Total Petroleum Hydrocarbon and Heavy Metal Remediation with *Jatropha curcas* L. Seedlings Grown on Spent Oil Polluted Soil

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Abstract: Soil contamination by Spent Lubricating Oil (SLO) has adverse environmental effect because of its highly toxic constituents including heavy metals and complex mixtures of heavy Polycyclic Aromatic Hydrocarbons (PAHs). The presence of this wide range of toxic metals and compounds is responsible for its perceived adverse effect on flora and fauna in particular and the environment in general. This study employed *Jatropha curcas* seedlings in phytoremediating organically amended and unamended soil contaminated with 0, 3 and 6% (w/w) SLO for 84 days. Total Petroleum Hydrocarbon (TPH) and heavy metal concentration of the phytoremediated soil were assessed every 28 days and at the beginning and the end of the experiment respectively. Phytoremediation with *J. curcas* seedlings was effective in the breakdown of TPH content of the polluted soil. At the end of the study, TPH reduction was higher in phytoremediated soil with up to 91.54% in amended soil and 84.81% in unamended soil at 3% level of pollution. For the three heavy metals investigated, there was a general reduction in their concentration at the end of the phytoremediation process in the order Pb>Cd>Fe. This study has shown that *J. curcas* seedlings together with organic soil amendment is a veritable tool for phytoremediation of SLO polluted soils.

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1. Introduction

In the review of millennium agenda that is just winding up; the Millennium Development Goals (MDGs), it was generally agreed that Africa's performance has not been very impressive. Sub Saharan Africa, particularly, performed poorly with poverty, hunger and diseases pervading every nook and cranny of the sub-continent. In Nigeria, for example, long years of corruption and bad governance has brought the country almost to her kneels. The over- reliance, of the country, on crude oil resources has led to the neglect of other important sectors such as agriculture and mining. Oil exploration has been without regard to the environment with oil spillages and pollution of the biota being a common spectacle.

Spent oil contamination of soils is another form of oil pollution with devastative environmental consequences in Nigeria and other developing countries of Sub Saharan Africa. Although insidious, the effect of such contamination and pollution is felt in agriculture, animal and human health as well as the general state of the environment. It is known that changes in soil properties associated with spent oil contamination often lead to water and oxygen deficit as well as shortage of available forms of Nitrogen and phosphorus (Wyszokowska and Kucharski, 2000). Contamination of soil environment can also limit soil protective functions, upset metabolic activities, functions and chemical characteristics as well as reduce fertility and ultimately affect plant production (Gang, Sun, Beudert and Hahnm, 1996).

The reason for the perceived dangerous effect of SLO is because it is a complex mixture of hydrocarbons and other organic compounds including some organometallic constituents (Buttler and Mason, 1997). The Polycyclic Aromatic Hydrocarbons and heavy metals inherent in SLO are known to be among the most toxic and hazardous pollutants in the environment (Boonchan, Britz, and Stanley, 2000). Uncontrolled and indiscriminate introduction of this category of pollutants into the environment in this part of the world, is eliciting great concern among scholars and environmentalists especially because of the insidious nature of SLO.

Adegoroye (1997) reported that in Nigeria and some developing countries about 80 million litres of waste engine oil is generated annually from mechanic workshops and discharged carelessly into the environment. This renders the environment unsightly and constitutes a potential threat to humans, animals and vegetation (Adelolowo, Alagbe, and Ayandele, *2006*). The SLO eventually end up on the farmlands, fish ponds, rivers, lakes etc. Bamiro and Osibanjo (2004) gave a more comprehensive estimate of SLO generation in Nigeria. It was reported that up to 200 million litres of spent lubricating or used oil is generated annually mainly from the industrial sector.

