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Adsorption of Petroleum Hydrocarbons from Crude Oil Polluted Soil Using Agro-waste

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Abstract

This study was carried out to ascertain the Adsorption of heavy metals from crude oil-polluted soil using agro-waste. Samples of garden soil with no history of crude oil pollution were spiked with 100mL of Bonny light crude oil and left for two weeks to simulate the condition of a major spill before adding different weights of palm bunch ash (0, P+NOPBA, 50g, 100g, and 150g). Preliminary results revealed alteration of chemical properties of soils, elevated heavy metals levels, and TPH content one month after spiking. Metal content increased significantly from ND (not detected) to Cr (1.41 mg/kg), Pb (1.18 mg/kg), Cd (0.30 mg/kg), and As (1.93 mg/kg) respectively. The initial TPH content was 176.81 mg/kg whereas, one month after spiking with crude oil, the value increased to 1,535.5 mg/kg indicating that the soil sample had undergone alteration concerning TPH. There was a dose-dependent decrease in TPH and heavy metal content of the crude oil-polluted soils with time. Net reductions of total petroleum hydrocarbon concerning treatment levels at the end of the experiment were P+US 1,409 (43.7%), P+50 g 1,320 (72.7%), P+ 100g 1,122 (87.9 %), P+150g 1,043 (98.9%). Overall, the net reduction in heavy metals and TPH was very low in the soil left under natural attenuation than in treated soils. Net reduction of heavy metals (Pb, Cr, Cd, and As) was as follows: P+US (23.7%), (37.8%), (26.9%),(31.8%), P+50 g (85.7%), (88.4%), (86.1%), (77.3%), P+100g (93.9%), (94.6%), (93.5%), (89.1%) and P+150g (98.9%), (99.9%), (98.7%), (93.7%). This study has established a marked degradation of the heavy metal and hydrocarbon contents of soil which indicated that agro-waste could be used for the remediation of crude oil-polluted soil. It is recommended that agro-waste be replaced with conventional fertilizer in the restoration of crudely contaminated soil.

Keywords: Adsorption, Crude oil, Heavy metals, Agrowaste, Imo State

1 Introduction

Petroleum hydrocarbon contamination of soil is a widespread global environmental concern [1]. Oil and fuel spills in soil are among the most extensive and environmentally damaging pollution problems as it is threatening human health and ecosystems [2], especially in cold regions. Biochemical and physicochemical properties of soil are deteriorated by refinery products and it also limits the growth and development of plants [3]. Petroleum hydrocarbon-contaminated soil causes organic pollution of underground water which restricts its use and causes economic loss, environmental problems and decreases the agricultural productivity of the soil [2]; [4]. Microorganisms, plants, animals, and humans are facing vulnerable situations because of the toxicity of petroleum hydrocarbons Petroleum hydrocarbon is released into the sea, normally during transportation, leading to the pollution of several sites, and can eventually reach the coasts [5]. Oil spills ranging from low-level discharges to catastrophic accidents threatened coastal environments; large spills commonly are followed by clean-up efforts [4], but complete containment is rare [6]. As the solubility of petroleum hydrocarbon in water is generally low, certain fractions of it float in water and form thin surface films, which will facilitate the agglomeration of particles and natural organic matter, and impact oxygen transfer [7]. Other heavier fractions will accumulate in the sediment at the bottom of the water, which may affect bottom-feeding fish and organisms [8]. Crude oil spillage on water affects aquatic life, by reducing the level of dissolved oxygen in the water, which adversely affects aquatic life [9].

