

Effects of Oxygen Diffusion on Physicochemical Characteristics of Petroleum Contaminated Sandy Loam Soil

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ABSTRACT: Soil pollution by petroleum and its products in Niger Delta region of Nigeria are predominant due to oil exploration, exploitation and transportation. The aim of this study was to examine the effects of oxygen diffusion on the physicochemical characteristics of petroleum contaminated sandy loam soils at 100cm depth when there is sufficient availability of indigenous microorganism in the polluted soils. The soil samples were analysis after pollution with Bonny light crude oil. The purpose of the pollution of the soil samples were to simulate conditions of major crude oil spill. Oxygen was diffused through the impacted soils in reactor C. Physicochemical properties of the sandy loam soils considered for analysis were pH, Electrical conductivity, Total organic carbon, Total Nitrogen, Moisture content, Phosphorous and Total petroleum hydrocarbon at two weeks' interval for six weeks using standard methods. The analysis indicated that bioremediation was faster in the soils at the depth of 100cm with oxygen diffusion. The study showed 75% reduction in TPH concentration, pH decreased from 6.47 to 6.34, Electrical conductivity increased from 92.1 to 169.9 μs , Moisture content decreased from 13 to 8%, Total organic carbon reduced from 2.301 to 1.716%, Total Nitrogen decreased from 0.199 to 0.032% and Phosphorous reduced from 0.08 to 0.003 mg/kg at the end of the study. Physicochemical properties of the treated soils showed similar soil characteristics as compared with the initial conditions of the soil before contamination with Bonny light crude oil. Therefore, this confirmed that oxygen diffusion through petroleum contaminated sandy loam soil was an effective method and a good approach for treatment of petroleum contaminated Sandy loam soil at the depth of 100cm and below.

Keywords: Hydrocarbon; Oxygen diffusion; Pollution; Sandy loam soils.

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I. INTRODUCTION

Bioremediation is a process that utilize microorganism or biological approach to transform different contaminants into harmless substance (Vidali, 2001). Bioremediation uses three basic methods such as natural attenuation, bio-stimulation and bio-augmentation (Chikere et al., 2009). For an efficient process of bioremediation to take place in the soil, the soil must have adequate concentration of nutrient and oxygen, to promote the actions of microorganism (Umeda et al., 2017). The microbes in soil contaminated with crude oil utilized more nutrients and this leads to a very fast depletion of the soil nutrients (Rahman et al., 2002).

Biodegradation of soil contamination with crude oil depends on environmental situation of the soil and on the chemical composition of the contaminant (Aldrett et al., 1997). The rates at which biodegradation occurs depends on the type of hydrocarbon present at the impacted site (Del Arco and de Franca, 2001). Compound that have lower grade of molecular weight like the aliphatic hydrocarbons degrades faster compared to those with heavier molecules (aromatic hydrocarbon). The species of microbes that cannot degrade the hydrocarbons with heavier molecules disappeared after the lighter hydrocarbons are degraded. (Ezra et al., 2000). The degree and rate of biodegradation are influenced by the type of soil in which the process occurs (Van hmm et al., 2003).

II. MATERIALS AND METHODS

2.1 SOIL SAMPLING

The Soil samples for this study were collected from Ewue Two Oil Field in Abua Odua Local Government Area of Rivers state. It was collected using hand soil auger. This location was considered because of frequent crude oil spill occurrence due to high rate of illegal refining activities. The soil samples collected were bulked together and put in a well labelled Polyethylene bags and glass bottles, sealed with aluminum foil, for Total Petroleum Hydrocarbon (TPH) analysis.