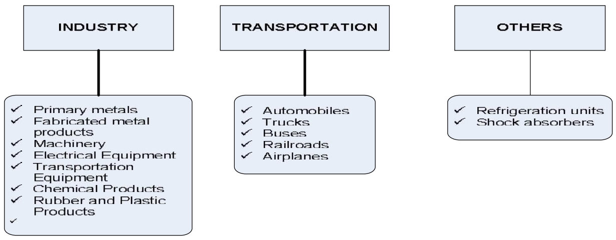

Figure 1: Major Sources of Spent Lubricating Oil Pollution


Plate 1: Improperly Disposed Spent Lubricating Oil by Road Side
Source: Bamiro and Osibanjo (2004)


Plate 2: Stream Polluted by Used Engine Oil
Source: Bamiro and Osibanjo (2004)

It has, therefore, become highly necessary to carry out remedial activities that will ensure that SLO is intercepted and removed from the environment to prevent it from impacting its toxic effects on the flora and fauna components. A veritable option that could be useful in this regard is phytoremediation.

Phytoremediation is the use of living green plants for the removal of contaminants (Nedunuri, *et al.,* 2007). It potentially offers unique, low cost solutions to many problems of soil contamination since it does not require expensive equipment or highly specialized personnel (Liu *et al.,* 2000).

The objective of this work, therefore, was to determine the potential of *Jatropha curcas* in phytoremediating spent oil polluted soil with special emphasis on the organic and inorganic components (Total Petroleum Hydrocarbon and Heavy metals) of the SLO. The choice of *Jatropha curcas* is borne out of the fact that it has been used on heavy metal polluted soils especially hexavalent chromium, in some countries of the world (Mangkoedihardjo, Ratnawati and Alfianti, 2008). Also, it is known to be a highly resistant species which thrives well in adverse conditions (The Biomass Project, 2000).

2. Materials and Methods

Study Area

The experiment was carried out at the Nursery Site of the Moist Forest Research Station (6^0 32′N $5.^0$ 58′E), Forestry Research Institute of Nigeria (FRIN) Benin City, Edo State, Nigeria. Benin City is within the tropical rainforest ecological zone of Nigeria.

Soil Sampling

Top soil (0 – 15cm) was collected from the Forest Floor of Acacia Plantation of the Research Station. The soil was thoroughly mixed and passed through a 2mm sieve to remove the non-soil particulate. The chemical and physical properties of the soil including heavy metal analysis were determined prior to introduction of the *J. curcas*. Contamination was done at 3 levels (0%, 3% and 6% w/w) of spent oil in 6kg top soil and two classes of soil were used based on amendment with organic manure (amended and unamended soils). In both cases, the soil was thoroughly homogenized.

Soil Preparation and Planting

Six kilogramme (6kg) polythene pots were utilized for the experiment. Experimental Design was 2 x 3 Factorial in a Randomized Complete Block Design (RCBD) and replicated 3 times. The first factor was soil amendment (amended and unamended soil) and the second factor was three levels of spent oil pollution (0%, 3% and 6% weight by weight (w/w)). Ten seedlings were used for each level of pollution, translating to 30 seedlings per replicate and a total of 180 seedlings (90 seedlings for amended soil and 90 seedlings for unamended soil) for the experimental setup.

The pots were filled with topsoil, thoroughly mixed for even distribution of introduced organic manure (10% w/w) and spent oil contaminants and watered to field capacity. Earlier raised seedlings of *J.*

curcas were transplanted to the polluted soils at 3 weeks after planting. The investigation was carried out for 12 weeks.

Based on the setup, the treatment combinations used in this study were:

Unamended (w/w)

0% level of contamination

6kg topsoil+0 kg spent lubricating oil **(NP_0)**

3% level of contamination

6kg topsoil + 0.18kg of spent lubricating oil **(NP_3)**

6% level of contamination

6kg topsoil+ 0.36kg of spent lubricating oil **(NP_6)**

Amended (w/w)

0% level of contamination

6kg topsoil+0 kg spent lubricating oil + 0.6 kg Organic Manure **(AP_0)**

3% level of contamination

6kg topsoil+ 0.18kg of spent lubricating oil + 0.6 kg Organic Manure (**AP_3**)

6% level of contamination

6kg topsoil+ 0.36kg of spent lubricating oil + 0.6 kg Organic Manure **(AP_6)**

Laboratory Analysis

Soil and organic waste Nitrogen content was determined using the Kjeldahl Method. Phosphorus and carbon contents were determined using ICP-OES and Furnace Methods respectively. Soil pH was determined using a Glass Electrode pH metre (Rent Model 720) in distilled water (A.O.A.C. 2003).

