OPEN ACCESS

Global Journal of Research in Engineering & Computer Sciences
ISSN: 2583-2727 (Online)
Volume 04| Issue 04 | July-Aug. | 2024
Journal homepage: https://gjrpublication.com/gjrecs/

Research Article

Nutrient-Enhanced Degradation of Polycyclic Aromatic Hydrocarbons in Crude Oil-Contaminated Soil: Plantain Peel Biochar versus Nitrogen Phosphorus Potassium Fertilizer

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DOI: 10.5281/zenodo.13309190 **Submission Date: 05 July 2024 | Published Date: 13 Aug. 2024**

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Abstract

This study compared the efficacy plantain peels biochar, and a commercial nitrogen phosphorus potassium fertilizer for the remediation of polycyclic aromatic hydrocarbons (PAHs)-contaminated soil at laboratory scale.

PAHs concentrations ranged from 0.245 to 348.04 mg/kg; with sum concentrations of 698.459, 694.213, and 687.892 mg/kg after 30, 60, and 90 day remediation, respectively for the control sample (sample without amendment). Benzo(a)pyrene had the least concentration, while Chrysene has the highest concentration. PAHs concentration degradation was quicker with PPB amendment option. That is, with PPB amendment option, the sum concentration of PAHs obtained were 649.743, 634.532, and 550.369 mg/kg after 30, 60, and 90 day experiment, respectively. Similarly, with the NPK option, PAHs concentrations were 663.961, 644.215, and 633.766 mg/kg, respectively for the different measurement intervals.

Consequently, to support quick reclamation and/or restoration of PAHs-contaminated soils, bioremediation should be encouraged and implemented. Overall, PPB is recommended for PAHs contaminated soil remediation.

Keywords: Agro-based waste; crude oil; contaminated soil; bioremediation.

1. INTRODUCTION

Soil pollution and environmental degradation caused by crude oil (petroleum hydrocarbon) spills is a common challenge many parts of the world. Soil pollution by crude oil is caused by several cases including crude oil exploration and exploitation activities, accidental spills, and sabotage; which has affected human health, and environmental health and its stability (CL: AIRE, 2015; Zhao et al., 2022).

Crude oil spill assisted environment pollution and degradation is well pronounced in the Niger Delta region of Nigeria because Niger Delta is the seat of the oil and gas industry in Nigeria. IN the Niger Delta region of Nigeria (Ogoni land in Rivers State), the United Nation Environment Program (UNEP) in 2011 reported that crude oil pollution has deeply affected soil, air, and water quality criteria and thus posing a serious threat to both human health and the environment. Like the Ogoni land, other crude oil producing states, especially, Bayelsa, Delta, Akwa Ibom, and Ondo states are faced with same challenges. The devastating environmental pollution and degradation and associate impacts in the Niger Delta region of Nigeria are primarily as a result of oil theft; oil bunkering; artisanal (illegal) refining of crude oil in Nigeria; technical or operational error; un-serviced oil infrastructure; and hazardous waste management. Crude oil is complex mixture of both organic and inorganic compounds. Its composition includes petroleum hydrocarbons, potentially toxic elements (PTEs) such as lead (Pb), zinc (Zn), cadmium (Cd), nickel (Ni), arsenic (As), titanium (Ti), silver (Ag) amongst others (Douglas et al., 2020). Soil pollution by PTEs has attracted substantial ecological concern due to their toxicity and bioaccumulation. Nigeria has been recorded largest natural gas reserve and the second largest oil reserve in Africa, thus faced with the challenges of environmental pollution and degradation. Thus, there is urgent need for contaminated land assessment, risk assessment, remediation and management in the Niger Delta, Nigeria. On the aspect of remediation, environment friendly, and cost-effective approach be investigated at laboratory scale before field trial.