Conventional methods for removal, reduction, or mitigation of toxic substances introduced into the soil or groundwater via anthropogenic activities and processes include: pump and treat systems, soil vapor extraction, incineration, and contaminant utility of each of these conventional methods of treating contaminated soil suffers from recognizable drawbacks and may involve some levels of risk [1]; [7]; [10]; [11]. The application of these conventional methods of treating Petroleum has continuously grown to be a mainstay of the Nigerian economy [12]. However, the exploration of petroleum has led to the pollution of land and waterways [13]. Soil is a primary recipient by design or accident of a myriad of waste products and chemicals used in modern society [14]. Over the years, in oil-rich and exploiting countries, efforts have increasingly been taken to remediate contaminated sites [4]. Different approaches; physical, chemical, and biological have been undertaken [15]. However,

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some of these are expensive while others have harmed the environment, particularly soil health and human livelihoods [16]. For example, excavation (physical approach) has logistics [2]; and transport constraints while incineration (chemical approach) adds greenhouse gases to the atmosphere leading to global warming) Biological approaches such as bioremediation are known to be the safest and most ecofriendly approach for the remediation of polluted soils [1]. Bioremediation is the use of biological agents, mainly microorganisms (e.g., yeast, fungi, or bacteria) or their enzymes to destroy or reduce the concentration of hazardous wastes on a contaminated site [17]. The potential of agro-waste (*E. guinensis*) in the adsorption of pollutants from the environment has not been fully exploited. It is on this basis that the study was undertaken to evaluate the adsorption of pollutants from crude oil-polluted soil using agro-waste.

2 Materials and Methods

2.1 Experimental site

The study was carried out at Hezekiah University Umudi which lies at latitude 5.3866 ° N and longitude 6.9916° E in the Orlu zone of Imo State. There was no history of crude oil pollution in the area at the time of carrying out this study.

2.2 Sample collection and processing

Sandy-loamy topsoil was collected within 0 – 20 cm depth from a fallow plot located in Hezekiah University Umudi after the removal of plant debris and exposed surface following the method [20]. Agro-waste (*E. guinensis*) was collected from an oil mill in Nkwerre L.G.A of Imo State while crude oil (Bonny light) was gotten from the Department of Chemical Sciences.

2.3 Sample preparation/analysis

Sample preparation followed the method of [21] with slight modifications. Collected soil samples were air-dried and sieved using a 2 mm sieve for routine post-soil analysis before treatment and adsorption trial. The Agrowaste (*E. guinensis*) was shredded into smaller pieces and sun-dried for two weeks. The shredded and dried bunch was combusted and ground into powdered form and later passed through a 2 mm standard mesh sieve.

2.4 Mineral element analysis

About 1 kg of the processed sample was taken to the laboratory for mineral constituents. Potassium and sodium were analyzed using the digested extracts of the samples and taking readings on Jenway Digital Flame Analyser PFP7. Calcium (Ca), Magnesium (Mg), Phosphorus (P), Zinc (Zn), Copper (Cu), Iron and (Fe), and Manganese (Mn), were determined using AA-7000 Atomic Absorption Spectrophotometer adopting a procedure recommended by Environmental Protection Agency (EPA, Method 3050B).

2.4 Heavy metal analysis

2.4.1 Digestion and analysis of soil samples

This followed the method of [21]. Briefly, about Ten grams (10 g) of the soil sample was placed into a 50 ml beaker and 5 ml of concentrated aqua-regia (HCl: HNO_3 in the ratio of 3:1) was then added. The mixture in the beakers was covered with a watch glass and boiled on a hot plate at 125 ^{0}C for 2hrs. A small portion of aqua-regia was added until the solution appeared light in color and clarity. The beaker wall and watch glass were then washed with deionized water and the samples were filtered to remove insoluble materials that could clog the atomizer. Then, the digest was allowed to cool at room temperature and filtered through a Whatman 1 filter paper in a 50 ml volumetric flask. The volume of the filtrate was made up to 50ml with distilled water. A control sample was prepared to allow a blank correction to be made. These were then aspirated into the GBC Avanta Atomic Absorption Spectrophotometer model for the determination of each metal (Cadmium, Arsenic, Chromium, and Lead), and the concentration expressed in mg/kg dry weight.

