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Polycyclic Aromatic Hydrocarbon (PAHs) Levels in Treated Spent Engine Oil Contaminated Soil from Automechanic Workshop Site in Wukari, Nigeria

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Abstract

Spent engine oils contain Polycyclic Aromatic Hydrocarbons (PAHs) and can contaminate soils when spilled. PAHs have been linked to cancer. Therefore, there is need to reduce the levels of PAH contamination in contaminated soils. In the present study, spent engine oil contaminated soil samples were collected and divided into three portions. Two of the contaminated soil samples were treated with 0.5M NaOH and heat, applied at 250^{0}C, respectively. Furthermore, a sample of uncontaminated soil was collected 10 meters away from the contaminated site. All the samples were extracted with appropriate solvents using sonication technique, cleaned up and then analyzed with gas chromatograph coupled to flame ionization detector. The results show that total PAHs concentration of contaminated soil sample was 333.6820 mg/kg, contaminated soil sample treated with 0.5M NaOH, 104.8540 mg/kg, contaminated soil sample treated with heat at 250^{0}C was 180.0905 mg/kg and the uncontaminated soil sample, 0.7084 mg/kg. The results showed that Wukari auto-mechanic village is contaminated with PAHs through indiscriminate disposal of spent engine oil. Furthermore, treatment with 0.5M NaOH was a more effective means of remediation relative to treatment with heat at 250^{o}C for two hours. The study will provide an appropriate guide for the artisans following enlightenment about the possible threat that contact with PAH contaminated soils could have on their health.

Keywords: Spent, engine oil, uncontaminated, soil, ionization, detector, remediation

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Introduction

In Nigeria, as in many other countries, hydrocarbons from petroleum contamination are widespread. Pollution arising from the disposal of used engine oil is a common environmental challenge in Nigeria due to the manner of their disposal [1]. The contamination of soils from spent engine oils could arise from mishandling, deliberate disposal, spilling and leakage of petroleum products such as gasoline, lubricating oils, diesel fuel [2].

Environmental pollution by Polycyclic Aromatic Hydrocarbons (PAHs) has being found to be a serious issue especially from automobile sources [3]. This is due to the health implication of their presence since they are not required for any function either by plants or animals. PAHs can either be from naturally occurring or synthetic /anthropogenic sources such as burning of industrial materials, wastes among other forms of combustion [4]. They could therefore be present in the air, water and soil. As population and technology increase and improve, there is also an indirect increase in environmental pollution causing a continuing and accelerating decline in the quality of the environment and its ability to sustain life. Among other parts of the environment, PAHs are usually deposited on the soils [5].

Their deposition enables them to be sorbed onto soil particulate matter so that they become less mobile and less available for removal [3, 6]. Its removal from soils however can be effected through heating, photo-oxidation, use of chemicals and environmentally friendly sources such as plants and microbial enzyme mediated approaches [6]. Biodegradation of PAHs have also being employed using banana skin, brewery spent grain and spent mushroom compost [7].

Spent engine oil contamination is becoming very common nowadays due to their generation from increasing number of vehicles and other machines alongside their use as recycled lubricants in machines and on metal surfaces for enhanced performance. These practices are responsible for why spent engine oils spread and percolate the soil and water sources around towns where they are disposed [7, 8].

Most studies on auto-repair workshop sites have been reported by some authors previously just that the studies majorly addressed the levels and effects of heavy metals and polycyclic aromatic hydrocarbons on the soil following disposal of the spent engine oils without considering the remediation of PAHs and heavy metals in the soil [8-12]. These used oils and solvents constitute most of the wastes commonly generated in auto-repair shops around cities in Nigeria [12, 13].

Waste oil is a mixture of different chemicals including petroleum hydrocarbons, chlorinated biphenyls, chlorodibenzofurans, additives, decomposition products and heavy metals that come from engine part as they wear away [14].

Polycyclic aromatic hydrocarbons result from incomplete combustion of materials in the environment such as garbage, petroleum products, coal, meat and tobacco [15]. PAHs belong to a group of over 100 hazardous substances. They are organic pollutants consisting of two or more fused-benzene aromatic rings.

PAHs have also been found in facilities where petroleum products or coal are used or where wood, cellulose, corn or oil is burned. People living near waste sites containing PAHs may be exposed through contact with contaminated air, water and soil. They are often seen as persistent organic pollutants and their tendency to persist in the environment tend to increase as the number of rings increase [16].

