Treatment of Spilled Auto-Mechanic Garage Oils in Soil Using Polyethylene Terephthalate (PET) Waste Materials

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Abstract: The work aimed at cleaning oil contaminated soil started with sorting and pulverizing pre-clean high density polyethylene terephthalate (PET) waste materials. The soil samples were collected from Farin-Gada Auto-Mechanic village located at Jos North Local Government area, Plateau State-Nigeria. They were prepared by homogenizing, crushing and sieving (mesh size ≤2mm), then extracted and purified for characterization of the total petroleum hydrocarbon (TPH) using gas chromatography mass spectroscopy (GCMS). For the heavy metals, the soil samples were air-dried, digested using mineral acids (HNO₃ and HCl) in appropriate proportion, then analyzed using atomic absorption spectrophotometer (AAS). The levels of nickel (Ni), lead (Pb), chromium (Cr), cadmium (Cd) and TPH were assessed in the sample used as control (uncontaminated) labelled as ‘CC-S’. Untreated soil (automobile mechanic garage soil) labelled ‘UT-S’, and treated soil labelled as ‘TT-S’. The results revealed that the untreated soil (UT-S) had high contaminants when compared to that of CC-S and TT-S. This suggest that anthropogenic activities is the major source of the soil contamination. On treating the contaminated soil with PET, the concentration of Ni, Pb, Cr and Cd in the UT-S was reduced by 18%, 4%, 84% and 66% respectively. Similarly, the total petroleum hydrocarbon (TPH) concentration reduced from 0.271 mg kg⁻¹ to 0.082 mg kg⁻¹ on treating UT-S with PET. The present work has established that pulverized waste polyethylene terephthalate (PET) can be processed and used to clean up oil contaminated soils.

Keywords: Treatment, Oil Contaminated Soil, Polyethylene Terephthalate (PET), Heavy Metals, Total Petroleum Hydrocarbon (TPH)

1. Introduction

Crude oil products are one of the common environmental contaminants/pollutants, and its spilling has a hazardous effect on both plants and animals living within the natural environment. However, the current rapid rate of industrialization, urbanization [1] and economic activities like mining [2], agriculture [3], industries and transportation [4, 5], can lead to contamination of environmental resources due to huge amount of waste they generate. For instance, gasoline, battery manufacture, metal plating, smelting, tanneries, petroleum refining, paint manufacture, pesticide, cosmetics, ceramics, pigment manufacture, printing and photographic industries, etc., are sources of 0 heavy metals such as cadmium, zinc, copper, nickel, lead, mercury, cobalt, manganese and chromium [6].

Also, spills from used automobile oils, worn machinery, used batteries, organic and inorganic chemicals used in oil as additives also release different heavy metals [7], and these heavy metals enter the environment through bio-magnifications in food chains [8]. Once these toxic substances penetrate into the ground, they have the capacity to pose a wide range of adverse environmental problems [9, 10]. For instance, a study [11] confirmed that garage and automotive workers are in danger of impending lead toxicity and facing abdominal colic, constipation, and central nervous system dysfunction. Other studies [12, 13] further confirmed that plants and leafy vegetables grown using wastewater and
polluted soil can accumulate toxic heavy metals above maximum limit [14], causing serious risk to human health when consumed [15].

In recent times, Shashemane City is characterized as one of the fastest growing cities and center of business in Oromia region. This will lead to rapid increase in automobile garage villages, and the implication of this is releases of different types of used lubricants and oil related pollutants that have significant potential impact into the environmental sectors [16] undertaking different operational activities. Mindful of the impact of activities in garages, an investigation [11] on impact of exposure to lead (Pb) by garage workers which relates to a similar study [13] in which it was discovered the effects of oil spills are not limited to the immediate environment alone. The immediate effects on humans, fish, animals, birds and wildlife in general, mainly are due to; direct contact with the spilled oil including breathing of volatilized oil components from the spillage, direct contact with the environment polluted with spilled oil components, such as drinking polluted water or breathing polluted dust particles and consumption of polluted food at any level within the food chain, with a higher risk for food pollution at the higher levels of the food chain i.e. humans and animals. Hence, the need to clean contaminated soils, that inform the need for this work which aims to prepare as readily available waste materials to treat soil polluted by used oil and lubricants.