2.4.2 Determination of Total Petroleum Hydrocarbon

Some quantity (2 g) of the sample was weighed into a clean extraction container. Exactly 20 ml of hexane (extraction solvent) was added to the sample, mixed thoroughly, and allowed to settle. The mixture was carefully filtered into a solvent-rinsed extraction bottle using filter paper fitted into Buchner funnels. The extract was concentrated to 2 ml and transferred for separation/clean-up.

2.5 Experimental Design/treatment combination

About 2000 cm^2 of plastic pots were filled with the topsoil and each was spiked with 100 ml of crude oil. The experiment was laid out in a Complete Randomized Design with three replicates per treatment and used for analysis. About 1 kg of each of the soil was properly mixed with 100 ml of crude oil to simulate the condition of a major spill. Each sample was tilled for 2 min twice a week and analyzed fortnightly for 60 days (2 months) for even mixing of the Agrowaste (*E. guinensis*) [22]. The soil physicochemical properties, mineral analysis of oil palm bunch ash, total petroleum hydrocarbon content, and selected heavy metal (Pb, As, Cr, and Cd) concentrations of soil samples were analyzed to track the rate of degradation in the treatments using standard methods.

2.6 Method of Data Analysis

Data were analyzed using Statistical Package for Social Sciences (SPSS) Incident 21.0 Software. Descriptive and one-way Analysis of Variance (ANOVA) for comparison was carried out and results were expressed in tables and charts as mean ± standard deviation (M±SD).

3 Results

Table 1 shows the elemental properties of palm bunch ash used for the study. The pH showed high alkalinity with a value of 8.5. The percentage moisture content was 32%. The values for micronutrient content of the Agrowaste (*E. guinensis*) fluctuated as follows: Potassium (40.04), Calcium (6.19), Magnesium (5.24), Sodium (6.02), Zinc (1.02), Iron (1.12), Copper (0.86), and Manganese (0.72) respectively.

Table 1: Elemental properties of Agrowaste (*E. guinensis*) used for the study

Parameters	Values
pH	8.5
Moisture content (%)	32
Potassium (ppm)	40.04
Calcium (ppm)	6.19
Magnesium (ppm)	5.24
Sodium (ppm)	6.02
Zinc (ppm)	1.02
Iron (ppm)	1.12
Copper (ppm)	0.86
Manganese (ppm)	0.72

The chemical properties of experimental soil samples before and after crude oil application is presented in table 2. The result indicated that crude oil contamination of the soil

altered the selected chemical properties of the soil one month after spiking. The values for pH, TN, OC, OM, and available phosphorus before and after spiking changed from 5.61, 0.04, 0.71, 1.23, 28.44 to 6.9, 0.02, 2.51, 4.46, and 8.40 mg/kg respectively. However, organic carbon content was higher in the polluted soil than in the unpolluted soil. Metal content increased significantly from ND (not detected) to Cr (1.41 mg/kg), Pb (1.18 mg/kg), Cd (0.30 mg/kg), and As (1.93 mg/kg) respectively. Generally, elevated concentrations of selected heavy metals were observed one month after spiking when compared with the unpolluted soil (before spiking). The initial TPH content was 176.81 mg/kg whereas, two weeks after spiking with crude oil, the value increased to 1,535.5 mg/kg indicating that the soil sample had undergone slight alteration for the TPH content of the soil. This value (1,535.5mg/kg) was found to be above the permissible limit of 1,000 mg/kg set by the Department of Petroleum Resources (DPR). However, the concentration of selected heavy metals in soil assayed after 2 months after exposure falls above the permissible limit of the standard set by WHO but was all below the permissible limit set by the Nigerian department of petroleum resources (DPR).