They have also been seen to show a wide range of toxicological effects though the primary focus has been on their mutagenic and carcinogenic potential. U.S Environmental Protection Agency (USEPA) had classified seven of the 16 priority listed PAHs as probably carcinogenic to humans [17]. Furthermore, benzo[a]pyrene, benzo[b]fluoranthene, chrysene, dibenzo[a,h]anthracene and indeno[1,2,3-cd]pyrene had been classified as carcinogenic PAHs [18]. Cancer refers to a medical condition which is characterized by the uncontrolled growth and spread of abnormal cells in a part of the body leading to formation of malignant tissues [19]. This means prolonged exposure to PAHs could pose threat to human health. An assessment of auto mechanics carried out for their renal status and haematological profile showed significantly higher values compared to non-mechanics [20]. This puts them at risk of kidney and blood related ailments too. There is therefore the need to evaluate the level of PAH contamination in the soils at Wukari auto-mechanic workshops and ascertain ways by which contamination levels could be reduced.

Materials and Methods

Study area

The study area is Wukari auto-mechanic village. Wukari is a local government area in Taraba State, Nigeria having its headquarters in Wukari town. The Donga river flows through the area and the Benue River form a boundary with Nasarawa State to the Northwest.

The mechanic village is located along Wukari-Takum road, a few meters from the town. It is an area mapped out by the government of Taraba State for artisans in automobile business which involves constant changing of used motor engine oil. The area is populated with residential homes and also peasant farming activities carried out around it.