2.2. EXPERIMENTAL DESIGN AND PROCEDURE

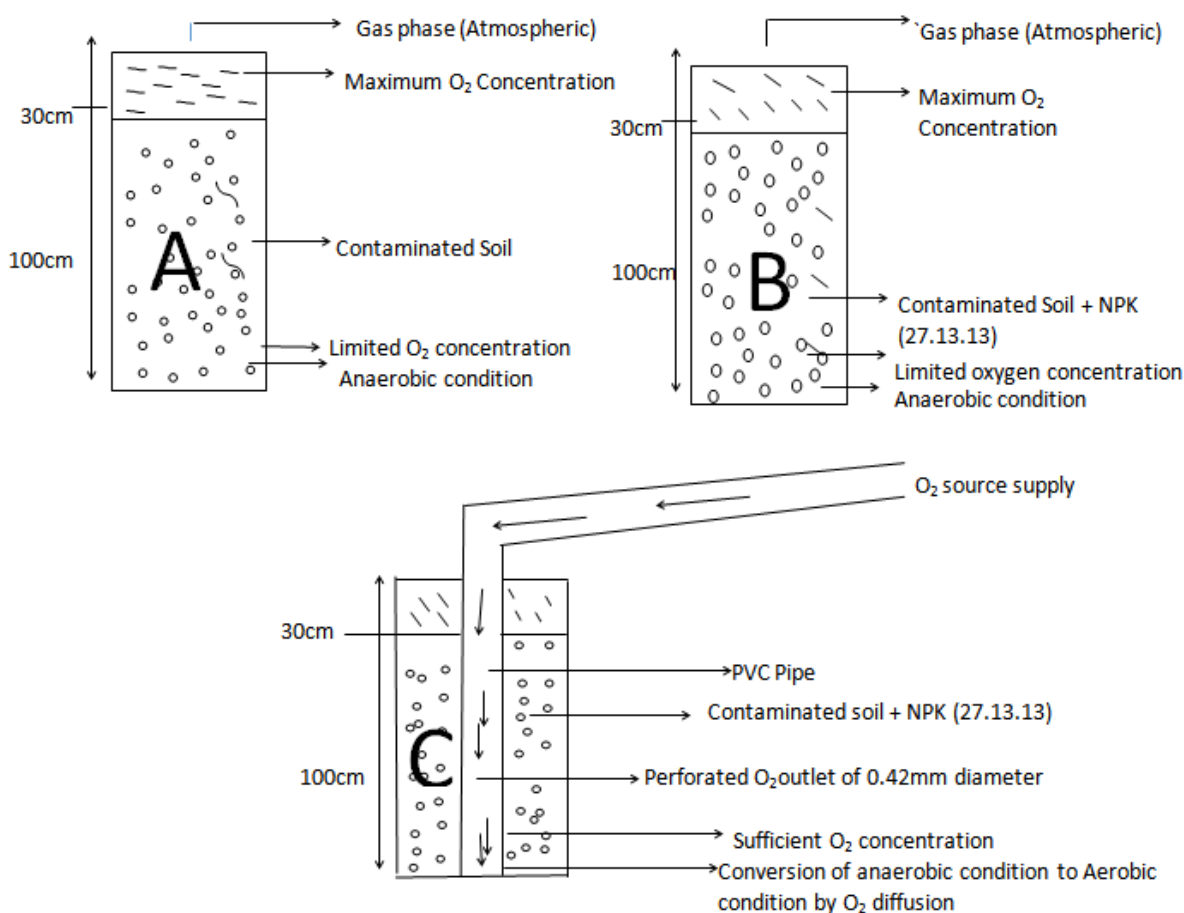


Figure 1. Schematic Diagram for Experimental Set up of Oxygen Diffusion in the Reactors.

Fifty kilogram (50kg) of soil samples were weighed into three 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 physicochemical 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. The three treated sandy loam soil samples were transferred into three separate batch reactors labelled A, B, and C, of one - meter depth and $4 \times 10^{-2} \text{m}^3$ volume, with application of 200mls of water every two days for 6 weeks to effect quick compartment and settlement of the soil samples. Soils in reactor C had a PVC pipe which transferred oxygen gas from the gas cylinder into soils in reactor C. The PVC pipe had perforation of 0.42mm in diameter of four perforations per 10 cm intervals along the length of the Pipe. The purpose of the perforations 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. (Umeda *et al.*, 2017). Soil samples were collected from the reactors at intervals of 2, 4 and 6 weeks for physicochemical properties and hydrocarbon utilizing bacteria analysis. Standard deviation (SD), using the STDEV function in Microsoft® Excel 2013, simple percentages and ANOVA were used to analyze the data.

2.3. DETERMINATION OF PHYSICOCHEMICAL PROPERTIES

The soil samples were characterized for particle size distribution and classification, pH, electrical conductivity, total petroleum hydrocarbon, moisture content, total organic content, total nitrogen, and phosphorous physicochemical properties were determined before and after contamination with crude oil using standard methods adopted from relevant literatures. Soil type classification and particle size analysis was carried out before contamination of the soil with crude oil by hydrometer method using sodium hexametaphosphate as

