Short Communication

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# Assessment of Total Petroleum Hydrocarbons Content of Soils Within Estate and Works Departments of Three Universities in Port Harcourt Housing Heavy-Duty Generators

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**ABSTRACT**

Soil samples were collected at random from the Estate and Works Departments of three universities in Port Harcourt, Rivers State, Nigeria, where heavy-duty diesel generators are mounted to provide electricity for the universities environments and assessed for total petroleum hydrocarbons contamination. Soxhlet extraction method was used for the extraction of the total petroleum hydrocarbons and due clean-up of the chromatographic column followed. Total petroleum hydrocarbons were analyzed with Gas Chromatography-Flame ionization detector (GC-FID) to detect and quantify the levels in the different stations. The results obtained from the soil samples from the different universities were; IAUE, 1475.56904 mg/Kg; RSU, 953.11949 mg/Kg and UNIPORT, 968.93886 mg/Kg with an average total petroleum hydrocarbons concentration of 1132.20913 mg/Kg. Total petroleum hydrocarbons contamination levels obtained in the three stations were higher than the 50mg/Kg limit allowed by DPR for agricultural soils. Source diagnostic ratios and indices used in analyzing the total petroleum hydrocarbons contamination revealed that the pollution was majorly due to anthropogenic origin, although other sources such as terrestrial vascular plants, biogenic origin also contributed. Since the contamination level of total petroleum hydrocarbons in the soils were greater than target limit, the universities should therefore provide remediation schemes at regular intervals to forestall any danger that it might pose to the workers in the departments.

**Abbreviations:** GC-FID: Gas Chromatography-Flame Ionization Detector; UNIPORT: University of Port Harcourt; RSU: Rivers State University; DPR: Department of Petroleum Resources; EGASPIN: Environmental Guidelines and Standards for the Petroleum Industries in Nigeria; LHC: Long Chain Hydrocarbons; SHC: Short Chain Hydrocarbons; ACL: Average Carbon Chain Length; CPI: Carbon Preference Index

## Short Communication

The fundamental and non-replaceable nature of the soil which is a natural link between other natural components like the air and water together complete the environmental cycle. The interactions between these natural components of the environment has helped in providing the essential necessities of life like food, water, fuel and support for man and other living organisms [1]. The soil which is

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very important for the survival of life when contaminated or polluted by petroleum hydrocarbons poses serious effect and danger to the environment worldwide and hence attracts the attention of the public. One major way which petroleum hydrocarbons enter into the environment is by the activities of man, which is not properly checked, managed or controlled [2]. Agricultural and industrial activities discharge waste into the environment (air, water and soil) [3]. Total petroleum hydrocarbons are now the main organic pollutant in the soil. The presence of total petroleum hydrocarbons in the soil have negatively impacted on human health, growth and proper functioning of other organisms in the soil space.

Soil contamination by total petroleum hydrocarbons has both a long-term and a short-term side effect on the soil quality and the proper functioning as well as the quality of food produced from the affected soil [1,4]. The production of energy is associated with transformation of 9-petroleum products and other chemicals. The process of crude oil exploration to the time of final consumption can lead to severe environmental pollution that may become irreparable [5]. The different chemicals that are produced from crude oil, when present in the environment can lead to several health problems in clean-up workers, which include pulmonary disease, neurological irregularities, skin colour change and pregnancy related issues in women. The pollution occasioned by total petroleum hydrocarbons can have a long-term or a short-term effect on the environment [6], man and other creatures that inhabit the surrounding where pollution is being noticed. Spills from fuels and oils in the soil causes severe environmental damage and pollution that potentially threaten the ecosystem and human life. The effect of the contamination of crude oil on the soil has negative impact due to its flow from soil and water to man [7].