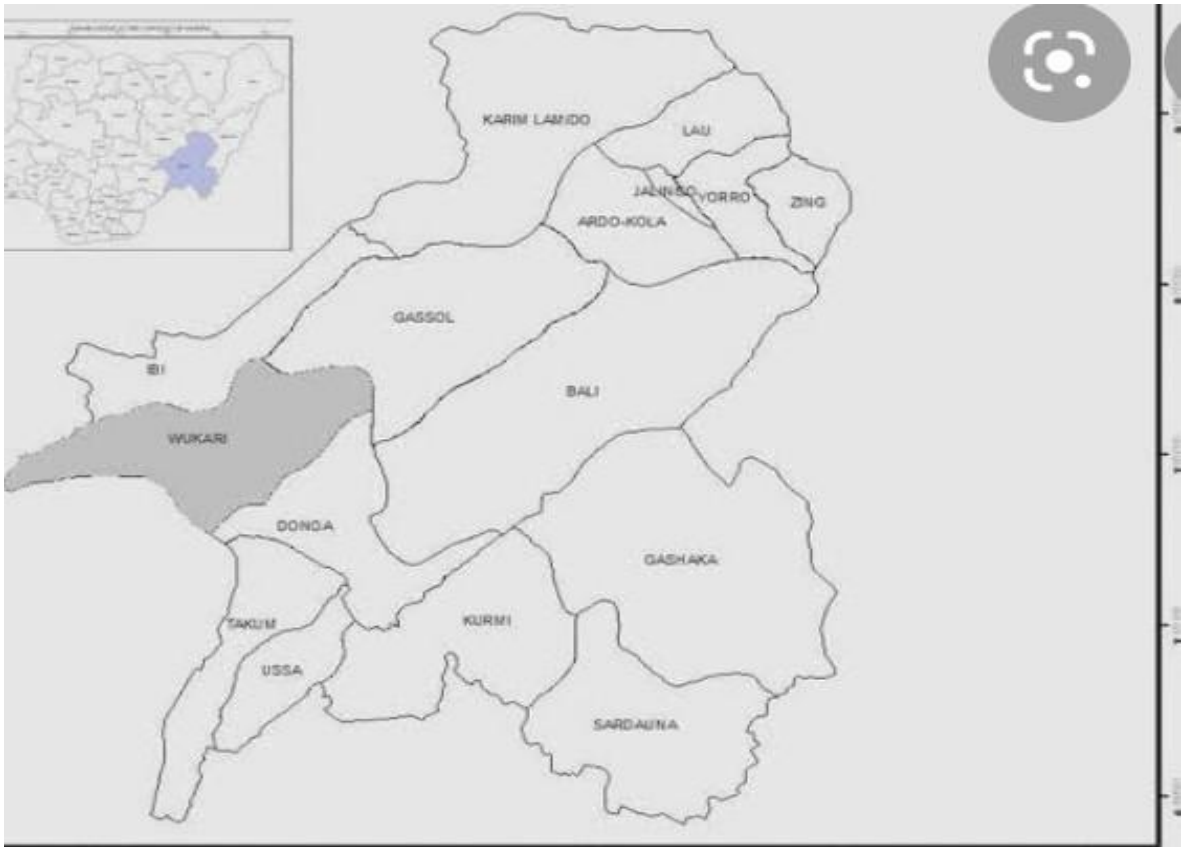

Figure 1: Map of Taraba State showing location of study area

Sample collection

The sample area was divided into two sub- areas; the contaminated site and non-contaminated site. At the contaminated site, Samples were collected from three points (workshops) at the auto-mechanic village using shovel to scrape off the top soil surface. The samples as collected were homogenized and divided into three portions and the second sample was collected about 10 meters away from the auto-mechanic workshop site where there were no spent engine oil contamination activities. The samples were each collected into black polythene bags to avoid the effect of UV light on them and then transported to the laboratory for pre-treatment and analysis.

Procedure for sample pretreatment

Treatment with 0.5M NaOH

Prior to analysis, 200 g of the contaminated soil sample was weighed and transferred into a beaker then 150 ml of the 0.5M sodium hydroxide solution was poured into the sample and stirred using a glass rod. The mixture was allowed to stay for two days to allow the solution to penetrate the sample properly. It was then kept at room temperature in a black polythene bag.

Treatment with heat

Prior to analysis, 200 g of the second portion of contaminated soil sample was weighed into a beaker and placed in the oven. The temperature knob was set at 250^{o}C and allowed to heat for 2 h after which the sample was removed and allowed to cool, then kept at room temperature.

Procedure for GC analysis

Extraction

The extraction was carried out using modified method 355^{0}C [21]. 3 g of the samples were weighed into a 250 ml capacity beaker and 50 ml of ratio 3:1 (75:25 ml) redistilled hexane -Dichloromethane mixture added. The beaker and its content were placed in a sonicator to extract the Hydrocarbon for about 30 min. The organic layer was filtered into the 250 ml capacity beaker and the extract dried by passing the filtrate through a funnel containing anhydrous sodium sulphate. The extract was then concentrated with a stream of nitrogen gas.

PAH separation

The concentrated oil was separated into the aliphatic profile and hydrocarbon profiles by packing the glass column with activated alumina. The treated alumina was packed into the column and cleaned properly with redistilled hexane. The extract was poured onto the alumina and was allowed to run with the aid of the redistilled hexane to remove the aliphatic profiles into the pre-cleaned 20 ml capacity glass container. The polycyclic aromatic hydrocarbon fraction was recovered by allowing the mixture of hexane and dichloromethane in ratio 3 to 1 to run through the alumina packed column and finally removed the most polar PAH by allowing the redistilled dichloromethane to run through the packed column then collected into the pre-cleaned borosilicate beaker.

The mixture was concentrated to 1.0 ml by stream of the nitrogen gas before the gas chromatograph analysis was carryout. 1 µL of the extract was injected into the gas chromatograph through the port. The gas chromatographic conditions are as indicated in Table 1.

Table 1: Condition of the gas chromatograph analysis

GC	Hp 6890 powered with Hp Chem. Station Rev. A09.01 (1206) Software.
Injection temperature	Split injection
Split Ratio	20:1
Carrier Gas	Nitrogen
Inlet temperature	250^{0}C
Column type	Hp-1
Column Dimension	30 m × 0.25 mm × 0.25 μm
Oven program	Initial temperature at 60^{0}C for 5 mm First Ramping at 15^{0}C/min for 14 min maintained for 3 min. Second Ramping at 10^{0}C/min for 5 min, maintained for 4 min
Detector	FID
Detector temperature	320^{0}C
Hydrogen pressure	28psi
Nitrogen Column pressure	30psi
Compressed air	32psi

Determination of % PAH Contamination Levels in contaminated Soil samples

The % PAH Contamination level (PCL) in the contaminated samples was developed to determine the extent of contamination of the soil samples. This was calculated using equation 1 below:

$$\%\ \boldsymbol{PCL = CUCSS - \frac{CUSS}{CUCSS}} \qquad \text{----------(1)}$$