2. Materials and Experiments

2.1. Study Area

Jos North is located in the Northern part of Plateau State situated between Latitudes 9°56'21.7" (9.9394°N) and Longitudes 8°5'8" (8.9022°E), elevation 1,200 meter (3,937 feet). The mechanic garage village being studied is located along Farin-Gada Jos North Local Government, Plateau State, Nigeria (Figure 2). It covers a large span of land, a busy area where automobile repairs and maintenance are carried out and serves as the major mechanic workshop for the Jos metropolis and other neighboring Local Governments and States [18]. Soil exposed to petroleum hydrocarbon and heavy metal contamination due to industrials activity and transportation was collected at different depth of (0.5m and 1.0m) from the automobile repair workshops close Farin-Gada Market.

2.2. Sampling Area

The study was done by obtaining samples from Farin-Gada Auto-Mechanic Village, Jos North Local Government area in Plateau State-Nigeria. It is a densely populated area where commercials maintenance of vehicles and other automobiles are done. It is located on latitude 9°56'21.7"North and longitude 8° 54'8" East of Greenwich meridian. The soil samples site location were recorded using a Geographical Positioning System (GPS), which provides precise location of the research area [19].

2.3. Soil Samples Collection

The soil samples were collected with the aid of a stainless steel sampling auger at different depth in amber bottles, that had been properly cleaned before soaking in 10% nitric acid and rinsed with distilled water. The sampling auger was washed with distilled water each time it is used for collection of sample before using it to collect another samples. Samples were then transported to the laboratory in clean plastic coolers and stored in the chiller awaiting analysis.

2.4. Sample Treatment Using the Pulverized PET Material

A reasonable quantity of high density polyethylene terephthalate (PET) waste polymeric bottles with properties earlier identified (Table 3) [20] were collected and clean before pulverizing into powder (mesh size ≤2.00mm) using the Cryo-milling technique under specified conditions [17]. It was also tested to ascertain the safety usage of the PET rubber, and to ensure that all PET rubber used does not serve as a contaminating agent other than the purpose it was employed for [21]. 50g of the polluted soil sample and 10g of the pulverized material (PET powder) was measured into 200ml of distilled water in a 500ml beaker. On stirring, the processed PET absorbed the oil contaminants which are hydrocarbon in nature, then scooped. A portion of the soil left was dried and digested for heavy metals analysis, while the second portion was used for Total Petroleum Hydrocarbon (TPH) analysis. The material used in this research work was polymeric materials that had been properly cleaned by soaking in 3% nitric acid, thereafter rinsed with distilled water.

2.5. Heavy Metals Analysis

The dried soil samples were ground and sieved through ≤ 2mm mesh-sized sieve mesh. 1.00g of each of the sieved sample was weighed into an Erlenmeyer flask, and 100ml of
The wet soil sample was air-dried and homogenized, after which the sample was crushed and filtered. 10.00g portion of the sample was weighed into the brown laboratory amber bottles. Thereafter 10ml of a mixture of Dichloromethane (DCM) and Acetone in the ratio of 1:1 added to the sample. A known concentration of O-terphenyl standard was also measured into the sample to help spike the samples. The mixture were further agitated using the vortex mixer for 20 minutes. This was then filtered using whatmann filter papers on a reasonable amount of anhydrous sodium sulphate (Na\textsubscript{2}SO\textsubscript{4}). The samples were then collected and further filter through a micro column parked with cotton wool and silica gel powder (60-120 mesh size). Anhydrous sodium sulphate (Na\textsubscript{2}SO\textsubscript{4}) was further added on the silica gel inside the column as drying agent for the polluted area, which represent untreated soil (automobile mechanic garage soil) labelled ‘UT-S’ had the concentration of 0.025mg kg\textsuperscript{-1} for Ni, 0.090-0.206 mg kg\textsuperscript{-1} for Pb, 0.090-1.280 mg kg\textsuperscript{-1} for Cr and 0.002-0.009 mg kg\textsuperscript{-1} for Cd as shown in Table 1. The relatively unpolluted soil (from a conserved area within University of Jos) representing the ‘Control sample’ showed a concentration of 0.025mg kg\textsuperscript{-1} 0.080mg kg\textsuperscript{-1} 0.090 mg kg\textsuperscript{-1}, and 0.002 mg kg\textsuperscript{-1} for Ni, Pb, Cr and Cd respectively. While for the polluted area, which represent untreated soil (automobile mechanic garage soil) labelled ‘UT-S’ had the concentration of Ni, Pb, Cr and Cd to be 0.039±0.01, 0.206±0.03, 1.280±0.14 and 0.009±0.06 mg kg\textsuperscript{-1} respectively. On treating the soil, the concentrations of all these metals reduced significantly. From this, the concentrations of Ni and Pb reduced by 18% and 4.00% respectively. Higher reduction was observed in Cr with 84% and Cd with 66% (Table 1 & Figure 3). This suggest the treatment method used in this research is effective and can even be adopted at a larger scale to treat similar oil contaminated soils.