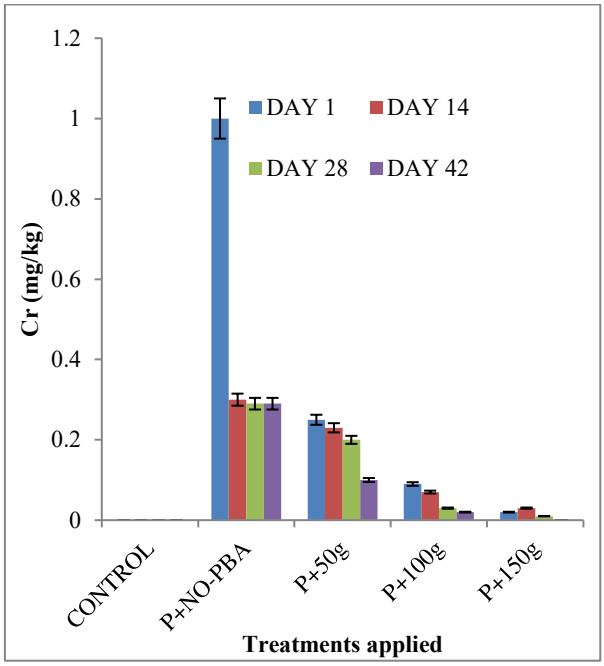

Figure 1: Mean concentration of chromium in crude oil-polluted soil at different sampling times

The effects of different concentrations of Agrowaste (*E. guinensis*) on the heavy metal content of crude oil-polluted soil over the study period is displayed in figures 1, 2, 3, and 4. There was a progressive decrease in the heavy metal content of the crude oil-polluted soils with a corresponding increase in treatment levels. The decrease in heavy metal content was dose-dependent. Samples treated with various levels of palm bunch ash performed better in terms of remediation when compared with soil samples left under natural attenuation (Polluted Untreated soil).

As seen in Figure 1, treatment with Agrowaste (*E. guinensis*) showed an optimum Total Petroleum Hydrocarbon content reduction for 50g, 100g, and 1500g treatment levels in a dose-dependent manner, with minimum reduction rate observed in polluted untreated soil and maximum reduction rate recorded at 100g and 150g treatment levels during the sampling periods.

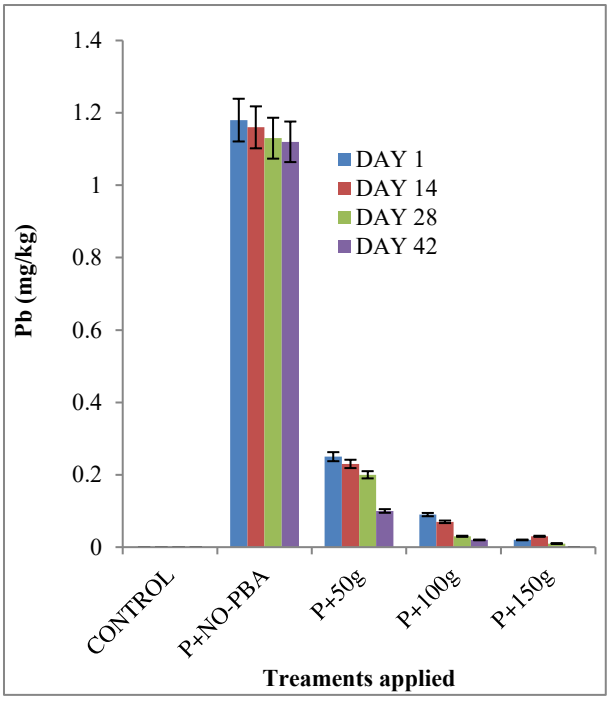

Figure 2: Mean concentration of Lead in crude oil-polluted soil at different sampling times

Table: Pre-experimental soil properties before and after spiking

Parameters	Before Spiking	After Spiking	DPR Limit	WHO Limit
pH	5.61	6.9	6-9	-
TN (%)	0.04	0.02	-	-
OC (%)	0.71	2.51	-	-
OM (%)	1.23	4.46	-	-
Av. P (cmol/kg)	5.44	1.60	-	-
Cr (mg/kg)	ND	1.41	100	0.02
Pb (mg/kg)	ND	1.18	85	0.01
Cd (mg/kg)	ND	0.30	0.8	0.02
As (mg/kg)	ND	1.93	29	0.009
TPH (mg/kg)	176.81	1,535.5	1,000	-