CUCSS=Concentration of untreated contaminated soil sample

CUSS=Concentration of uncontaminated soil sample

Determination of % PAH remediation levels in treated samples

The % PAH Remediation level (PRL) in treated samples was developed to ascertain the extent of remediation due to treatment with 0.5M NaOH and heating at 250°C for 2 h. This was calculated using equation 2 below;

$$\%\ \boldsymbol{PRL = CUCSS - \frac{CTCSS}{CUCSS}} \qquad \text{----------(2)}$$

CUCSS=Concentration of untreated contaminated soil sample

CTCSS=Concentration of treated contaminated soil sample

Results and Discussion

The concentration of the 16 EPA target PAHs components (mg/kg) in the soil sample collected from Wukari Auto-mechanic site were determined. Two portions of the contaminated samples were treated with 0.5M NaOH and by heating for 2 hours at 250ºC respectively while the third portion was not treated. The control was collected few meters away from the spent engine oil contaminated site. The results of all the samples are presented in Table 2.

The concentration (mg/kg) of the 16 EPA PAHs investigated in the untreated contaminated soil sample as shown in Table 2 revealed that Naphthalene had the highest concentration of 78.1851 and Indeno[1,2,3-cd]pyrene, the least with the concentration of 2.3033. The other PAHs in decreasing order are Acenaphthylene (57.6974) >Acenaphthene (45.3266) >Anthracene (31.3429) >Fluoranthene (23.7040) >Phenanthrene (23.5700) > Chrysene (13.4171) >Fluoranthene (12.3296) >benzo[k]fluoranthene (8.5184) >benzo[a]anthracene (8.0773) >pyrene(7.1065) >benzo[a]pyrene (6.8609) >benzo[g,h,I]pyrene (6.8531) >benzo[b]fluoranthene (5.9441) >dibenzo[a,h]anthracene (2.4447).

The presence of PAHs with high potential for carcinogenicity in the untreated contaminated soil sample was in concentrations ranging between 5.9441-13.4171 mg/kg. They include benzo[a]anthracene (8.0773); chrysene (13.4171); benzo[b]fluoranthene (5.9441); and benzo[a] pyrene (6.8609). The concentrations determined for these PAHs were higher than reported in a study carried out by Obini *et al.* [8] which had concentrations ranging between 0.0162-0.0453 mg/kg. Also, in a study carried out in Edemudu(EU) and Gracebill road (GB) in Akwaibom state by Ekanem *et al.* [2] at the vicinity of automobile workshops during the dry and wet seasons gave results which showed that the PAH values from soils 10 cm deep for the carcinogenic PAHs, Chrysene and B[a]P in mg/kg were (0.28,0.26) and (0.23,0.29) respectively for EdemUdulocation and at Gracebill road the PAHs concentrations in both seasons were (ND, ND) and (0.15,0.18). The concentrations in these locations were lower than determined in the present study. Generally, the concentrations of the PAHs in the untreated contaminated soil samples were higher when compared to the Netherlands Maximum Permissible Concentration [22] except benzo[g,h,I]pyrene which fell within the permissible limits.

Furthermore, presented in Table 2 are the concentrations of the treated contaminated soil samples. The result showed a decrease in concentration of the PAHs for both treatments using 0.5M NaOH and heating at 250ºC for 2 h when compared to that of the untreated contaminated sample. Total concentrations of the sample treated with heat at 250°C for two hours was 180.0905 mg/kg and contaminated soil sample treated with 0.5M NaOH is 104.8540 mg/kg. The sample treated with 0.5M sodium hydroxide revealed a greater decrease when compared with that involving heating at 250°C. The reason for this is not clear but probably the hydroxide may have converted most of the PAHs to another form other than the USEPA listed PAHs which may have accounted for the decrease in their concentrations.

The PAH concentrations determined for the treated soil samples were also found to be higher when compared to Netherlands maximum permissible concentration (mg/kg) except benzo[g,h,I]pyrene and chrysene which fell below the Netherlands Maximum Permissible Concentration limits. Therefore it may suggest that treatment with higher concentrations of the hydroxide could improve the remediation of the contaminated soil samples.