Efforts have been made on the remediation of hydrocarbon contaminated site, using different remediation methods. For instance, biostimulation has been used (Lim et al., 2016; Wu et al., 2016, 2019; Zhang et al., 2020); bioaugmentation (Nwankwegu and Onwosi, 2017; Wu et al., 2019); Bio attenuation (Machando et al., 2019; Agarry and Latinwo, 2015); Bioventing (Sharma, 2019; Trulli et al., 2016; Camenzuli and Freidman, 2015); Landfarming (Brown et al., 2017; Wang et al., 2016; Guarino et al., 2017); Bio-piles (Wu and Coulon, 2015; Raju and Scalvenzi, 2018); Composting (Renet al., 2018; Saum et al., 2018); Windrow (Jiang et al., 2016; Azubuike et al., 2016); Verm remediation (Shi et al., 2020; Njoku et al., 2016); Mycoremediation (Kumar et al., 2018; Ali et al., 2017; Anderson and Juday, 2016). However, all contaminated land treatment and/or remediation methods have their advantages and disadvantages. Douglas et al. (2024) used agro-based waste for the remediation of crude oil contaminated. The authors compared the effectiveness of cattle manure and poultry manure and concluded that poultry manure outperformed cattle manure. For the selection of agro-based waste for environment friendly and cost-effective means of remediating crude oil contaminated land sites, the authors recommend further studies to consider other agro-wastes. Consequently, this study compared the potential of plantain peel biochar and a commercial nitrogen phosphorus potassium (NPK: 20-10-10) for the remediation of polycyclic aromatic hydrocarbon (PAHs)-contaminated soil at laboratory scale.

2. MATERIALS AND METHODS

2.1 Soil Sampling

4kg uncontaminated soil sample was collected from the Niger Delta University, Department of Agricultural and Environmental Engineering Research Farm, Amassoma, Bayelsa State, Nigeria. The soil sample was collected from top 0-20cm layer using a hand trowel and taken with polythene bags to the laboratory.

2.2 Soil Sample Preparation for Hydrocarbon Analysis

The4kg soil sample was sieved using 4 mm sieve, then three sets of samples were measured out (1kg each) into three different sample pots (A, B, and C). The three set of samples were spiked with 150ml crude oil each, and stirred thoroughly with different spatulas. Sample C was used as a control.

2.3 Preparation of Plantain Peel Biochar

A bounce of plantain was bought from Swali Ultra-Modern Market (SAM), Bayelsa State, Nigeria. The plantains were peeled to obtain the peels. The peels were sliced to smaller pieces and sun dried at ambient condition for 2-3 days. The dried plantain peels were packaged were carbonized using furnace at low-temperature (300°C) to produce the plantain peel biochar (PPB).

2.4 Preparation of commercial NPK Fertilizer Biochar

A commercial nitrogen phosphorus and potassium (NPK: 20-10-10) fertilizer was bought from Market. Prior to application as an amendment, the fertilizer (in lumps) was grounded using mortar and pestle in the laboratory, and sieved using 2mm sieve.

2.5 PAHs Analysis by GC-MS

PAHs analysis by gas chromatography coupled to mass spectrometry (GC-MS) strictly followed the method described in Douglas et al., 2024. PAHs were analyzed in both the control, PPB biochar amended, and commercial NPK fertilizer amended soils after 30, 60 and 90 days laboratory experiment.

3. Results and Discussion

3.1 Effect of PPB and commercial NPK fertilizer on PAHs degradation

The effect of PPB and NPK fertilizer on the degradation of PAHs in crude was evaluated and presented in Table 3.1. The results of PAHs concentrations obtained after 30 day, 60 day, and 90 days remediation period were 649.743 mg/kg, 634.532 mg/kg, and 550.369 mg/kg, respectively. The control sample concentrations after 30, 60, and 90 day period were698.459, 694.213, and 687.892 mg/kg, respectively. Results show that the use of PPB for contaminated soil is a promising approach for hydrocarbons-contaminated soil reclamation. Based on the results, the removal efficiencies obtained were6.97; 9.15; and 21. 2% after 30 day, 60 day, and 90 day, respectively (Table 3.2). Result shows that the removal efficiency increases with remediation time period. The removal efficiency of total petroleum hydrocarbon using poultry manure and cattle manure in 45 day laboratory experiment (Douglas et al., 2024). With poultry manure, the removal efficiency was 36%; with it was 23% using cattle manure. Considering the remediation intervals (i.e., 45 day and 60 day), the results did not vary markedly. Another study also reported 79% and 71% degradation while remediating total petroleum hydrocarbon (TPH)-contaminated soil in 28 day laboratory experiment (Obiakalaiji et al., 2015). Though the performance is better than the performance obtained in this study, the current study focused on PAHs, hence cannot be compared.The potential of fresh beef cattle manure for total hydrocarbon contents biodegradation in crude oil-contaminated soil was investigated (Soretire et al., 2017).66% biodegradation efficiency was achieved after eight weeks, and the authors attributed the effectiveness to the by-product of rumen compartment which is known to be a reservoir of diverse species of micro-organisms reported previously (Jannsen and Kirs, 2008).

