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Research Article

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Bioremediation of Petroleum Refinery Oily Sludge in Topical Soil

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Abstract

An enhanced applied bioremediation project was carried out with the aim to establish a pilot plan for preparation of a future large scale ex-situ bioremediation of refinery oily sludge in a tropical soil in West Africa. Prior to this experiment, primary feasibility studies were conducted by developing a consortium of indigenous oil degrading micro-organisms, by producing a local oil dispersant named CNN_{2000} (produced by recycling agricultural waste) and establishing that chemical fertilizer NPK 20:10:10 used at 2400 kg/ha will be the appropriate amount to be applied to polluted soil. For the experiment proper, four hundred (400) litres of waste refinery oil were emulsified with CNN_{2000} and sprayed in a 100 m^2 prepared plot, the microorganisms' consortium and fertilizer were applied and the polluted plot was watered, tilled frequently and observed for six months. The treatment resulted in 94% removal of total petroleum hydrocarbon (TPH) in six months compare to 19% removal of TPH in control plot. Chromatographic analysis revealed that the alkane and aromatic fractions of TPH reduced by 95% and 92%, respectively, while the NSO fraction instead increased. The population of introduced consortium remained stable in the treated plot even after six months. The physical and chemical properties of the treated soil improved during the study period. The practical outcome from this project has revealed that, 40,000 litres of waste refinery oil could be treated in 1 hectare plot (10,000 m^2) at a cost estimate of approximately US 3.20 dollars per m^2. This cost could be reduced to about US 1.90 dollar per m^2 during subsequent treatments. This project has enabled us to finally establish a cost effective local baseline technique for efficient bioremediation of waste refinery oil in tropical soil.

Keywords: Oily sludge; Emulsifier; Enhanced applied bioremediation; Land farming remediation; Petroleum hydrocarbon; Tropical soil

Introduction

Refinery processing of crude oil generates oily sludge as waste end products which are poorly biodegradable constituting environmental problems with substantial implication for economic development and human health. Oily sludge is generated in the refinery principally from periodic cleaning of crude oil and refinery products storage tanks. One of the major problems faced by oil refinery is the safe disposal of oily sludge in the environment; this is because many of the constituent of oily sludge are carcinogenic and potent immuno-toxicants [1]. A number of methods exist that are used to decontaminate the affected sites. Amongst these methods, most refineries often adopt bioremediation. Briefly, bioremediation is an ecological sound and state-of-the art technique that employs natural ecological processes to completely eliminate toxic contaminants. The technique makes use of living organisms (bacteria, fungi, some algae, and even plant) to reduce or eliminate toxic pollutants. These organisms may be either naturally occurring or laboratory cultivated and they either eat up the contaminant (e.g. organic compounds) or assimilate within them all harmful compounds (e.g. heavy metals) from the surrounding, thereby rendering the region virtually contaminant free. The process uses relatively low cost, low-technology technique which generally has a high public acceptance and can be carried out on site (in-situ) or off site (ex-situ). Bioremediation could be enhanced with the use of fertilizers, compost, bulking agent and some chemicals including oil dispersant. With the addition of enhancing agents that support microbes and increase the availability of the substrate, clean up proceeds much more rapidly [1-4].

Bioremediation of waste oil in soil (land farming) also called land spreading, had been extensively carried out in different parts of the world for almost 4 decades now. Early studies were published in USA by Dotson et al. [5] and Kincannon [6]. By mid 70's other authors reported on oily sludge land farming including Salome (1974) in UK, and Fusey [7] in USA. In the late 70's, further studies on bioremediation by land farming were reported by a number of authors namely Grove [8] in UK; Dibble and Bartha [9] in South Africa; and Weldom in USA. Intensive studies have been carried out in the 80's as well as the 90's to include Gomez-Bordonado and Almagro-Huertas in Spain; Bossert et al. [10] in USA; Shalubhai et al. [11] in the Middle East; Todd et al. [12] in USA; Rosenberg et al. [13] in Israel; Prado-Jatar et al. [14] in Venezuela; Genouw et al. [15] in Belgium; and Adams and Jackson in Nigeria. In this last decade, improve studies on the traditional methods and development of molecular environmental genetics to give a more comprehensive understanding on the process have been catalogued and this include reports from Mishra et al. [1] in India, Zucchi et al. [16] in Italy, Christopher and Kitts [17] in USA; and Marc et al. [18] in Spain to name some few.