the dispersing agent (Ayotamuno et al., 2011). The soil structural classification was obtained, using the United State Department of Agriculture (USDA, 1987) soil textural classification scheme using TAL[®] for Windows software. The pH level of the soil samples were determined in the laboratory using Hanna HI 2211 pH/ORP meter according to ASTM (1999) method D4972. The pH was determined by dipping the electrode into a 1:25 soil: water suspension that was stirred and allowed to equilibrate for one hour. Electrical conductivity of the soil samples were measured using, Hanna EC 214 conductivity meter. Total organic carbon was determined using wet combustion method as reported by Ayotamuno *et al.* (2006). Soil Moisture Content was obtained by application of APHA (1998) standard as described in Ayotamuno *et al.* (2011). Total Petroleum Hydrocarbon was analyzed by using Gas Chromatograph-Flame Ionization Detector (GC-FID) Model, HP 5890 Series II, U.S.A., after extraction of hydrocarbon content by applying ASTM (1999) method D3921 (Umeda, et al., 2017). Total nitrogen was determined by using APHA (1998) method, 4500-NO₃ B. Phosphorous was analyzed by using APHA (1998) method, 4500-PO₄³⁻

III. RESULTS AND DISCUSSION

Table 1 shows the initial conditions and particle size distribution analysis of the soil, as sand 78.4%, Silt 6.8% and Clay 14.7%. This indicated that the soil was Sandy loam, with bulk density of 1.201g/cm³ and porosity 0.547.

Table 1. Physicochemical characteristics of Sandy loam soil before pollution (Result represent mean ± standard deviation of three replicates)

| Sand (%) | Silt (%) | Clay (%) | MC (%) | pH | EC (µs) | TPH (PPM) | TOC (%) | TN (%) | P (mg/kg) |
|----------|----------|----------|--------|-----------|----------|-----------|---------|---------|-----------|
| 78.44 | 6.80 | 14.76 | 32±2 | 6.32±0.06 | 87.2±0.8 | 9.60±0.1 | 1.014±1 | 0.088±1 | 0.10±1 |

Bulk density = 1.201g/cm³, Porosity = 0.547

Table 2. *Physicochemical characteristics of Sandy loam soil 3days after pollution, prior to remediation (Result represent mean ± standard deviation of three replicates)*

| pH | EC (µs) | TPH (PPM) | MC (%) | TOC (%) | TN (%) | P (mg/kg) | Oxygen Conc.(mg/l) |
|------------|---------|---------------|----------|-------------|----------|-----------|--------------------|
| 6.47 ±0.49 | 92.1 ±2 | 1373.59 ±0.62 | 13 ±0.87 | 2.301 ±0.20 | 0.199 ±1 | 0.08 ±001 | |
| A | | | | | | | 0.95± 1 |
| B | | | | | | | 1.01±0.02 |
| C | | | | | | | 2.50±4 |

Table 2 indicates the physicochemical characteristics of Sandy loam soil 3days after contamination with crude oil. Comparison of Table 1 and Table 2 showed that the concentration of the physicochemical parameters of sandy loam soil increased after contamination with crude oil, except, moisture content which decreased. The change in concentration of the parameters was as a result of the contamination of the soils with crude oil which introduced contaminants into the soils.

3.1 Variation of pH with Time

Figure 2 shows the variation of pH with time for Sandy loam soil in reactor A, B and C. In Figure 2, the pH values in the soils in reactor A, B and C decreased after pollution with crude oil. These were as a result of the effect of pollution of the soil with crude oil which introduced pollutants into the soils. Figures 2 showed a continuous decrease in pH value in soils in reactor A and B with Time while soils in reactor C showed an increase in pH value with Time for the entire period of bioremediation.