The resultant effect of soil pollution by total petroleum hydrocarbons include the deterioration of the physical, chemical and biochemical properties of the soil, limitation in the growth of plants, deficiency of oxygen and water, shortage of nitrogen and phosphorus-based nutrients in the soil [7,8]. Hydrocarbon pollution or contamination leads to increase in the acidic value and concentration level of heavy metals in the soil [9,10]. The effect of such pollution on the soil is achieved by initiating deviations from provisional values thereby resulting in negative environmental and health challenges. The use of such lands become limited due to total petroleum hydrocarbons contamination and therefore requires immediate remediation activities, which if not put in place may affect the groundwater, rivers and the surrounding environment, thereby causing more harm [11]. The contamination of the soil by hydrocarbons have a resultant effect on the pH value by lowering it and grossly affects agricultural and horticultural crop output by lowering production [12,13].

The contamination/pollution of the site introduces different compounds into the soil which have differing chemical characteristics from the original mixture. The volatile components of the mixture easily evaporate while the heavier compounds seep through the soil and contaminate groundwater [14,15]. Soil particles may be attached to by these pollutants and stay over a long period, while microorganisms in the soil may break others down through degradation processes [16]. This work focused on the concentration of total petroleum hydrocarbon in soils in the immediate vicinity at the Estate and Works Departments of three universities in Port Harcourt, Rivers State, Nigeria, where heavy duty generating sets are kept as back-up electricity source in the universities.

## Materials and Method

### Selection of Site

Soil samples were collected from three universities in Port Harcourt, Rivers State, Nigeria. The universities are, University of Port Harcourt (UNIPORT), Rivers State University (RSU) and Ignatius Ajuru University of Education (IAUE). The samples were collected within the vicinity of the Estate and Works Departments of the various universities, where heavy duty generators are planted to supply power to the various campuses due to the inability of the National Power Generating and Distributing Companies to provide adequate power.

### Sample Collection

With the aid of soil auger, soil samples were collected at a depth range of 1 to 1.5m from the various sites. Four soil samples collected at random within the sites were combined together to form a 500g composite sample. The collected samples were then homogenized with the aid of a ceramic mortar and pestle plastic that was washed and dried previously. The soil samples were then kept in a thoroughly in clean plain amber bottles in order to avoid being contaminated. The soil samples were then placed in an ice-chest container. The collected samples were then transported to the laboratory for treatment before being analyzed for total petroleum hydrocarbons.

### Extraction of Soil Sample

Analytical weighing balance was used to weigh out 10g of the homogenized soil sample for extraction purposes, and then put into an amber bottle with the aid of a spatula. Addition of 5g anhydrous sodium sulphate into the amber bottle was made in order to remove moisture from the homogenized soil sample and then stirred with a magnetic stirrer for thorough mixing. 300 μg/ml concentration of 1-chlorooctadecane was added as a standard surrogate to the soil sample and 300ml of dichloromethane was then added to the amber bottle containing the soil sample as the extracting solvent. The amber bottle was thoroughly corked to prevent air from entering in and a mechanical shaker was used in agitating the samples for a period of 6 hours under room temperature conditions. The agitated samples were allowed to settle gently for an interval of one-hour and filtration was made using 11mm filter paper into a beaker that was previously washed and cleaned for that purpose and then allowed to concentrate to 1ml through evaporation under room temperature conditions in a fume cupboard [17,18].

### Soil Sample Clean-Up Procedure

The column clean-up was performed by the introduction of a glass-wool into a chromatographic column thoroughly washed, then silica gel was added into a beaker. Dichloromethane was added to the beaker containing the silica gel to make it slurry, and then introduced into the column. Sodium sulphate (anhydrous form) was then added, which was followed by n-pentane into the chromatographic column. The already concentrated sample was then mixed with cyclohexane and put into a beaker and then added into the already prepared column. Pentane was used in the eluting of the samples and then collected below the column. More pentane was being added for more elution of the sample to take place. The soil samples were evaporated to dryness after the column was rinsed with dichloromethane. The method of [19] and LAWI [18] were used in treating the soil samples.