Draw back from the above development indicates that although bioremediation has been cited in Africa, much need to be done. Bioremediation in Sub-Saharan Africa especially in the oil-rich – Gulf of Guinea, is at its pioneering stage. Presently few reports on bioremediation exist in the region. But petroleum industry established in the region some 45 years ago and the devastating effects as a result of these activities had led recently to a tripartite conflict in part of the region opposing the - population-oil companies-and the respective government. A recent example is that of the Ogoni Region in the South Eastern (Niger Delta Gulf of Guinea) in Nigeria, where the Ogonis' armed militia opposed government's armed forced. These militia were requesting the government to summon the oil companies in the region for environmental cleanup and compensation. Lack of safe disposal of oily waste contributed partly in the conflict. In anticipation to such crisis, we therefore found it necessary to investigate through experimental research, the possibility to develop a cost effective local baseline technique for efficient bioremediation of waste refinery oil in our environment, so as to cushion the effect of such crisis on

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the environment and on people. Prior to this experiment, primary feasibility studies were conducted by -a) determining the optimal condition for microbial seeding and nutrient amendment, and by -b) producing waste oil emulsion, using locally produced oil dispersant. This research is presented here as a pilot plant in preparation for a future large scale ex-situ bioremediation of refinery oily sludge in our soil environment.

Materials and Methods

Study area

This research was carried out in Buea, Cameroon, a country situated in the oil rich Gulf of Guinea of Africa. The city of Buea is situated at some 800 m altitude above sea level in the flank of Mount Cameroon (4100 m).

Collection of materials

Oily Sludge: The oily sludge was obtained from an open- to the- air primary storage basin of the activated sludge treatment plant of the country refinery (SO.NA.RA Refinery, Limbe, Cameroon) situated some 40 km (24 miles) away from Buea city. Oil dispersant/emulsifier used to emulsify oily sludge was the dispersant CNN_{2000} formulated by recycling tropical agricultural waste [19] dispersant CNN_{2000} was tested for its toxicity potential and was found to be environmental friendly. Fertilizer NPK 20:10:10 was purchased from a local agricultural shop. The consortium of microorganisms used for microbial seeding was that described by Nkeng et al. [20].

Preparation of experimental plots

The experimentation was carried out in a planting land covered with dense vegetation. An area of 37 m x 37 m (1,369 m^2) was cleared to prepare the plots. Three plots of 10 m x 10 m (100 m^2) were prepared. One of these plots was set apart for pilot plot and the second tagged Control plot A (to receive neat oily sludge) and the third control plot B (non-polluted control plot). The distance between consecutive plots was 1.5 meter.

Treatment of pilot plot

In this plot 400 litres of waste refinery oil was emulsified with dispersant CNN_{2000} (mixing ratio was 1 g oil / 1.5 ml dispersant) and sprayed on the 100 m^2 demarcated zone. The consortium of micro-organisms for seeding oily sludge polluted soil was added at predetermined amount; the organisms were Bacillus subtilis, Aspergillus sp, and Penicillium sp; they were harvested from stock cultures, and primary feasibility studies demonstrated that, the following inoculum's concentrations that could produce excellent results would average: 32 x 10^8 CFU/ml for Bacillus subtilis, 23 x 10^6 CFU/ml for Aspergillus sp and 29 x 10^6 CFU/ml for Penicillium sp. The dilutions were prepared and pooled in a 20 litres volume and sprayed in every 25 m^2 area. Fertilizer NPK 20:10:10 was applied once every week for one month, then once every two weeks for 2nd and 3rd months, the concentration was reduced by 50 % in the 4th and 5th months and by 70% in the 6th month. Fertilizer was applied at 2400 kg/ha as indicated by the feasibility studies.