### 3. Result and Discussion

#### 3.1. Heavy Metals And Total Petroleum Hydrocarbon Concentrations

The Control (CC-S), Treated (TT-S) and Untreated (UT-S) soil samples under investigation revealed varying concentrations of all the four heavy metals (Ni, Pb, Cr, and Cd) analyzed. The concentrations were in the range of 0.025-0.039 mg kg\textsuperscript{-1} for Ni, 0.080-0.206 mg kg\textsuperscript{-1} for Pb, 0.090-1.280 mg kg\textsuperscript{-1} for Cr and 0.002-0.009 mg kg\textsuperscript{-1} for Cd. This research employed a treatment approach that used high workshops with 362.60 ± 185.84 and 428.55 ± 119.00 mg kg\textsuperscript{-1} in related activities. For instance, and earlier work [23] reported contamination widely reported in other places and sites involved TPH concentrations at Petrol Stations had minimum of 399.83 ± 106.19 and maximum of 450.83 ± 90.58 mg kg\textsuperscript{-1} and Mechanic workshops with 362.60 ± 185.84 and 428.55 ± 119.00 mg kg\textsuperscript{-1}. This research employed a treatment approach that used high density PET to treat the contaminated soil (UT-S) from which the treated sample (TT-S) significantly recorded a low concentration of 0.081 mg kg\textsuperscript{-1} (Table 2 and Figure 5). The Control sample (CC-S) did not show any contamination before it gets out of place, even though it is slightly higher than the TPH concentration of 0.002-0.153mg kg\textsuperscript{-1} present in the control sample (CC-S). Giving the trends of contamination widely reported in other places and sites involved in related activities. For instance, and earlier work [23] reported TPH concentrations at Petrol Stations had minimum of 399.83 ± 106.19 and maximum of 450.83 ± 90.58 mg kg\textsuperscript{-1} and Mechanic workshops with 362.60 ± 185.84 and 428.55 ± 119.00 mg kg\textsuperscript{-1}. This research employed a treatment approach that used high density PET to treat the contaminated soil (UT-S) from which the treated sample (TT-S) significantly recorded a low concentration of 0.081 mg kg\textsuperscript{-1}.

#### 3.2. Total Petroleum Hydrocarbon (TPH) Concentrations

From this work, the total petroleum hydrocarbon (TPH) concentration lies between 0.002-0.174 mg kg\textsuperscript{-1} in the auto-mechanic garage contaminated soil (UT-S). This is quite low when compared with range for a similar work of 486 to 4,438.7 mg kg\textsuperscript{-1} at 0–15 cm depth reported in a similar work [22]. From this, there is a ray of hope as the relatively low concentration obtained in this work will pave way to regulate the contamination before it gets out of place, even though it is slightly higher than the TPH concentration of 0.002-0.153mg kg\textsuperscript{-1} present in the control sample (CC-S). The samples were then collected and further filter through a micro column parked with cotton wool and silica gel powder (60-120 mesh size). Anhydrous sodium sulphate (Na\textsubscript{2}SO\textsubscript{4}) was further added on the silica gel inside the column as drying agent before the sample was passed through it. The purified samples were finally concentrated to 3ml before detecting them using Gas Chromatography-Mass Spectroscopy (GC-MS).