### Separation and Detection of Soil Samples

Agilent 6890N Gas Chromatography-Flame Ionization Detector (GC-FID) was used to detect the level of total petroleum hydrocarbons in soils from the stations under investigation [20]. Injection of 3μl concentration of the previously clean-up sample was done with the aid of a micro-syringe into the GC-vial. The syringe was first cleansed by using blank dichloromethane to be injected through the syringe. The cleaning process of the syringe was done three times before being used for sample analysis. After rinsing the sample with the eluted soil sample, the sample was then introduced into the chromatographic column by the rinsed micro-syringe for the separation of the different components of the of analysis. The Flame Ionization Detector then detects the various components within the sample. At a certain chromatogram, the total petroleum components are being resolved and measured in mg/Kg for that soil sample.

## Results and Discussion

The degree of contamination due to the presence of total petroleum hydrocarbons in the soils from the Estate and Works Department's vicinity of the Universities are shown in Table 1. The result showed that total petroleum hydrocarbons concentrations were; IAUE, 1475.56904mg/Kg, RSU, 953.14203mg/Kg and UNIPORT, 968.93886mg/Kg. The result revealed that concentration levels were in the order; IAUE > UNIPORT > RSU. The values obtained in the three sites were all greater than the limit of total petroleum hydrocarbons allowed by the Department of Petroleum Resources (DPR), [21] of 50mg/Kg. The results obtained in this work were less than that obtained in the study of soil housing heavy-duty diesel generators in Delta Park of the University of Port Harcourt which was 19692.1±4.1mg/Kg in site P1 (0m) and greater than or within the same range with the result obtained in site 2 (200m) [22]. The results obtained were also higher, lower or within the same range with the work of Nwankwo, et al. [11] in evaluating soil contamination due to the spillage of oil in Akinima, Rivers State.

**Table 1:** Level of Concentration of Total Petroleum Hydrocarbons in the Soils of the Estate and works Departments of the Universities.

| Carbon Length (mg/Kg) | Universities | | |
|---|---|---|---|
| | IAUE | RSU | UNIPORT |
| C8 | 4.42145 | - | 4.40920 |
| C9 | - | - | - |
| C10 | - | - | - |
| C11 | 2.14677 | 2.01837 | 2.60133 |
| C12 | 3.35384 | 3.05701 | 3.21101 |
| C13 | 7.01018 | 5.73828 | 4.89693 |
| C14 | 21.74394 | 16.73274 | 18.72247 |
| C15 | 18.66803 | 14.00476 | 12.03548 |
| C16 | 45.84241 | 12.52497 | 16.51023 |
| C17 | 49.53566 | 19.96827 | 18.88639 |
| C18 | 135.62842 | 99.35272 | 93.36628 |
| C19 | 89.8452 | 30.02676 | 35.83295 |
| C20 | 83.50204 | 58.11582 | 58.32146 |
| C21 | 82.68804 | 74.39992 | 74.99342 |
| C22 | 17.02611 | 16.30559 | 19.30957 |
| C23 | 31.42845 | 24.50720 | 23.72050 |
| C24 | 29.26306 | 27.34844 | 25.43832 |
| C25 | 43.95466 | 33.11769 | 35.10964 |

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| | | | |
|---|---|---|---|
| C26 | 167.09702 | 14.73968 | 16.79836 |
| C27 | 35.07342 | 26.98035 | 24.97821 |
| C28 | 81.6706 | 15.50924 | 16.49205 |
| C29 | 90.79549 | 47.22181 | 47.11380 |
| C30 | 25.77430 | 25.01272 | 28.03269 |
| C31 | 58.98981 | 56.10310 | 57.31005 |
| C32 | 55.33079 | 49.87378 | 46.97482 |
| C33 | 67.20622 | 58.85809 | 64.35609 |
| C34 | 21.49957 | 22.46751 | 22.56355 |
| C35 | 44.79957 | 44.84194 | 43.78499 |
| C36 | 19.44003 | 25.15220 | 21.52510 |
| C37 | 74.94762 | 63.06201 | 66.05310 |
| C38 | 39.56833 | 31.08509 | 33.06915 |
| C39 | 15.34741 | 19.24156 | 17.64758 |
| C40 | 11.97005 | 15.77423 | 14.87414 |
| Total | 1475.56904 | 953.11949 | 968.93886 |