LEGEND: WHO – world health organization (2016); DPR = Department of Petroleum Resources (2002)

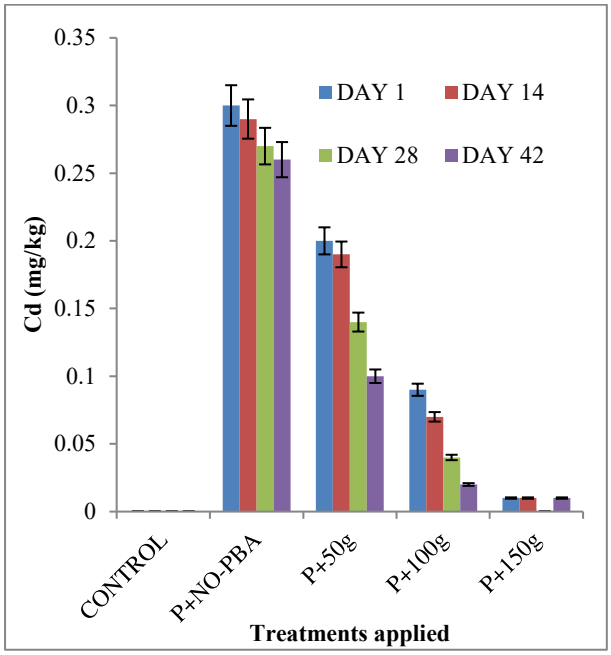

Figure 3: Mean concentration of Cadmium in crude oil-polluted soil at different sampling times

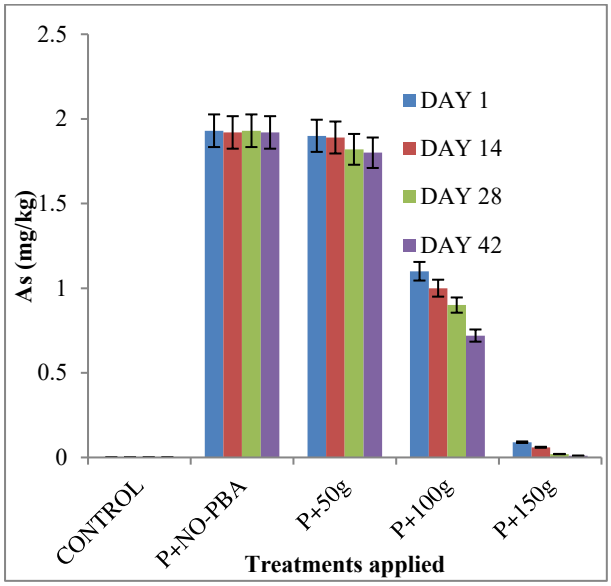

Figure 4: Mean concentration of Lead in crude oil-polluted soil at different sampling times

The percentage reduction of heavy metals and total petroleum hydrocarbon content of post-treated soils are displayed in tables 2 and 3. The result revealed a variation in the reduction rate of crude oil-polluted soils with the increase in treatment levels. Net reductions of total petroleum hydrocarbon for treatment levels at the end of the experiment were as follows: P+US 1,409 (43.7%), P+50 g 1,320 (72.7%), P+ 100g 1,122 (87.9 %), P+150g 1,043 (98.9%). Overall, the net reduction of heavy metals and TPH was very low in the soil left under natural attenuation than in treated soils. Net reduction of heavy metals (Pb, Cr, Cd, and As) was as follows: P+US (23.7%), (37.8%), (26.9%), (31.8%), P+50 g (85.7%), (88.4%), (86.1%), (77.3%), P+100g (93.9%), (94.6%), (93.5%), (89.1%) and P+150g (98.9%), (99.9%), (98.7%), (93.7%).