Similarly, with the commercial NPK fertilizer, the PAHs concentrations after 30 day, 60 day, and 90 day remediation period were 663.961 mg/kg, 644.215 mg/kg, and 633.766 mg/kg, respectively. The results are presented in Table 3.1. Comparing with the control sample concentrations:698.459, 694.213, and 687.892 mg/kg after 30, 60, and 90 day; NPK fertilizer is also promising for hydrocarbon-contaminated soil remediation. From the results, PAHs removal efficiencies were 4.94, 7.05, and 9. 26% after 30 day, 60 day, and 90 day, respectively (Table 3.2). The removal efficiencies are low compared to those obtained with PPB in this study, and those reported (Douglas et al., 2024). This may be attributed to the difference in composition of the different amendments used or soil types. Therefore, further study should i) investigate the efficacies of these nutrients in a single study to select best option for cost-effective remediation of crude-oil contaminated soils; and ii) soil types, as soil types affect bioremediation efficiency.

Table3.1: Results for control, PPB and commercial NPK fertilizer assisted biodegradation of polycyclic aromatic hydrocarbons (PAHs) in crude oil contaminated soils at laboratory scale.

Hydrocarbon	**Control experiment without amendment (in days)**	**Remediation with PPB amendment (in days)**			**Remediation with NPK amendment (in days)**		
PAHs (mg/kg)	**4d**	**30d**	**60d**	**90d**	**30d**	**60d**	**90d**
Acenaphthylene	35.089	29.56	27.859	26.415	31.864	28.625	27.851
Acenaphthene	6.987	4.954	4.274	4.022	5.942	5.197	4.685
Fluorene	0.735	0.516	0.468	0.397	0.607	0.516	0.401
Phenanthrene	77.018	69.135	67.451	65.941	71.025	67.541	66.475
Fluoranthene	70.021	63.142	61.587	59.082	65.714	63.875	62.081
Pyrene	82.978	75.421	73.847	71.206	77.041	75. 208	74.802
Benz(a)anthracene	69.987	60.659	57.451	56.012	62.965	58.514	57.094
Chrysene	348.04	340.473	336.897	334.278	342.643	339.897	336.107
Benzo(k)fluoranthene	5.861	4.976	4.107	3.814	4.896	4.107	3.705
Benzo(a)pyrene	0.245	0.104	0.097	0.054	0.184	0.102	0.092
Indeno (1, 2, 3-c, d) pyrene	0.578	0.309	0.196	0.105	0.477	0.306	0.196
Dibenz(a,h)anthracene	0.296	0.107	0.089	0.062	0.198	0.118	0.098
Benzo(g,h,i) pyrylene	0.624	0.387	0.209	0.187	0.405	0.209	0.179
Total PAHs	**698.459**	**649.743**	**634.532**	**550.369**	**663.961**	**644.215**	**633.766**

Table 3.2: Results for polycyclic aromatic hydrocarbons (PAHs) concentration removal efficiencies based on PPB and commercial NPK fertilizer assisted biodegradation.