Alinnor, et al. [23] also showed that contamination of soil by total petroleum hydrocarbons decreased with increase in depth with recorded values of 519952, 2341, and 1116.96mg/Kg between 0.00-2.0m depth which is greater on the average when compared to this work. The work of Ibezue [24], on soil samples within Gokana, Rivers State revealed total petroleum hydrocarbon concentration level of 13949.42 and 8279.35mg/Kg, which were far higher than that obtained in this work. However, values of total petroleum hydrocarbons concentrations in soils obtained by Okop, et al. (2012) were 289, 417 and 178mg/Kg for top soil, subsoil and greater depth respectively, Li, et al. [25], Adeniyi, et al. [26], were within the range of 100-500mg/Kg, were all lower than the values obtained in this study. The results of this study as shown in Table 1, showed that accumulation of total petroleum hydrocarbons took place in all the stations and the highest value obtained in IAUE is an indication that diesel generators were used more often in IAUE than the other universities under investigation.

Further observation showed that supply of electricity by Electricity Distribution Company is in the order RSU > UNIPORT >IAUE, as the level of hydrocarbons pollution has revealed. The low and non-presence of the $C_8$-$C_{10}$ range might be as a result of volatility, biodegradation, leaching and reactions [23], associated with the range or probably due to the use of diesel engines, since diesel range is from $C_{11}$ above. The low presence of the $C_8$-$C_{10}$ range observed in IAUE and UNIPORT might have resulted from repair works on the generators during the period of study since petrol is often used in washing off grease and oil in engine parts during maintenance and servicing of heavy-duty machines. The study showed that the soil within the vicinity of the Estate and Works Departments in the Universities housing the heavy-duty generators were grossly polluted since they far exceeded the required acceptable level of Environmental Guidelines and Standards for the Petroleum Industries in Nigeria (EGASPIN) [27]. The anthropogenic behavior of man was the major reason behind this high level of contamination and pollution, since it is difficult for contamination of such level by organic pollutants to occur naturally [28]. The level of total petroleum hydrocarbons contamination is a global tool used in establishing soil clean-up targets as allowed by regulatory agencies involved [29,30].

The values measured in this work has established that the total petroleum hydrocarbons concentrations in the study areas of the universities were far more than the that recommended by the national regulatory body [31]. The observations from this work will further aid the monitoring and tracking of hydrocarbon contaminants in the soils of the study area and can therefore be of great help during remediation processes within the universities' communities. This observation agreed with Makadia, et al. [32] and Pinedo, et al. [33] (Table 1). The importance of total petroleum hydrocarbons as a tool in identifying the source of pollution and contamination of the soils within the Universities' Estate and Works Departments are shown in Table 2. Useful mathematical indices and ratios such as $C_{15}$-$C_{19}$ (odd number TPH), $C_{18}$-$C_{22}$ (even number TPH), low molecular weight to high molecular weight hydrocarbons (L/H), long chain hydrocarbons (LHC) to short chain hydrocarbons (SHC), average carbon chain length (ACL) and carbon preference index (CPI) were used in the identification of the origins of total petroleum hydrocarbons in the soil of the study area [34,35].