Table 2: PAH concentration [mg/kg] of soil samples collected at Wukari mechanic village

PAH	CUCSS	CTCSS at 250^{o}C for 2 h	CTCSS at 0.5M NaoH	CUSS	Netherland MPC [22] (mg/kg)
Naphthylene	78.1851	45.3704	27.8048	0.2501	0.14
Acenapthylene	57.6974	30.9865	20.1040	0.1503	-
Acenapthene	45.3266	21.7298	14.7260	0.1471	-
Fluorene	12.3296	6.1514	3.8671	0.0199	-
Phenanthrene	23.5710	11.8166	4.9914	0.0108	0.51
Anthracene	31.3429	20.1086	8.6735	0.0136	0.12
Fluoranthene	23.7040	12.1927	6.5883	0.0100	2.60
Pyrene	7.1065	3.7493	1.9449	0.0297	-
Benzo[a] Anthracene	8.0773	4.2207	2.8574	0.0371	0.25
Chrysene	13.4171	7.1572	4.8123	0.0196	10.70
Benzo[b] fluoranthene	5.9441	3.2117	1.7630	0.0095	-
Benzo[k] fluoronthene	8.5184	4.5985	2.5606	0.0037	2.40
Benzo[a] pyrene	6.8609	3.8087	2.2434	0.0033	0.26
Indeno[1,2,3-cd] pyrene	2.3033	1.2037	0.5253	0.0025	-
Dibenzo[a,h] Anthracene	2.4447	1.2660	0.5408	0.0006	-
Benzo[g,h,i] pyrene	6.8531	2.5187	0.8509	0.0006	7.50
Total	**333.6820**	**180.0905**	**104.8540**	**0.7084**	

CUCSS: Concentration of Untreated contaminated Soil Sample; **CTCSS:** Concentration of Treated Contaminated Soil Sample; **MPC:** Maximum permissible concentration; **CUSS:** Concentration of Uncontaminated soil sample