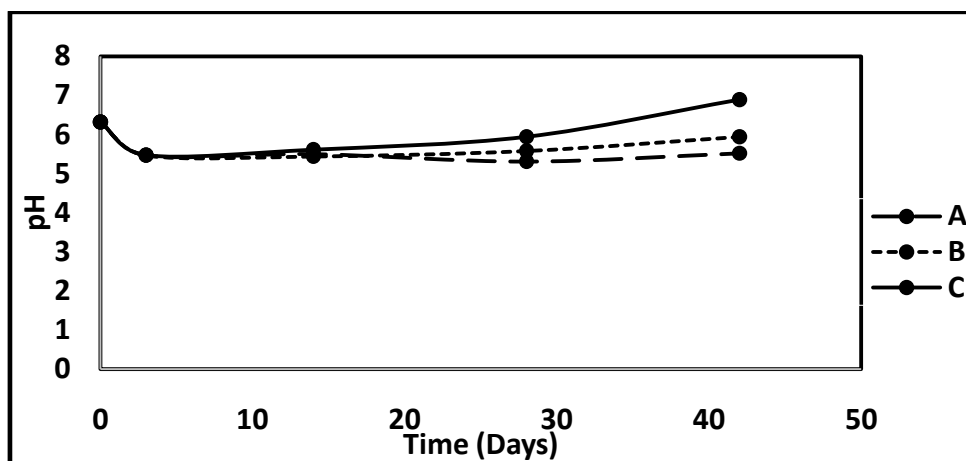


Figure 2: Variation of pH with Time

The increase in acidity level in soils in reactor A and B were as a result of the release of metabolites by microorganism due to insufficient oxygen to enhance bioremediation by aerobic conditions at 100cm depth in Sandy loam soils. The decrease in acidity level in soils in reactor C were due to the process of complete aerobic conditions experienced by the microorganism as explained by Ekperusi&Aigbdion (2015). This may be possible due to provision of oxygen at 100cm depth through oxygen diffusion.

3.2 Variation of Electrical conductivity with time

Figures 3 show the variation of Electrical Conductivity with Time in soil in reactors A, B, and C. The graph shows the initial level of Electrical Conductivity (EC) in sandy loam soil rised after pollution with Crude oil. This is due to the presence of trace ionic elements or metals released into the soils by the crude oil pollution which initiated a change in the initial level of the Electrical Conductivity of the soil (Obiri&Nwanbete, 2001). Figures 3 showed a decrease in amount of Electrical Conductivity in soils in reactor A and B within the second week, but thereafter, slight increase was noticed for the entire period of six weeks of the bioremediation process. The soils in reactor C in Figure 3 was constant within the second week, but thereafter, Electrical Conductivity level decreased rapidly towards the sixth week of bioremediation. Electrical Conductivity (EC) is the measure of the salinity of the soil.

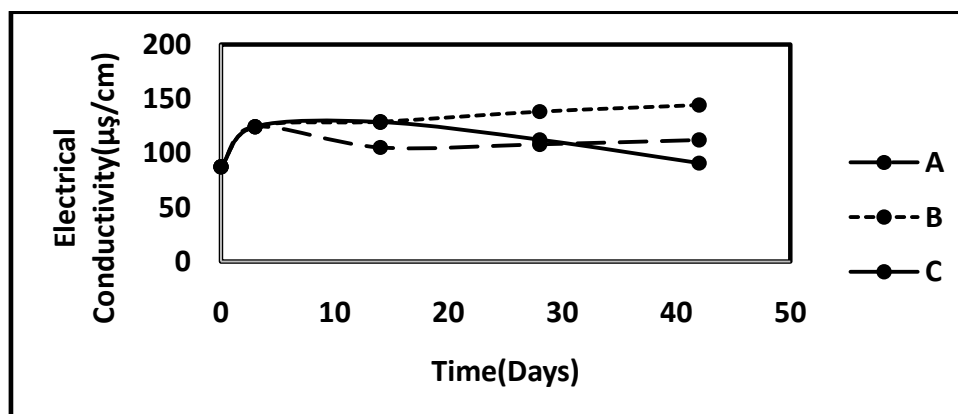


Figure 3: Variation of Electrical conductivity with time

The applied Fertilizer (NPK 27:13:13) is a salt, so the treated options (soils in reactors B and C) are expected to have higher level of Electrical Conductivity due to the application of NPK 27:13:13 as nutrients. (Ayotamuno *et al.*, 2009). The decrease in amount of Electrical Conductivity in soils in reactor C may be as a result of the utilization of the nutrients by the microorganisms during biodegradation of hydrocarbons. This may be as a result of sufficient oxygen provided to the microbes at 100cm depth due to oxygen diffusion which gave rise to an increase in microbial population.

3.3 Variation of Total Petroleum Hydrocarbon with time

Figures 4 shows the variation of Total Petroleum Hydrocarbon with Time in soils in reactors A, B and C. It indicated that the initial concentration of Total Petroleum Hydrocarbon (TPH) in the soil increased after pollution with Crude oil. This is because of the introduction of carbon compounds into the soils because of crude oil pollution (Obiakalajeet *et al.*, 2015). Also, the graphs revealed that, there were decrease in concentration of hydrocarbons in the soil samples with Time after three days of contamination of the soil with crude oil for the entire period of bioremediation. Also Figure 4a- 4c showed the chromatogram of petroleum contaminated Sandy loam in reactorC for with respect to Time respectively.