The ratio of long chain hydrocarbons to short chain hydrocarbons (LHC/SHC) is used in order to assess how abundant phytoplankton or vascular plants are in the soil environment [36]. Total petroleum hydrocarbons less than or equal to $C_{26}$ are known as short chain hydrocarbons while those greater $C_{26}$ are referred to as long chain hydrocarbons. The LHC used in this calculation were $C_{27}$+ $C_{29}$+$C_{31}$ while SHC were $C_{15}$+$C_{17}$+$C_{19}$. LHCs originates from vascular plants while SHCs originates from plankton and benthic algae [36]. The results obtained from this study varied from 1.8461 to 2.03603 with an average value of 1.71970 in the soils affected by total petroleum hydrocarbons due to heavy-duty diesel engines used to supply electricity. The ratio between 0.21-0.80 range establishes that phytoplankton dominates, 2.38-4.33 is an indication of mixed origin, but when the ratio is greater than 4, there is the dominance of terrestrial plant waxes [37]. The results obtained was an indication that the source of pollution in the soils might be from mixed origin.

The L/H is a ratio that establishes the quantity of low molecular weight total petroleum hydrocarbons ($C_{15}$-$C_{20}$) compared to the high molecular weight hydrocarbons ($C_{21}$-$C_{40}$). It is an important instrument for understanding the hydrocarbons sources in the environments [36,38]. The results obtained in Table 2 showed that the L/H ratio was within 0.33558-0.4123 range with an average value of 0.36372. The summation of the ratio of L/H when less than 1 implies hydrocarbons from higher plants, marine animals and water bacteria, plankton and petroleum sources while ratio greater than 2 is an indication of fresh oil. The study revealed that the L/H ratio was < 1, therefore the source may be due to hydrocarbons from higher plants. Considering the distribution pattern of total petroleum hydrocarbons in the soils under study as shown in Table 2, ranged from 953.11949 to 1475.56904 mg/Kg, with an average of 1132.20913 mg/Kg in the samples analyzed. This clearly indicated that specific total petroleum hydrocarbons were present.

The high presence of $C_{16}$, $C_{18}$, and $C_{20}$ was an indication of hydrocarbons of anthropogenic origin while that from microbial biogenic origin is visible with the presence of $C_{12}$ and $C_{14}$. $C_{15}$, $C_{17}$ and $C_{19}$ were also present, was an indication of phytoplankton and algae of soil biogenic origin [39,40]. The dominance of even number hydrocarbons over odd number hydrocarbons was an indication of anthropogenic origin while that due to $C_{25}$-$C_{35}$ hydrocarbons was an input from biogenic sources (terrestrial and vascular plants) [41,42]. The determination of $C_{31}/C_{19}$ ratio of total petroleum hydrocarbons in the soils of the studied areas was to ascertain hydrocarbons of terrestrial origin to those of marine origin. $C_{31}$ hydrocarbons is an indication of terrestrial biogenic origin while $C_{19}$ hydrocarbons provides information on the presence of biogenic origin [36]. $C_{31}/C_{19}$ values more than 0.4 is an indication that the total petroleum hydrocarbons present were not from marine origin as observed by Ahmed, et al. [38]. The $C_{31}/C_{19}$ hydrocarbon ratio obtained in this work ranged between 0.65657 to 1.86844 with stations average of 1.3748 which indicated that the hydrocarbon pollution is terrestrial in nature. This assertion agreed with Adeniji, et al. [42].

The carbon preference index (CPI), measures the ratio of odd to even numbered hydrocarbons and is calculated mathematically as:

$$CPI_{25-35} = 0.5\ [(C_{25} - C_{33} / C_{24} - C_{32})] + [(C_{25} - C_{33}) / C_{26} - C_{34})]$$

This index is a useful assessment to note the dominance of natural total petroleum hydrocarbons over anthropogenic hydrocarbons [44]. CPI values greater than 1 (ranging from 3-10) is an indication of biogenic materials such as epicuticular waxes in algae or terrestrial vascular plants and a dominance of odd numbered hydrocarbons. When CPI values are close to 1, it suggests the possibility of hydrocarbons from petrochemical source [45]. Observation from Table 2 revealed that CPI values ranged between 1.04594-2.60460 with an average value of 2.08124, which is an indication of aliphatic hydrocarbons from natural origin.