Remediation time (day, d)	**Amendments**	**Conc. of PAHs (mg/kg)**	**Variation(mg/kg)**	**Removal efficiency (%): Formula = [(C_s - C_t)/C_s] × 100**
30	PPB	649.743	14.218	6.97
	NPKf	663.961		4.94
60	PPB	634.532	9.683	9.15
	NPKf	644.215		7.05

90	PPB	550.369	83.076	21.2
	NPKf	633.766		9.26

NPKf= nitrogen phosphorus and potassium; **PPB**= plantain peel biochar; **C_s**= initial concentration of PAH; **C_t**= concentration of PAHs time, t.

3.2 Regression Analysis between the Two Amendment Options

Regression and analysis were carried out investigate the relationship between the two amendment options (Figure 3.1-3.3). The R-square value of 0.9999 is an indication that a good relationship existed between the two (n = 2) different remediation options (agro-waste PPB and commercial NPK fertilizer) evaluated after 30 day remediation period (Figure 3.1). Similarly, the R-square value (0.9999) obtained (Figure 3.2) is an indication that a good relationship existed between PPB and commercial NKP fertilizer) amendment options after 60 day remediation period.

R-square value of 0.9999 was obtained from the plot of degraded PAHs concentration due NPK amendment versus PPB amendment after 90 day (Figure 3.3). The R-square value indicates that a good relationship existed between the two different remediation options evaluated.

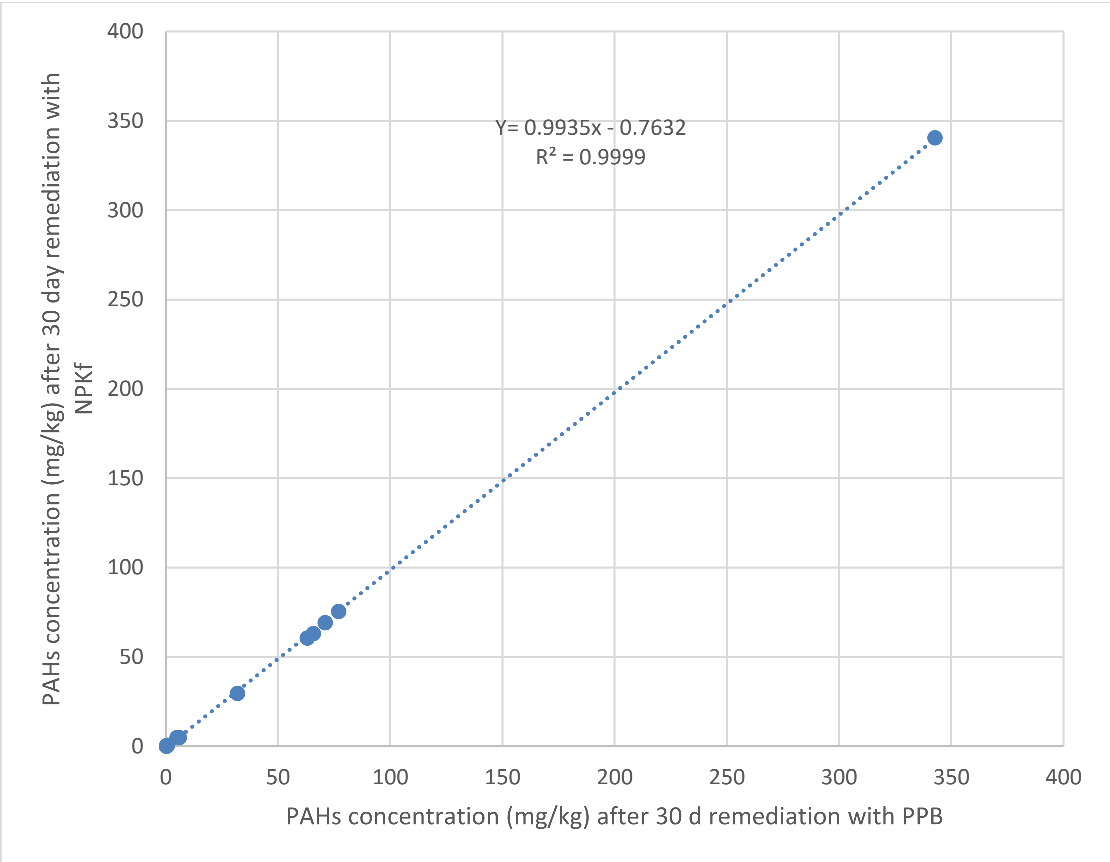

Figure 3.1: A plot of regression analysis showing the relationship that existed between PPB amendments and commercial fertilizer (NPK) for PAHs in crude oil contaminated soil after 30 days.