Treatment of control plots

Control plot A was polluted with neat oily sludge, while control plot B was left free from pollution.

Soil sampling

On the pilot plot, 10 random spots 1m apart to a depth of 30 cm were augured and pooled together (composite soil samples) monthly and transferred into sterile polythene bags and labeled. In addition 5 random spots were collected at the 60 cm at the beginning and at the end of experiment to determine the extent of seepage of oily sludge in soil. On the control polluted plot A, 4 random samples were collected monthly and pooled to obtain composite samples. All samples were sent for analysis in the laboratory immediately after collection.

Processing of the specimen

The extraction of Total Petroleum Hydrocarbon (TPH) from oily sludge contaminated soil, was carried out according to the method described by Mishra et al. [1]. The fractionation of TPH using chromatographic analysis was performed as described by Mishra et al. [1]. The soil physicochemical properties were carried out as described elsewhere [21] and the heavy metals were determined using the method described by Sengar et al. [22].

Results

The composition of oily sludge used in this study is presented in Table 1. The sludge has a high solid content (74%). The TPH content was 19%. Among the 3 fractions of TPH (alkane, aromatic and NSO), the alkane fraction was present in the highest proportion.

The physico-chemical characteristics at the remediation site were presented in Table 2. The pH value did not vary much in control plot A, but an increase was observed in pilot plot. The total nitrogen and available phosphorus and potassium content also increased in pilot plot while a decrease in organic carbon was observed.

constituents	Concentration (% wt/wt)a
Oily sludge	
Oily content (TPH)	19.0 ± 0.10
Solid content	74.1 ± 0.34
Water content	06.9 ± 0.28
TPH	
Alkane fraction	57.6 ± 0.1
Aromatic fraction	31.0 ± 0.3
NSO fraction	11.4 ± 0.1
Asphaltene	ND

ND = Not Determined
[a]means standard deviation for 3 samples

Table 1: Composition of oily sludge collected from the refinery.

Parameters	Unpolluted Plot	Pilot plot (Treated polluted soil)		Control plotA (Neat oily sludge)	
		Day zero	180 days (6 months)	Day zero	180 days (6 months)
pH	4.7 ± 0.04	5.9 ± 0.06	7.4 ± 0.04	4.6 ± 0.2	4.8 ± 0.1
Moisture content (%)	16.2 ± 2	14.1± 3.0	12.2 ± 4	19.1 ± 1	14.3 ± 3
Organic carbon (%)	3.1± 0.04	3.6 ± 0.02	2.2 ± 0.03	3.4 ± 0.01	3.2 ± 0.3
Total nitrogen (%)	0.12 ± 0.05	0.137± 0.07	1.24 ± 0.02	0.071± 0.001	0.066± 0.01
Availalable Phosphorus (mg/kg)	20 ± 0.014	31.6 ± 0.12	43.2 ± 0.18	20.0 ± 0.015	18. ± 0.01
Potassium (meq / kg)	0.015 ± 0.03	0.021± 0.01	0.032±0.02	0.018 ± 0.03	0.014± 0.02

[a]soil samples were collected at 30 cm depth

Table 2: Physical and chemical composition of soila at bioremediation site.

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Heavy metals analysis revealed 5 elements in the oily sludge. These elements were also estimated at the bioremediation site. The results indicated that, despite the high value of lead (Pb), the concentrations of these metals in the oily sludge and in the treated plot were much lower according to standard and these concentrations were not toxic to most soil organisms. The seepage of heavy metals in the soil of the bioremediation site was also monitored.