Heavy metals determination was done by the Atomic Absorption Spectrophotometry (AAS) after digestion in aqua regia (1:3 of HCl - HNO3) (A.O.A.C 2003). Total Petroleum Hydrocarbon (TPH) analysis was done using the method of Adesodun and Mbagwu (2008). Ten grammes (10g) of soil was weighed into 50ml flask and 20ml Toluene (Analar Grade) was added. After shaking for 30minutes on an Orbital Shaker, the liquid phase of the extract was measured at 420 nanometre (nm) using DR/4000 spectrophotometer. The TPH in the soil was estimated with reference to a standard curve derived from fresh used engine oil diluted with toluene.

3. Results

Physicochemical Parameters of Soil Used for Phytoremediation

The physicochemical properties of the soil and organic manure are presented in Table 1. The soil was predominantly sandy with a pH of 6.85.

Total petroleum hydrocarbon (TPH) variation in Phytoremediated Soil

The net percentage loss of TPH of phytoremediated soil (Table 2) reveals higher percentage reduction in TPH at 0, 3 and 6% levels of contamination for amended soil as compared to non-amended soil (29.84, 44.02, 32.52; 12.89, 34.59, 23.20%) at the end of 28 days of remediation.

The same trend existed up till the 84th day of remediation; higher percentage reductions were recorded in amended soil as against non-amended soil (31.14, 33.39, 15.02; 19.93, 29.90, 61.81 13.08%). Fig. 2 gives a graphical representation of the total percentage reduction of TPH, for the two soil types at three levels of pollution by the end of the study. Percentage reduction was higher in amended soil (73.49, 91.54, 85.01%) as compared to non-amended soil (63.88, 84.81, 73.77%) at 0, 3 and 6% levels of pollution respectively.

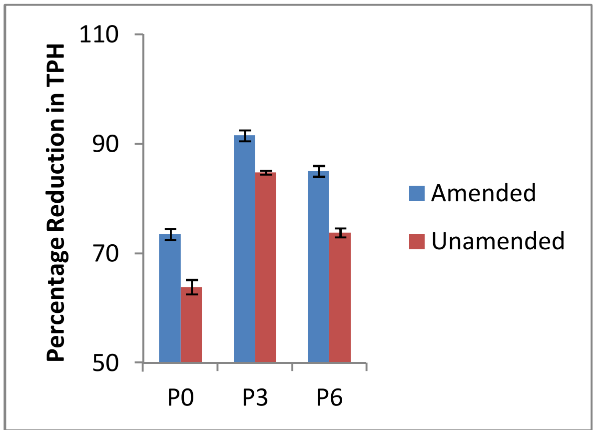

Figure 2: Percentage Reduction of TPH of Phytoremediated Soil at Day 84

P_0, P_3 and P_6 – 0,3 and 6% Pollution Levels

Table 1: Physicochemical Properties of Soil and Organic Manure Used for Phytoremediation at FRIN, 2013

PARAMETERS	ORGANIC MANURE	SOIL
Organic Carbon (g/kg)	36.38	0.93
% Sand	6.26	91.84
% Silt	0.00	4.67
% Clay	0.00	3.49
Ph	5.78	6.85
N (g/kg)	2.13	0.12
Ca (Cmol/kg)	1.18	0.008
Mg (Cmol/kg)	0.67	0.003
K (Cmol/kg)	1.03	0.011
Na (Cmol/kg)	0.49	0.006
Pb (mg/kg)	2.37	2.21
Cd (mg/kg)	0.18	0.43
Fe (mg/kg)	186.30	6.20

Heavy Metal Variation of Phytoremediated Soil

The changes in heavy metal concentration as a result of phytoremediation are represented graphically in Fig 3. It was observed that heavy metal concentrations in organic manure amended soil were higher than the unamended soil. There was a general decline in concentration of heavy metals for both soil types. The mean percentage reduction followed the order Pb>Cd>Fe respectively across the 3 levels of pollution for the two soil categories (Table 3).