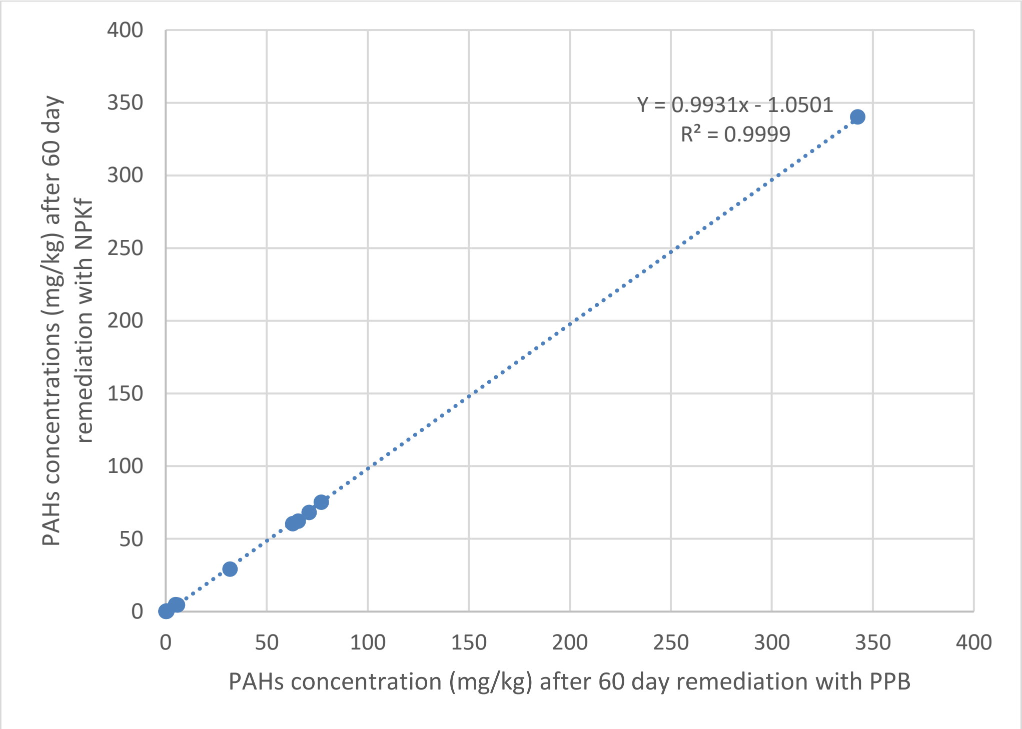

Figure 3.2: A plot of regression analysis showing the relationship that existed between PPB amendments and commercial fertilizer (NPK) for PAHs in crude oil contaminated soil after 60 days.