The biodegradation rate of oily sludge is presented in Figure 1. A degradation rate of 94% of TPH was recorded in the pilot plot as against 14% in the control plot A after six months experimental period. Figure 2 indicates the concentrations of TPH fractions in the soil samples. The alkane and the aromatic fractions reduced by 95% and 92%, respectively, while the concentration of NSO instead increased.

Discussion

The aim of this study was to initiate a pilot plot for the biodegradation of oily sludge using the enhanced applied bioremediation technique by land farming. Our results revealed the reduction of TPH was much higher (94%) in the pilot plot than in the control plot (14%). This pattern of waste oil degradation have been reported by a number of authors including Rosenberg et al. [13] Prado-Jatar et al. [14] Adams and Jackson [23] Mishra et al. [1] Zucchi et al. [16] Marc et al. [18].

The efficiency of the biodegradation of oily sludge was also ascertained based on the results of the changes in the physical and chemical properties of the polluted test plots. Changes were either increased or decrease in the mean values of the soil parameters (Tables 2 and 3). The pH value of the pilot plot increased from 4.9 to 7.5. This result indicating increase in the pH value in pilot plot is contrary to the reports by some authors who observed instead a decrease in pH due to the production of organic acids in the medium and also to the application of chemical fertilizer in test plots that help to produce higher concentrations of organic acids than the unfertilized soil [6,24]. The increase in pH value in this study could be as result of the treatment of the oily sludge with emulsifier CNN_{2000} whose pH value (11.4) is

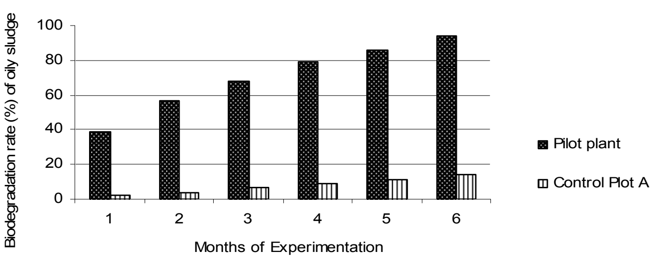

Figure 1: Biodegradation rate (%) of oily sludge in soil.

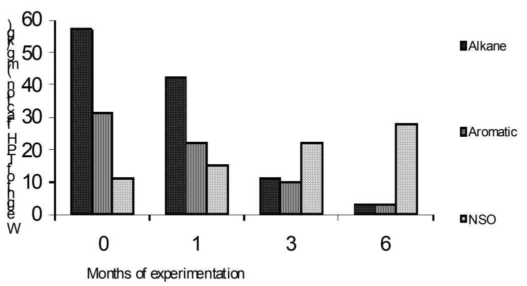

Figure 2: Concentration of TPH fraction in pilot plot.

Heavy metals	Oily sludge	Pilot plot (Day zero)		Pilot plot (180 days)	
		30cm	60cm	30 cm	60cm
Cd	0.013± 0.00	0.018± 0.01	0.022 ± 0.01	0.028 ± 0.00	0.010 ± 0.01
Cr	0.035 ± 0.00	0.039 ± 0.00	0.044 ± 0.00	0.049 ± 0.01	0.028 ± 0.00
As	0.005 ± 0.00	0.007 ± 0.00	0.009 ± 0.00	0.004 ± 0.00	0.002 ± 0.00
Pb	36.4 ± 0.15	39.6 ± 0.2	35.1± 0.3	49.6 ± 0.2	14.3 ± 0.00
Ni	0.042 ± 0.10	0.044 ± 0.00	0.032 ± 0.01	0.056 ± 0.00	0.019 ± 0.00

[a]values are means standard deviations for 3 samples

Table 3: Concentration of some heavy metalsa (mg/kg) in oily sludge and at bio-remediation site.

known to be strongly alkaline [19]. The contact of the emulsified oily sludge (alkaline pH) with soil (acidic pH) brought about a buffering effect such that the pH of the treated plot increased and stabilizes at about pH 7.5 (Table 2). This modification in the pH of the soil will favor the micro-organisms growth such as that of bacteria [10]. These findings inferred that emulsifier CNN_{2000} use to produce oil emulsion offered an additional advantage in that its presence increases the pH of the soil to a value that favor microbial growth and therefore limiting the use of liming to reduce soil acidity before bioremediation in acidic soil environment.