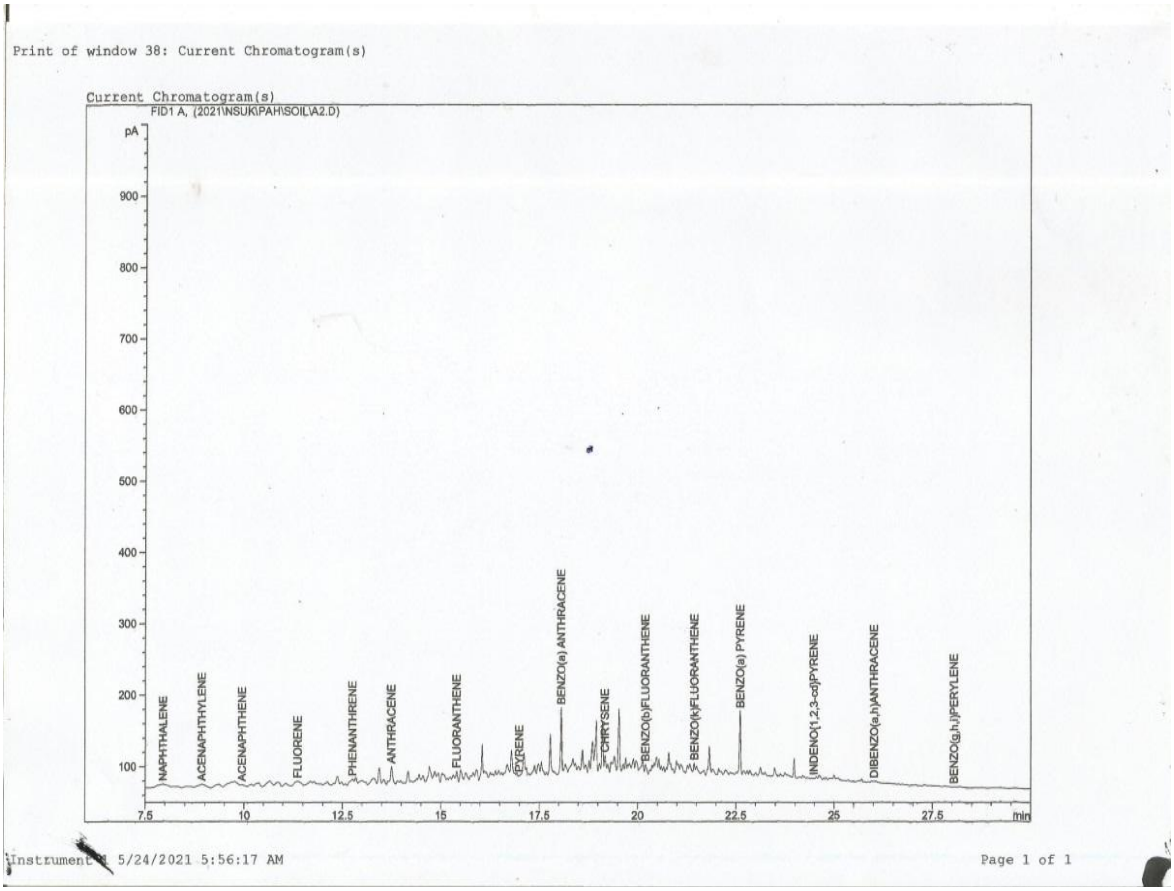

Figure 2: Chromatogram showing PAHs in the contaminated untreated soil samples

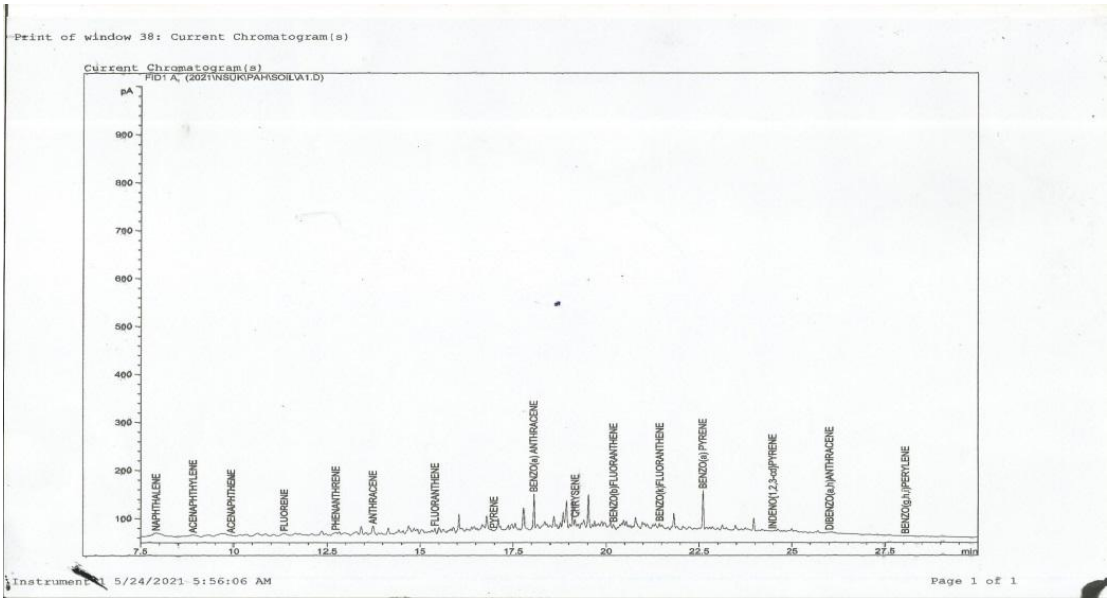

Figure 3: Chromatogram showing PAHs in the contaminated soil samples treated with 0.5M NaOH