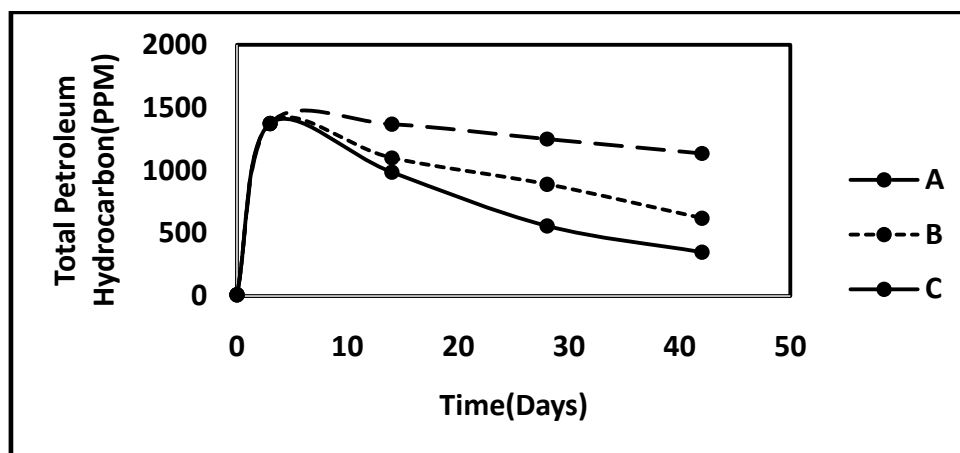


Figure 4: Variation of Total Petroleum Hydrocarbon with time

They indicated that the concentrations of the carbon atoms decreased with time. The peak of each of the carbon components also decreased with Time. The decrease in concentration of Total Petroleum Hydrocarbon in the soils showed that, the indigenous bacterial in the impacted soils have the ability to degrade petroleum hydrocarbon since they could use it as source of carbon and energy (Obiakalajeet *et al.*, 2015). The reductions in percentage of hydrocarbon concentration in Sandy loam soils were obtained as 17%, 55% and 75% for soils in reactor A, B and C respectively. The highest percentage degradation of Total Petroleum Hydrocarbon noticed in soils in reactor C may be attributed to an increase in population of microorganism because of availability of sufficient oxygen concentration in the soils at 100cm depth as a result of oxygen diffusion, therebyutilizingthe hydrocarbons as source of energy by the Microorganism.

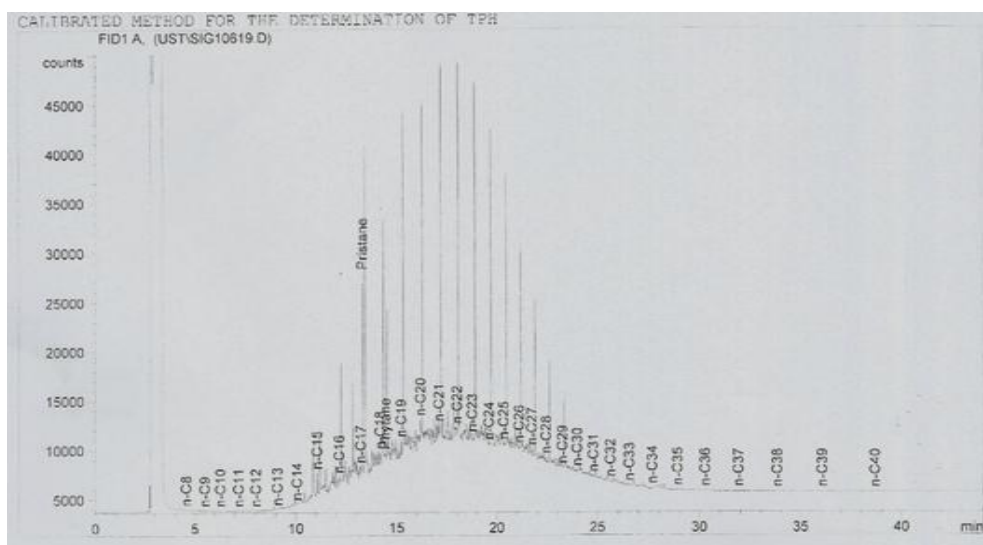


Figure 4a. Chromatogram of petroleum contaminated Sandy loam soil (Two weeks)