The total nitrogen and available phosphorus of the treated soil increased during the incubation period. These results agreed with findings of Mishra et al. [1], Odokuma and Dickson [21], Amadi et al. [24]. However the concentration of available phosphorus was very high in the experimental soil as compare to other soils where value of 2 mg/kg is found. The soil used in this study was a near to the coast soil and Donahue et al. [25] reported that, high values of available phosphorus are common in soils derived from coastal plain. The increase in nitrogen and phosphorus contents in pilot plot might probably be attributed to the nitrogenous and phosphorus bearing fertilizer NPK applied frequently in soil. The main value of potassium increased slightly in pilot plot. Potassium element is involved in the alkalinity of the soil. The alkaline nature of the emulsifier CNN_{2000} composed of KOH [19] could have affected the soil such that the exchangeable base K+ increases in value. Organic carbon in soil was reduced from 3.6% to 2.2% at the end of the study in pilot plot which indicates that TPH content of oily sludge had been lowered. We also observed that, the concentration of heavy metals in pilot plot (30 cm horizon) at the end of the study was much higher than in soil at time zero due to addition of heavy metals in the surface of the soil from the oily sludge after biodegradation of TPH. However the concentration of the heavy metals at 60 cm horizon decreased at the end of the study, and this probably established that degradation of TPH in soil did not result in leaching heavy metals in soil.

Chromatographic analysis of the TPH fractions in pilot plot indicates that, the alkane and the aromatic fractions decreased while the NSO increased indicating a conversion of alkane and/or aromatics to NSO fraction. The alkane degraded most readily (95%) than the aromatics (92%). These results agreed with the works by Mishra et al. [1], Shalubhai [11] who reported that susceptibility of to biodegradation of various oil fractions decreased in order from alkane> aromatic> asphaltic.

Soil was also sampled at the end of experiment and investigated for microbial community. The microorganisms of the consortium were isolated in the range of 10^5 to 10^7 CFU/g indicating a stable growth in the pilot plot. Vegetation appeared in the pilot plot at the end of 3rd month. This study was carried out in 2007, after termination of the project we purposely left the experimental area untouched for a four year observation. At the end of each year, vegetation density increase; and by 2011, the pilot plot was covered with very dense vegetation.

The cost evaluation of the study was also considered. As with any demonstration trial, the costs associated with undertaking a trial are far higher than a full scale commercial operation due to economies achieved through scale CL:AIRE [25]. Nevertheless an estimation of cost was made using the following assumptions: a) the volume of the materials (oily sludge) for treatment in 1 ha (10,000 m^2) would be 40,000 litres, and b) costs for excavation (collection), haulage, welfare and security are excluded. Based on the cost for the trial and these assumptions, the treatment cost would be US $ 3.20 per m^2. Exclusion of any trial costs incurred such as, transportation of liquid emulsifier to treatment site, additional analytical costs, literature search and reporting would lower the costs so that cost of US $ 2.7 per m^2 could be anticipated. If the extraction/production of the emulsifier (CNN_{2000}) could be envisaged at the site of the experimentation, the cost will be lowered to approximately US $ 1.9 per m^2.

Conclusion

The study has revealed that, the simultaneous application of consortium of organisms, chemical fertilizer, and an alkaline emulsifier in soil polluted with refinery oily sludge enhances the biodegradation of TPH. Consequently, the enhanced applied bioremediation of waste oil by land farming is a method that could be considered to initiate the treatment of the refinery oily sludge in our soil environment.

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