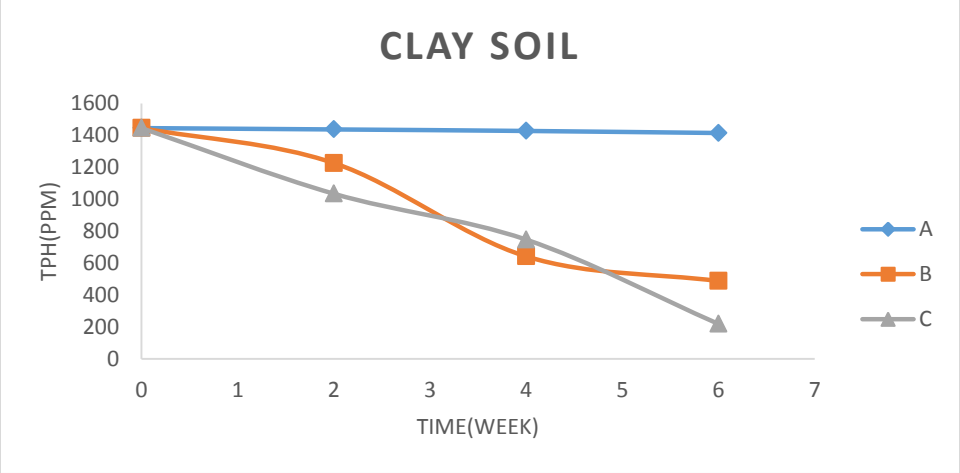

Figure 3: Graph of TPH concentration against Time for clay soil

Figure 1 shows that, in sandy soil, Total Petroleum Hydrocarbon (TPH) concentration decreased from 1136.79ppm after contamination to 1056.74ppm for soil in reactor A, Soils in reactor B reduced from 1136.79 ppm – 536.3ppm, while the soil in reactor C followed the same trend by decreasing from 1136.79ppm – 325.00ppm within the study period of six weeks.

Figure 2 indicates that, Total Petroleum Hydrocarbon (TPH) concentration for sandy loam soil in reactor A reduces from 1373.59ppm – 1134.74ppm. The TPH values for soil in reactor B, reduced from 1373.59 – 619.73ppm and, the TPH values for soil in reactor C, reduced from 1373.59 – 348.58ppm. Figure 3 shows, the behavior of Total Petroleum Hydrocarbon (TPH) in clay soils in reactor A, B and C. Total Petroleum Hydrocarbon in soils in reactor A reduced from 1446.75ppm – 1415.96 ppm, while TPH concentration in soil in reactor B reduced from 1446.75ppm – 488.20ppm. Soils in reactor C recorded TPH reduction in concentration from 1446.75 – 219.20ppm. Table 1- 3 indicated percentage reduction in hydrocarbon as follows. Sandy soils in reactor A recorded TPH reduction of 7%. Soils in reactor B, 53%, and Soils in reactor C registered 71%. Sandy loam soil samples in reactor A, B, and C had a TPH percentage reduction of 17.4%, 55% and 75% respectively. Also, the clay soils in reactor A, decreased in TPH by 2.1%, those in reactor B recorded 66% and the soils in reactor C had 85%. The reduction in concentration of Total Petroleum Hydrocarbon in the sandy soil, sandy loam and clay soil indicates that, the indigenous bacterial in the hydrocarbon impacted soil have the potentials to degrade petroleum hydrocarbon since they could use it as source of carbon and energy (Obiakalaije et al, 2015). The ability in reduction of hydrocarbon in crude oil contaminated soil at the depth of 100cm and above

may be attributed to higher growth in population of micro -organism because of sufficient Oxygen concentration in the impacted soil due to Oxygen diffusion through the crude oil contaminated soils. Statistically, the effect of time was highly significant at $P<0.05$ for soils in reactor C for sandy, sandy loam and clay soil.

Conclusion

From the above results obtained from the three soils considered in this work, it is obvious to conclude that, Oxygen diffusion through petroleum contaminated soils at depth beyond Agricultural soil have the ability of increasing indigenous microbial population of Hydrocarbon Utilizing Bacteria (HUB) in the crude oil impacted soils of Niger Delta, which uses the carbon source for energy, thereby reducing the concentration of Total Petroleum Hydrocarbon(TPH) in the impacted soils at depth of 100cm and above.

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