Table 2: Net Percentage Loss of Total Petroleum Hydrocarbon (TPH) in Phytoremediated Soil

Treatments	TPH% Loss		
	Day28(4 weeks)	Day 56(8 weeks)	Day 84(12 weeks)
NP_0	12.89 ±1.635	47.35 ±7.400	19.93 ± 9.815
NP_3	34.59 ±2.370	66.82 ±1.185	29.90 ± 1.510
NP_6	23.20 ±0.460	63.56 ±1.875	61.81 ± 13.735
AP_0	29.84 ±0 .845	45.12 ±1.055	31.14 ± 0.270
AP_3	44.02 ±3.810	77.18 ±1.825	33.39 ± 0.010
AP_6	32.52 ±3.195	73.83 ±0.525	15.02 ± 0.525

TPH – Total Petroleum Hydrocarbon; N - Unamended Soil;
A - Amended Soil; P_0, P_3 and P_6 – 0,3 and 6% Pollution Levels

Table 3: Percentage Reduction in Heavy Metal Contents of Contaminated Soil at 12 Weeks

SAMPLE	Pb(%)	Cd (%)	Fe(%)
NP_0	51.48±0.430	39.32±7.74	20.45±1.900
NP_3	51.97±0.390	29.77±1.685	9.38±1.210
NP_6	25.76±0.050	28.66±3.060	8.91±0.130
AP_0	28.17±0.250	32.85±2.450	9.43±0.100
AP_3	22.80±0.515	20.70±0.800	8.05±0.460
AP_6	31.59±0.375	9.93±1.080	7.10±0.495

N - Unamended Soil
A - Amended Soil
P_0, P_3 and P_6 – 0,3 and 6% Pollution Levels

Lead (Pb)

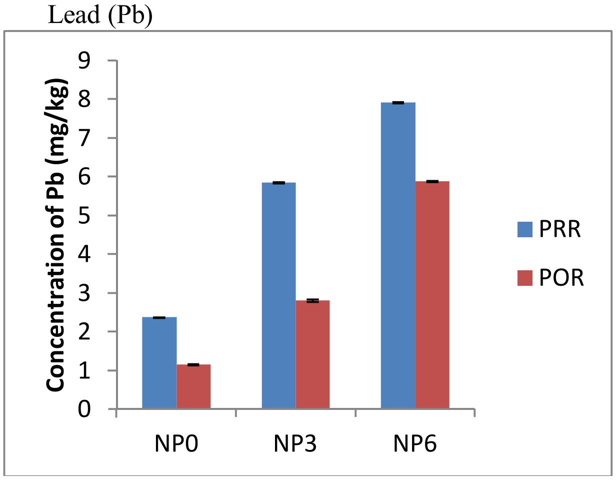

Figure 3a

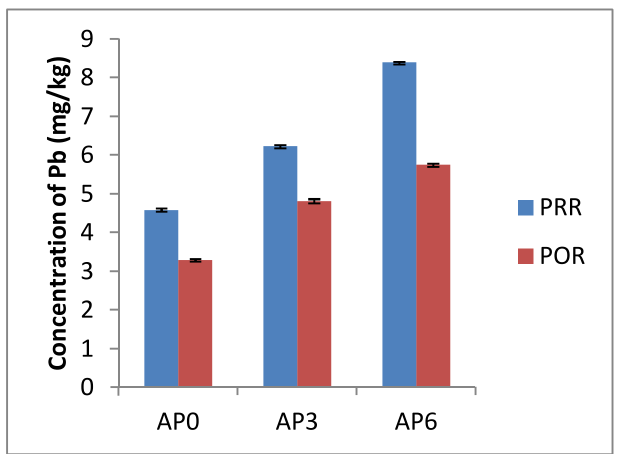

Figure 3b

Iron (Fe)