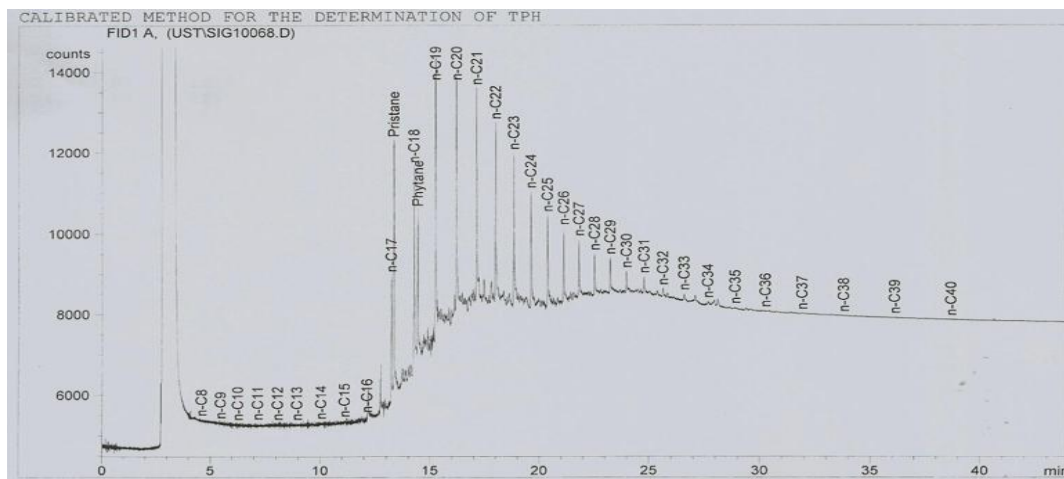


Figure 4b. Chromatogram of petroleum contaminated Sandy loam soil (Four weeks)

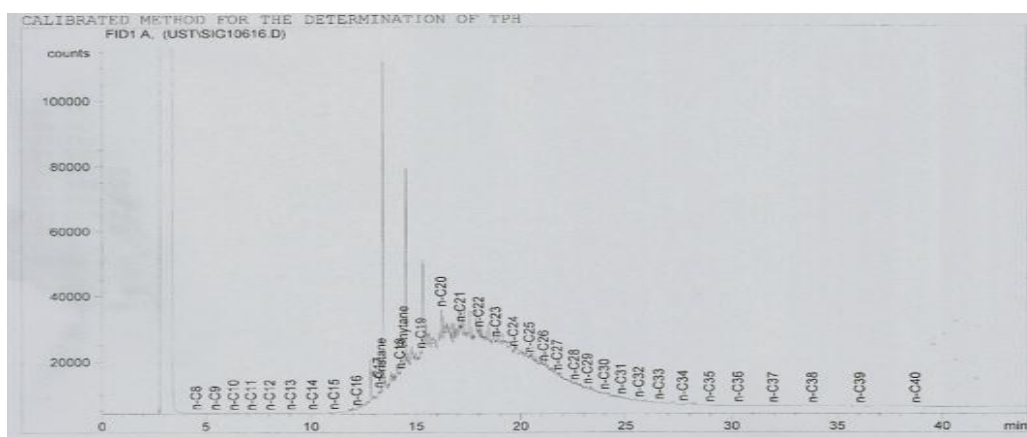


Figure 4c. Chromatogram of petroleum contaminated Sandy loam soil (six weeks)

3.4 Variation of Moisture Content with time

Figure 5 show the variation of Moisture Content with Time in soils in reactor A, B and C for Sandy loam soils. The graphs revealed that the percentage of Moisture Content reduced in the soils after contamination with crude oil. This was because, the inner part of the soil particles could not get wet due to formation of hydrophobic layer (Ayotamuno *et al.*, 2006). The moisture content of the soil increased within the second week of bioremediation process, thereafter it decreased as the process moved towards the last week of remediation.