The average carbon length (ACL) is calculated using the relation [46,47].

$$ACL\,value = \frac{25(nC_{25}) + 27(nC_{27}) + 29(nC_{29}) + 31(nC_{31}) + 33(nC_{33})}{C_{25} + C_{27} + C_{29} + C_{31} + C_{33}}$$

This is a useful index in evaluating odd carbon dominance per molecule in environmental samples. The ratio helps in establishing the link between higher plants and normal alkanes with the environment. Non-polluted sites give constant values and the values depletes when an area is polluted with petroleum hydrocarbons [48]. The results obtained from the soil under study were within 29.47578 to 29. 79370 with an average of 29.66491. The results showed that the three stations were operated under the same conditions and the origin of hydrocarbon pollution in the areas were basically anthropogenic. The results showed higher values which was an indication of heavier petroleum hydrocarbons. This is corroborated by Adeniji, et al. [43] (Tables 2 & 3).

**Table 2:** Diagnostic Ratios and Source Identification of TPH in the Soils of the Estate and Works Departments of the Universities.

| Parameter | Universities | | | Mean |
|---|---|---|---|---|
| | IAUE | RSU | UNIPORT | |
| TPH | 1475.5604 | 953.11949 | 968.93886 | 1132.20913 |
| $C_{15}$-$C_{19}$ (Odd Number TPH) | 156.04998 | 63.99979 | 66.75482 | 95.60153 |
| $C_{18}$-$C_{22}$ (Even Number TPH) | 236.15657 | 13.77413 | 170.99731 | 19364267 |
| $C_{25}$-$C_{35}$ | 692.19191 | 394.72609 | 403.51425 | 496.81075 |
| LHC/SHC | 1.18461 | 2.03603 | 1.93847 | 1.71970 |
| L/H [($C_{15}$-$C_{20}$)/($C_{21}$-$C_{40}$)] | 0.41723 | 0.33835 | 0.33558 | 0.36372 |
| $C_{31}/C_{19}$ | 0.65657 | 1.86844 | 1.59937 | 1.3748 |
| CPI | 1.04594 | 2.59318 | 2.60460 | 2.08124 |
| ACL | 29.47578 | 29.72524 | 2.60460 | 2.08124 |

**Table 3:** Molecular Ratios and Indices for the Identification of Total Petroleum Hydrocarbons Source in the Soil.

| Ratios/Indices | Biogenic Origin (Plants/Microorganisms) | | | Petrochemical/Anthropogenic Origin | |
|---|---|---|---|---|---|
| | Terrestrial | Mixed | Marine | Degraded Oil | Fresh Oil |
| CPI | <1 | - | ~1 | ~1 or < 1 | - |
| LMW/HMW | >0.4 | - | ~1 | ~1 | >2 |
| nC31/nC19 | >4 | - | ≤0.4 | - | - |
| LHC/SHC | | 2.38-4.33 | 0.21-0.80 | - | - |
| ACL | Higher and Constant (close range) | | | Depletes and fluctuates (wide range) | |

## Conclusion

The present study evaluated the presence of total petroleum hydrocarbons in the soils from the Estate and Works Departments, where heavy-duty generators are mounted and found out that there was pollution of the soils as a result of the generator usage. The soil was polluted with hydrocarbons range of $C_{11}$ to $C_{40}$ which was an indication the generators actually used diesel and lubricating oil in their daily operation works. The evaluation of different ratios and indices to ascertain the pollution sources of total petroleum hydrocarbons in the soils from the universities' power stations. The results showed that the major source of hydrocarbons in the soils was anthropogenic. The relevant authorities should therefore put in place health institutions that can easily be assessed by the workers in the generator houses and effective remediation processes be put in place immediately or restore proper electricity through the national grid.

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