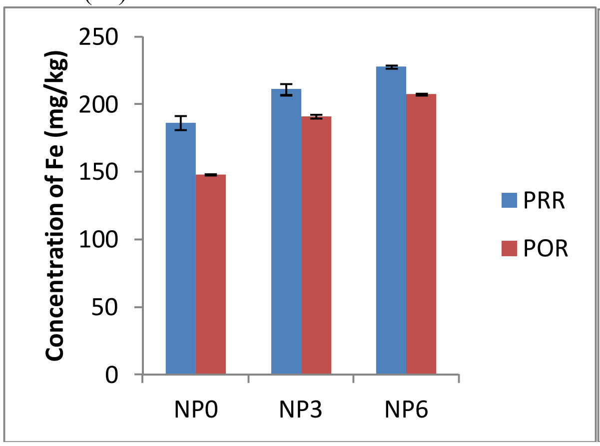

Figure 3c

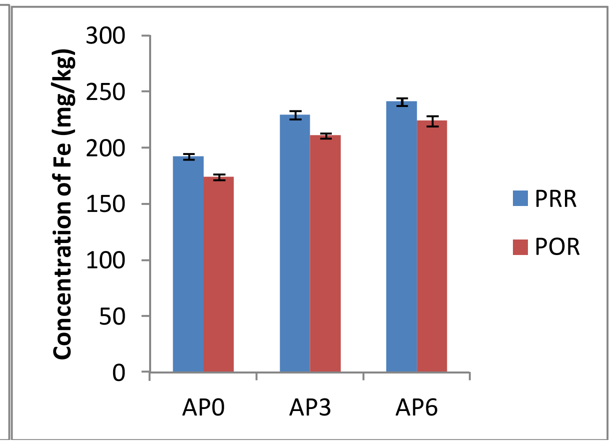

Figure 3d

Cadmium (Cd)

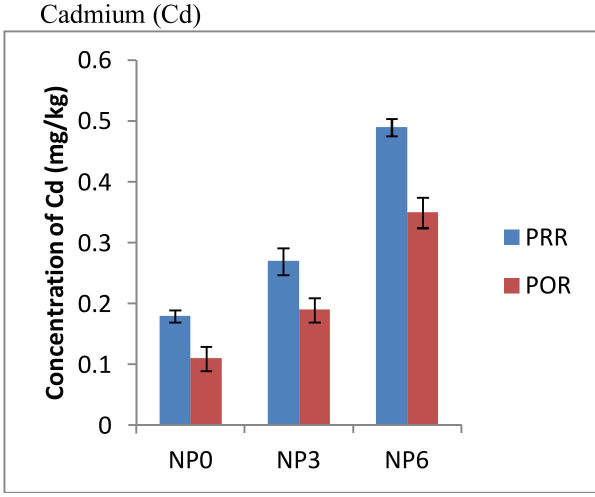

Figure 3e

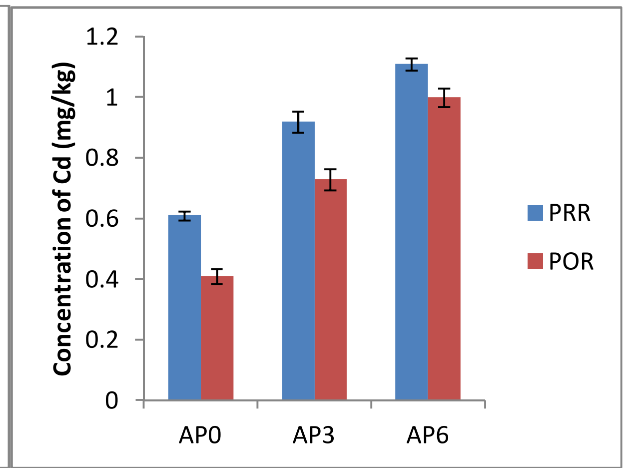

Figure 3f

Figures 3a-3f: Lead, Iron and Cadmium Concentration in SLO Polluted Soil for Amended and Unamended Soil before and after Phytoremediation.