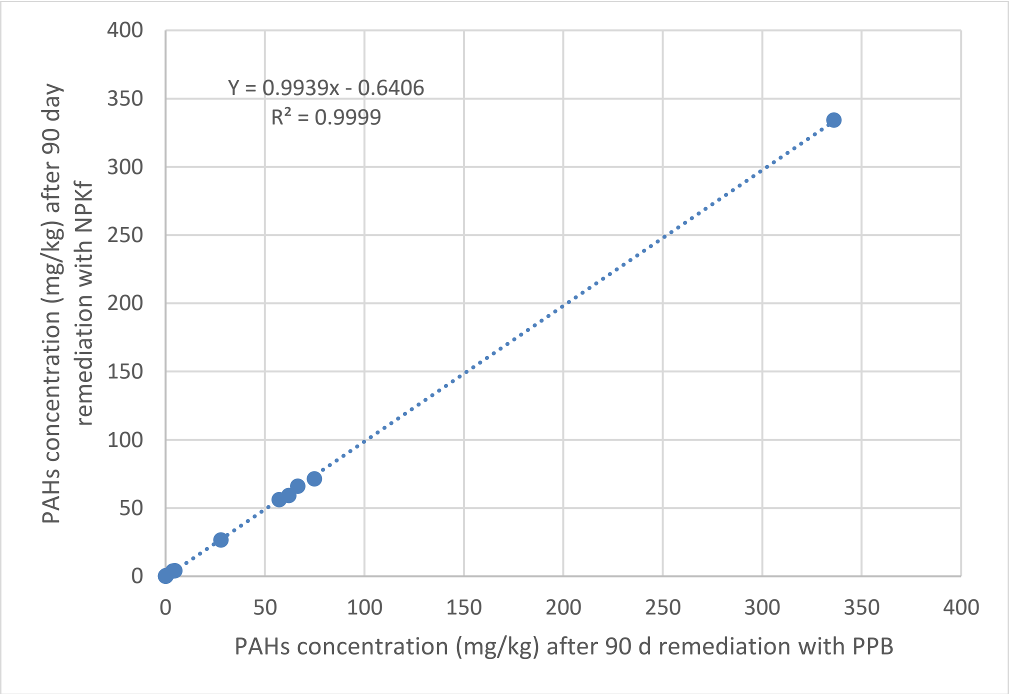

Figure 3.3: A plot of regression analysis showing the relationship that existed between PPB amendments and commercial fertilizer (NPK) for PAHs in crude oil contaminated soil after 90 days.

The summary results of plots generated from the various amendment options applied in this research work, remediation time intervals, regression equations, R-square values, and the level of relationship are presented in Table 3.3 below. The different R-square values show that a very good relationship existed in all the amendment options evaluated in this study.

Table 3.3: Summary results of plots of various amendment options, remediation time intervals, regression equations, R-square values, and the level of relationship.

Plot	Remediation time	Regression equation	R-square value	Relationship
NPK vs PPB	30 days	y = 0.9935x-0.7632	0.999	Very strong
NPK vs PPB	60 days	y = 0.9931x-1.0501	0.999	Very strong
NPK vs PPB	60 days	y = 0.9939x-0.6404	0.999	Very strong

4. Conclusions

Environmental challenges triggered by petroleum hydrocarbon pollution cut across human and environment health. As such, efforts have been intensified in environmental remediation to mitigating these challenges in cost-effective and sustainable manners. On that premise, this study assessed the potential of agro-based waste (plantain peel biochar), and a commercial nitrogen phosphorus potassium (NPK) fertilizer for the remediation of PAHs-contaminated soil at laboratory scale.

From the results the following conclusions are made:

1. With PPB amendment option, the sum concentration of PAHs obtained were 649.743, 634.532, and 550.369 mg/kg after 30, 60, and 90 day experiment period, respectively. Similarly, PAHs concentrations were observed to be 663.961, 644.215, and 633.766 mg/kg, respectively for the three different measurement intervals. Thus, PPB remediation option performed better than the commercial NPK fertilizer option.
2. To support quick reclamation and/or restoration of PAHs contaminated soil and/or land sites, bioremediation should be encouraged and implemented.
3. Further study should expand the remediation time interval. This will enable remediation experts using agricultural wastes (in this case, biochar) to understand the long lasting potential of biochar in the soil.
4. Further study should investigate the potential of other agricultural wastes for contaminated soil remediation. This will help uncover the hidden potentials of the abandoned agricultural wastes, and encourage their usage as nutrients for environmental remediation.

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CITATION

Lawrence C. A., & Douglas K.R. (2024). Nutrient-Enhanced Degradation of Polycyclic Aromatic Hydrocarbons in Crude Oil-Contaminated Soil: Plantain Peel Biochar versus Nitrogen Phosphorus Potassium Fertilizer. In Global Journal of Research in Engineering & Computer Sciences (Vol. 4, Number 4, pp. 114–120). https://doi.org/10.5281/zenodo.13309190