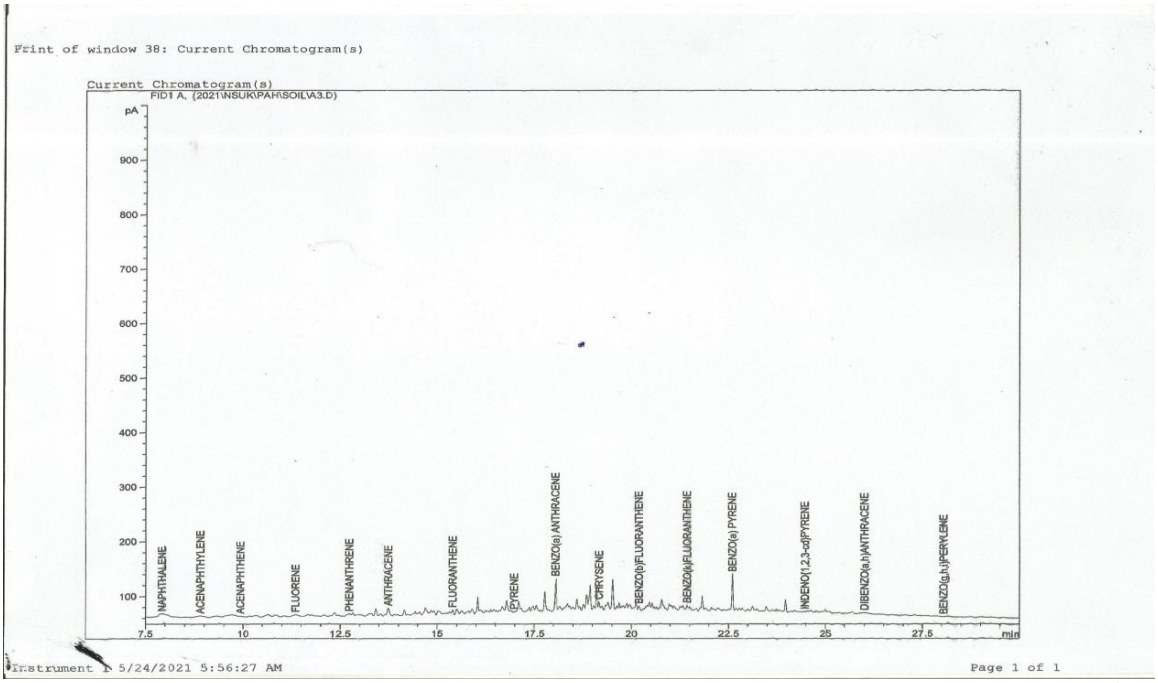

Figure 4: Chromatogram showing PAHs in the contaminated soil samples treated with heat at 250^{o}C