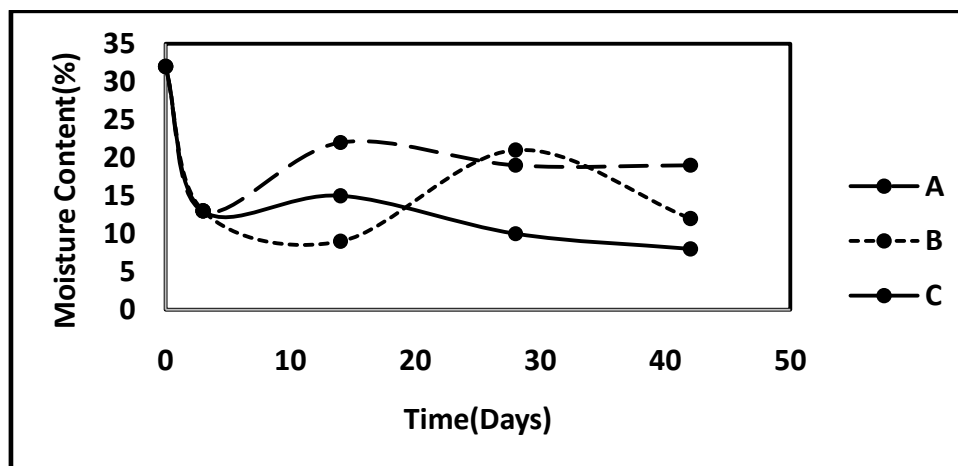


Figure 5: Variation of Moisture Content with Time

These were as a result of evaporation and continuous drying of the soils which maintained an optimum moisture level for biodegradation process (Ayotamuno *et al.*, 2006). The soils in reactor C show more reduction in moisture content compared to soils in reactor A and B. These were as a result of biochemical reactions which took place because of rapid biodegradation processes of petroleum contaminated soils at 100cm depth. These may have occurred due to sufficient oxygen provided by oxygen diffusion at 100cm depth which enhanced the actions of biodegradation of the hydrocarbons by microorganisms.

3.5 Variation of Total Organic Carbon with time

Figure 6 shows the variation of Total Organic Carbon with time. The concentration of Total Organic Carbon in soils in reactor A, increased sparingly from 2.420% - 2.986% for the entire period of remediation, while those for soils samples in reactor B and C increased rapidly within the second and fourth week, but later decreased with time towards the end of the remediation process.

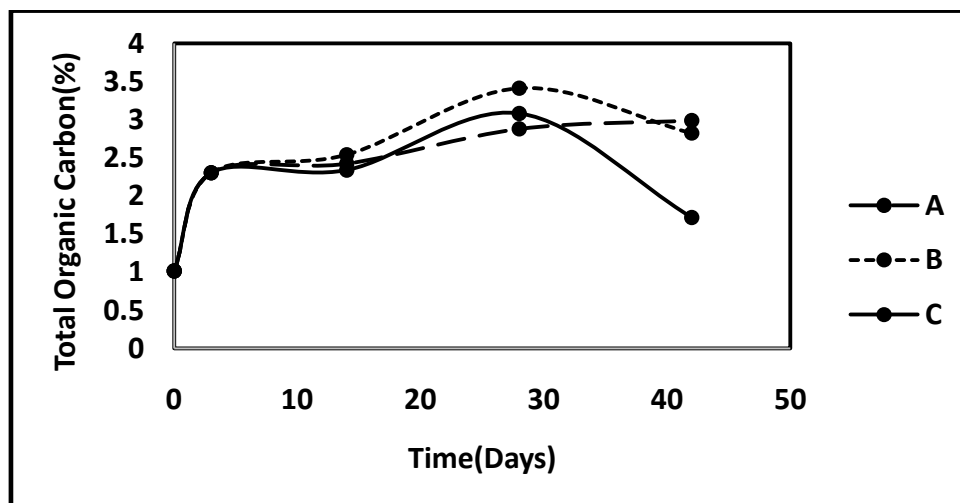


Figure 6: Variation of Total Organic Carbon with time, Sandy loam soil

The decrease was as a result of loss of organic carbon in the form of carbon dioxide due to microbial activities (Prakash and Karegan, 2010). The decrease in concentration of TOC in soil samples in reactor C, may be appreciably because of available sufficient oxygen which enabled the microorganism to consume more quantity of organic carbon for energy during degradation of hydrocarbons.

3.6 Variation of Total Nitrogen with time

Figure 7 shows the variation of Total Nitrogen with Time in soils in reactors A, B and C for Sandy loam and. Figure 7 shows a constant increase in percentage of Total Nitrogen for soils in reactor A and B, but a decrease for soils in reactor C. The reduction in percentage of Total Nitrogen in soils in reactor C revealed that oxygen diffusion at 100cm depth promoted an increase in population of microorganism.