### Table 1. Heavy Metal concentration (X±SD) (mg/kg) in the Control (CC-S), Untreated (UT-S) and Treated (TT-S) Soil Samples.

| Sample Code | Ni     | Pb     | Cr     | Cd     |
|-------------|--------|--------|--------|--------|
| CC-S        | 0.025±0.04 | 0.080±0.08 | 0.090±0.06 | 0.002±0.02 |
| UT-S        | 0.039±0.01 | 0.206±0.03 | 1.280±0.14 | 0.009±0.06 |
| TT-S        | 0.032±0.04 | 0.198±0.06 | 0.199±0.17 | 0.003±0.01 |
| Quantity Removed | 0.007 | 0.008 | 1.081 | 0.006 |
| % Removed   | 18.00   | 04.00   | 084.00 | 66.00 |

**Figure 3.** Heavy Metals Concentration (mg/kg) in Control (CC-S), Untreated (UT-S) and Treated (TT-S) Soil Samples.

**Table 2.** Profile and concentration (mg/kg) of TPH in the Control (CC-S),
Untreated (UT-S) and Treated (TT-S) Soil Samples.

| Compounds                  | CC-S | UT-S | TT-S |
|----------------------------|------|------|------|
| Decane                     | 0.002| 0.004| 0.002|
| Undecane                   | 0.002| 0.001| 0.000|
| Tridecane                  | 0.001| 0.002| 0.001|
| Tetradecane                | 0.008| 0.010| 0.000|
| Pentadecane                | 0.004| 0.008| 0.002|
| Hexadecane                 | 0.001| 0.001| 0.000|
| Heptadecane                | 0.002| 0.003| 0.001|
| 2,6,10,14-tetramethyl-pentadecane | 0.001| 0.000| 0.000|
| 1,2-dimethyl-cyclooctane  | 0.153| 0.175| 0.046|
| Octadecane                 | 0.022| 0.036| 0.003|
| Nonadecane                 | 0.001| 0.007| 0.005|
| Eicosane                   | 0.001| 0.003| 0.002|
| Heneicosane                | 0.007| 0.013| 0.008|
| Hexacosane                 | 0.002| 0.008| 0.007|
| Tetracosane                | 0.001| 0.002| 0.002|
| Octacosane                 | 0.000| 0.000| 0.002|
| 9-octyl-Heptadecane        | 0.000| 0.001| 0.000|
| 7-hexyl-Docosane           | 0.000| 0.000| 0.001|

Table 3. Properties of PET used.

| Property                | Value    |
|-------------------------|----------|
| Density                 | 0.93 to 0.97 g/cm³ |
| Chemical Property       | Inert    |
| Heat Resistant          | >100°C   |
| Melting Point           | 135°C    |

Source [20].

Figure 4. TPH’s Concentration (mg/kg) in Control (CC-S), Untreated (UT-S) and Treated (TT-S) Soil Samples.

Figure 5. Summary Concentration of THP in Control (CC-S), Untreated (UT-S) & Treated (TT-S) Samples.

Figure 6. Spectra for the direct infusion of Treated (TT-S) and Untreated Soil Samples (UT-S).

4. Conclusion

Pulverized high density polyethylene terephthalate (PET) waste bottles used for cleaning oil and lubricant contaminated soil was found to be effective, as considerable quantity of pollutants like TPH were removed from the contaminated soil by 70%. While heavy metals like Cr and Cd were significantly cleaned up by 84% and 66% respectively. This goes on to give positive indication that waste PET materials instead of being environmental nuisance, can be processed to clean up these categories of pollutants in oil
polluted soils. In addition, to deal with the problem from the root, it is advisable that all automobile repairs done in different cities be restricted to mechanic villages, to enhance ease of collection, preservation, recycling and re-use of spent oils. This will put an end to indiscriminate disposal of spent automobile oils which results in excessive pollution of topsoil.

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