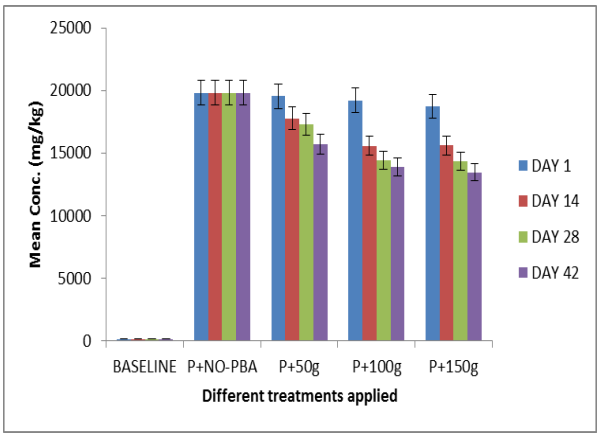

Figure 5: Total Petroleum Hydrocarbon content of soils treated with oil palm bunch ash

Table 2: Percentage degradation of Total petroleum hydrocarbon content (THC) reduction (week 4 post-analysis)

Treatment	TPH (Residual)	Net Reduction (%)
Control	ND	ND
P+US	1,409	43.7
P+50 g	1,320	72.7
P+ 100g	1,122	87.9
P+150g	1,043	98.9

Legend: ND = not detected.

4 Discussion

Results from this study implied that crude oil pollution significantly altered soil properties like pH, TN, OC, OM, and available phosphorus as well as the presence of selected heavy metals. Alteration of soil properties emanating from crude oil pollution had been reported by [23]. [24] also reported elevated levels of heavy metals in crude oil-polluted soil. [25] Reported that crude oil pollution increased the pH of the soil. The high organic carbon content value observed in polluted soil is in line with the findings of [26] who reported that soil organic carbon content increased due to the presence of hydrocarbon in the crude oil.

There was a marked decrease in TN, OM OC, and available phosphorus content of the soil two weeks after spiking. A decrease in soil nutrient content as a result of crude oil pollution had been reported by previous authors which is in line with the findings in this study [27]. The reduction in the level of available soil nutrients and rise in the level of heavy metals is in line with the report of [28]. The high pH (8.5) value of Agrowaste (*E. guinensis*) shows it is an alkaline medium. Therefore, pH of this sample indicates that the sample may be used to reduce the acidity of soils [29]. The high Calcium content of the sample confirms that it may be a useful liming agent [30]. This pH will also support nitrogen fixation and decomposition activities which are known to be hindered in strongly acidic soils [3]. This result corresponds with the physicochemical analysis conducted by [31]. According to [32], Agrowaste (*E. guinensis*) has been reported to contain rich nutrients such as nitrogen, phosphorous, potassium, calcium and magnesium. Thus, the alkaline properties and rich nutrient contents of the palm bunch ash used in this study could be a better source of remediation practice in crude oil polluted soil with an acidic pH and nutrient depleted characteristics [4]. There was a progressive decreased in heavy metal content of

soil with increased treatment levels at various periods of sampling.

Table 3: Net percentage reduction of heavy metals in the soil at the end of the study (4 weeks)

	Percentage Reduction in soil				
		Pb	Cr	Cd	As
Control	ND	ND	ND	ND	
P+US		23.7	37.8	26.9	31.8
P+50 g	85.7	88.4	86.1	77.3	
P+ 100 g	93.9	94.6	93.5	89.1	
P+150 g	98.9	99.9	98.7	93.7	