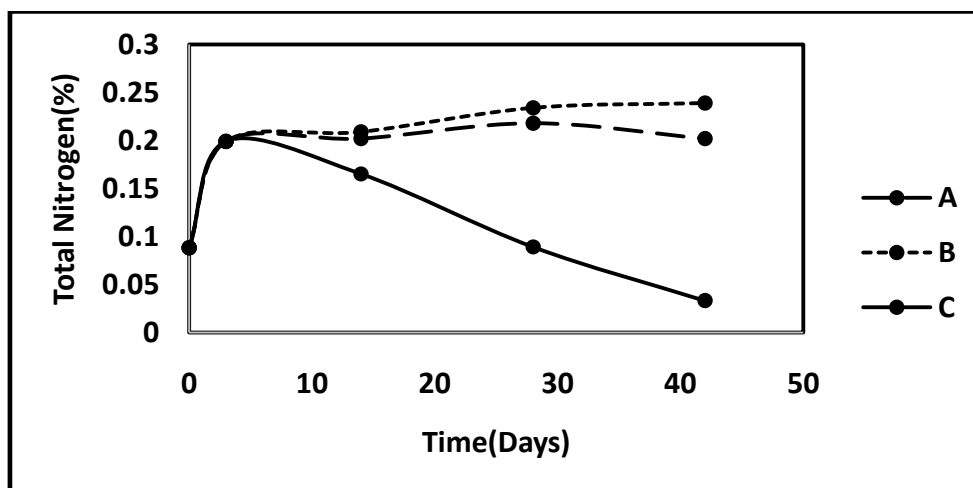


Figure 7: Variation of Total Nitrogen with time

This rise in population of the microbes gave rise to higher demand for nitrate in the soil by the microbes for use in biodegradation of hydrocarbons in the petroleum contaminated soils thereby depleting the percentage of Total Nitrogen in the soils despite the addition of artificial Fertilizer (NPK 27:13:13) to enhance the soil nutrients. Also, the decrease in amount of nitrate in soils in reactor C was because of the usage of the nitrate by the larger population of microorganism for digestion processes. (Liorodache and Borza, 2011).

3.7 Variation of Phosphorous with time

Figure 8 shows the variation of Phosphorous with Time in soils in reactor A, B and C for Sandy loam soils. In soils in reactor A, there was a slight decrease in percentage of phosphate in the soils, while the soils in reactor B experienced a constant rise in percentage of phosphate for the entire bioremediation period, but with a rapid decrease in percentage of phosphate in soils in reactor C.

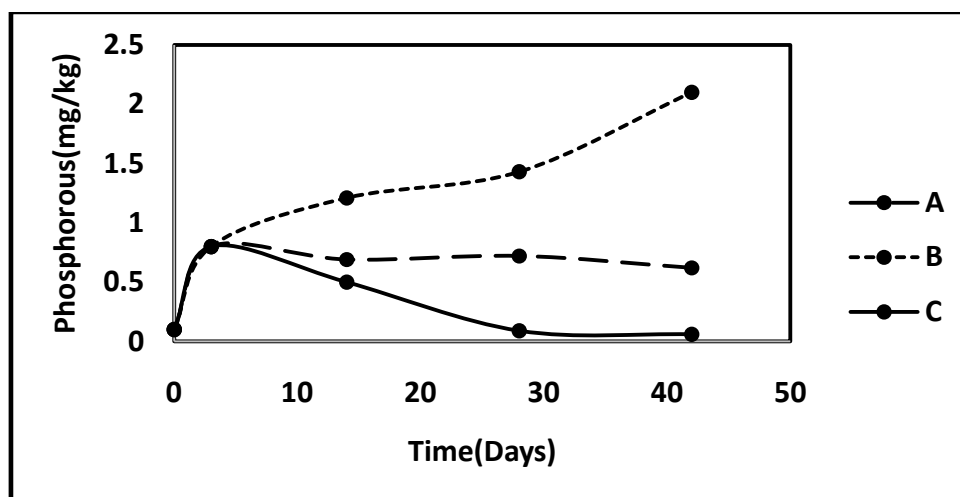


Figure 8: Variation of Phosphorous with time, Sandy loam soil

The rapid rise in percentage of phosphate in soils in reactor A and B indicated low amount of microorganism to utilize the phosphate at 100cm depth due to low amount of oxygen to promote their existence and growth in population, hence there was less demand for Phosphate as nutrient, unlike the soils in reactor C, where rapid decrease in concentrations of phosphate were experienced, because of high demand for phosphate by microorganism as nutrient during biodegradation of hydrocarbon. The higher population of microorganism at 100cm depth were facilitated by oxygen diffusion which provided sufficient oxygen to enhanced increase in microbial population to degrade hydrocarbon content.

IV. CONCLUSION

Oxygen diffusion through petroleum contaminated Sandy loam soils showed significant effect on the physicochemical properties of Sandy loam soil at 100cm depth. The results indicated that, the physicochemical characteristics of the treated Sandy loam soils have similar soil behavior when compared with initial properties of the soils before contamination with Bonny light crude oil. Therefore, this confirmed that oxygen diffusion through petroleum contaminated sandy loam soil was an effective method and a good approach for treatment of petroleum contaminated Sandy loam soil at the depth of 100cm.

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