N - Unamended Soil; A - Amended Soil; P_0, P_3 and P_6 – 0,3 and 6% Pollution Levels; PRR- Pre-remediated concentration; POR- Post remediated concentration

4. Discussion and Conclusion

This study revealed after 12 weeks of phytoremediation, a general minimal reduction in the concentrations of heavy metals in the order (Pb>Cd>Fe). The least percentage reduction in concentration was observed in Fe which interestingly had the highest concentration (186.30 – 241.30 mg/kg).

The effectiveness of *Jatropha curcas* in the removal of heavy metals from polluted soil has been well studied. Jamil *et al.,* (2009) reported, in their work, that *J. curcas* especially when supplied with basic plant nutrients, can be very useful in accumulating heavy metals such as Cu, Mn, Cr, and Fe. Mangkoedihardjo and Surahmaida (2008) stated that *J. curcas* could be used in the phytoremediation of heavy metals especially if the initial maximum concentration is not more that 50mg/kg. This might be the reason why, in this study, percentage reduction in Fe concentration was least after phytoremediation since its concentration far exceeded 50mg/kg.

Other studies on the applicability of *J. curcas* in phytoremediation include that of Surendra and Rana, (2010) who investigated the translocation and tolerance of *J.curcas* to Fe and concluded that the species is suitable for phytoremediation of Fe-contaminated wasteland soils. Also, Mathiyazhagan and Natarajan (2013) investigated the use of *Vigna unguiculata, Vigna radiate, Jatropha curcas* and *Oryza sativa* in the phytoremediation of Cd, Pb, Zn, Cr, Mn and Fe polluted waste dump site and concluded that *J. curcas* compared to others showed reasonable uptake of the elements. More importantly, Chehregani and Behrouz (2007) explained that the plant being a species of *Euphorbiaceae* family could be effective in removing Pb and Cd as demonstrated by *Euphorbia cheirandenia.* This might explain why Pb and Cd had the highest reduction in concentration after phytoremediation in this study.

This study also examined the biodegradation of the organic component of the SLO in the form of Total Petroleum Hydrocarbon (TPH). From the results, there was reduction in TPH within the first 28 days of phytoremediation. Organic amendment enhanced the rate of degradation. This is in line with the submission of Winnike-McMillan *et al.,* (2003) that organic matter enhances the bioavailability of hydrophobic organic pollutants in hydrocarbon polluted soils and also in accordance with Abioye *et al.,* (2009) who reported enhanced biodegradation of SLO in polluted soil with the use of organic wastes; Banana Skin, Spent Mushroom Compost and Brewery Spent Grain. In the same vein, Agamuthu, Abioye and Azeez, (2010) utilized J. *curcas* in the phytoremediation of SLO contaminated soil at 1 and 2.5% levels of contamination. It was reported that the addition of organic waste to *J. curcas,* remediated the soil rapidly by enhancing the removal of SLO from the contaminated soil. The enhanced TPH degradation in the phytoremediated soil could be due to increased activities of rhizospheric bacteria within the root zone of the *J. curcas* plants. Plants can enhance biodegradation of organic pollutants by microbes in their rhizosphere through the process of phytostimulation or rhizodegradation according to Nwoko *et al*., (2007).

In conclusion, this study has shown that *J. curcas* seedlings together with organic soil amendment is a veritable tool for phytoremediation of SLO polluted soils as well as phytoextraction of Pb, Cd, Fe in SLO polluted soils.

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