Legend: ND= not detected

The highest reduction rate was observed in 100g and 150g compared with other treatment levels; whereas the lowest reduction rate was noticed in polluted untreated soils left under natural attenuation. Results showed that the unpolluted soil had low values of total petroleum hydrocarbon (TPH) throughout the investigation. The reduction of TPH from Day 1 to Day 42 in polluted untreated soil was slow in comparison with other treatment levels. This slow reduction observed could be attributed to natural attenuation which hindered indigenous microorganisms to use available nutrients as both carbon and nitrogen sources to degrade hydrocarbon compounds [33]. Results of polluted soil without Agrowaste (*E. guinensis*) showed an increase in TPH value on Day 1 compared to the value obtained for the unpolluted soil, followed by minimal reduction from Day 14 to Day 42 of the experiment. The reason for the high concentrations of TPH observed on Day 1 is due to the pollution of soil with crude oil in such a quantity as to simulate natural pollution. Results of Polluted soil treated with 50g Agrowaste (*E. guinensis*) showed a significant reduction in TPH concentrations from Day 1 to Day 42. A marked reduction observed during the first two weeks of treatment with Agrowaste (*E. guinensis*) could be due to the ability of microorganisms to use the Agrowaste (*E. guinensis*) as both carbon and nitrogen sources to degrade the hydrocarbon compounds in the crude oil. [34] Had reported that amendment of 100g contaminated soil with 30g organic nutrients led to the loss of 40% TPH. There was a significant reduction in TPH values throughout the investigation, which also resulted in the loss of TPH values. From this, it could be deduced that the use of Agrowaste (*E. guinensis*) effectively stimulated organisms into the utilization of crude oil. All the polluted soil treated with different weights (that is, 50g, 100g to 150g) of Agrowaste (*E. guinensis*) followed the same trend as that of polluted soil treated with 50g Agrowaste (*E. guinensis*). There was a decrease in TPH values throughout the experiment for all the soils with different weights of Agrowaste (*E. guinensis*) in a dose-dependent manner. From the observation, PS + 50g Agrowaste (*E. guinensis*) showed the lowest percentage reduction in TPH value (72.7%), followed by PS + 100g PBA (87.9.5%), and PS + 150g which recorded a percentage reduction with a value of 98.9% respectively. [35] had reported that the quantity of manure is important for bioremediation (in supplying nutrients). Indicating that 150g weights of Agrowaste (*E. guinensis*) could be an optimum treatment recipe for crude oil degradation.

5 Conclusion

The treatment of crude oil-contaminated soil with agro-waste revealed that after one month of treatment, the chemical properties of the soil were adversely affected. Addition of different weights of agro-waste aided in the complete disappearance of the saturated hydrocarbons four (4) weeks after exposure. This shows that Agrowaste (*E. guinensis*) which has been confirmed to possess a rich concentration of nutrients and potassium ions species can contribute to the bioremediation of crude oil-polluted soil. This study has also revealed a marked degradation of the hydrocarbons which may be through the activities of microorganisms or emulsification.

Recommendations

- Palm bunch ash could be used in the bioremediation of crude oil-polluted soil
- The reaction mechanism and effect of environmental factors should be analyzed on the remediation of heavy metals from crude oil-contaminated soil using Agrowaste (*E. guinensis*).
- The use of Agrowaste (*E. guinensis*) could be replaced with conventional fertilizer in the restoration of crudely contaminated soil.
- There is a need for further studies on this subject to elucidate the actual mechanism of degradation that took place in this study.

Ethical issue

Authors are aware of and comply with, best practices in publication ethics specifically about authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests, and compliance with policies on research ethics. Authors adhere to publication requirements that the submitted work is original and has not been published elsewhere in any language. Also, all procedures performed in studies involving human participants were following the ethical standards of the institutional and/or national research committee and with the 1964 Helsinki declaration and its later amendments or comparable ethical standards. All procedures performed in this study involving animals were following the ethical standards of the institution or practice at which the studies were conducted.

Competing interests

The authors declare that no conflict of interest would prejudice the impartiality of this scientific work.

Authors' contribution

All authors of this study have a complete contribution to data collection, data analyses, and manuscript writing.

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