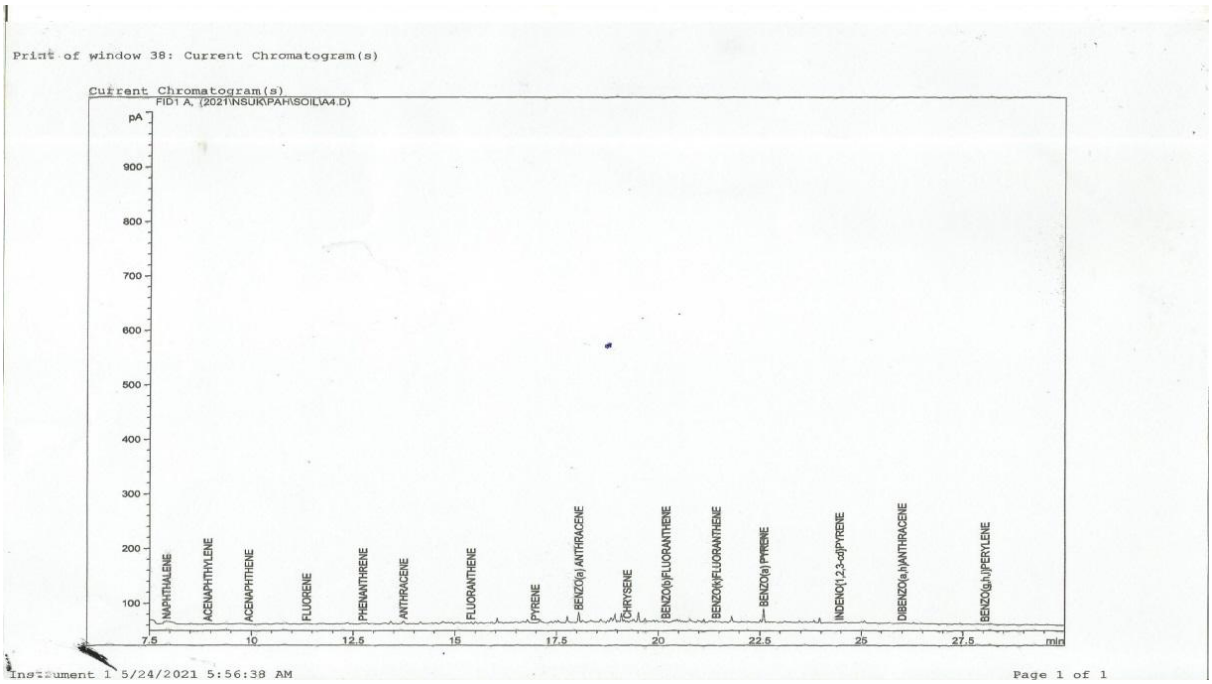

Figure 5: Chromatogram showing PAHs in the uncontaminated soil samples

It is evident from Table 3 that all the sixteen USEPA listed PAHs heavily contaminated Wukari auto-mobile workshop sites as against the background concentrations of the area with total concentration 0.7084 mg/kg. In fact, the heavy molecular weight PAHs like Benzo[b]Fluoranthene (99.96%), Benzo[a]pyrene (99.95%), Indeno[1,2,3-cd]pyrene (99.89%), Dibenz[a.h]Anthracene (99.89 %) and Benzo[g,h,i]perylne (99.99%) alongside some lower molecular weight PAHs such as phenanthrene (99.95%), Anthracene and Fluoranthene (99.96%) were those that contaminated the soils most. In a study carried out on PAH in soils around fuel stations in Wukari, dibenz[a,h]anthracene(18.6%) and Naphthalene (18.2%) were reported to be the most abundantly contaminated PAHs [22]. This report contrasted with the present study which revealed that before remediation approaches were employed all the 16 USEPA listed PAHs were very contaminated in the ranges of 98.68-99.99% with naphthalene being the least contaminated. This further indicates that spent engine oils contain more PAH including carcinogenic PAHs than petrol and from other activities that generate PAHs around fuel filling stations.

Table 3: % PAH contamination levels before treatment

PAH	% PAH Contamination levels in the soil samples
Naphthalene	98.68
Acenapthylene	99.74
Acenapthene	99.68
Fluorence	99.84
Phenanthrene	99.95
Anthracene	99.96
Fluoranthene	99.96
Pyrene	99.44
Benzo[a]Anthracene	99.54
Chrysene	99,85
Benzo[b]Fluoranthene	99.84
Benzo[k]fluoranthene	99.96
Benzo[a]pyrene	99.95
Indono[1,2,3-c,d] pyrene	99.89
Dibenzo[a,h] Anthracene	99.98
Benzo[g,h,i]perylene	99.99

Table 4: % PAH remediation levels after treatment

PAH	% PRL in CTCSS at 250°C for 2hrs	% PRL in CTCSS at 0.5M NaOH
Naphthalene	41.97	64.43
Acenapthylene	46.29	65.16
Acenapthene	52.06	67.51
Fluorence	50.12	68.64
Phenanthrene	49.87	78.82
Anthracene	35.84	72.32
Fluoranthene	48.56	72.21
Pyrene	47.24	72.63
Benzo[a]Anthracene	47.75	64.62
Chrysene	46.66	64.13
Benzo[b]Fluoranthene	45..97	70.34
Benzo[k]fluoranthene	46.02	69.94
Benzo[a]pyrene	44.49	67.30
Indono[1,2,3-c,d] pyrene	47.74	77.19
Dibenzo[a,h] Anthracene	48.21	77.88
Benzo[g,h,i]perylene	63.25	87.58

The % PAH Remediation Levels of the samples following their treatments are presented in Table 4. The % remediation levels of the soil sample treated with heat at 250°C for two hours was in the range of (35.84-63.25) while contaminated soil samples treated with 0.5M NaOH ranged between (64.43-87.58). It is worthy to note that benzo[g,h,i]perylene which contaminated the soil most (99.99%) was the most remediated following treatment with 0.5M NaOH (87.58%). It is evident from the study that treatment with 0.5M sodium hydroxide is a more effective means of remediation when compared with heating at 250°C for two hours. This position seems to be asserted by Ferrarese *et al.* [16] that chemical oxidation proved to be a more effective remediation technique, although heating for a longer period of time or higher temperature may give favorable remediation of the PAHs from the contaminated soil samples. The results also revealed that the low molecular weight PAHs have high concentrations in a typical soil sample contaminated with used engine oil. However higher molecular weight PAHs may tend to persist over time in the environment [16].

The uncontaminated soil sample showed that carcinogenic PAHs concentration ranged between (0.0033 - 0.0371) (mg/kg) which were lower compared to the Maximum Permissible Concentration. The low value of the recorded concentration of the uncontaminated soil sample collected few meters away from the workshops were spent engine activities took place should not be taken for granted, since there are no threshold concentration below which carcinogenic effect of PAHs does not occur. This could on bioaccumulation in body cells become a threat to life. The carcinogenic PAHs were present in concentrations of varying degrees in the contaminated soil sample. Consequently, it is possible that the population of people living and working in and around the Wukari mechanic village could be predisposed to high risk of cancer through contaminated soil and water.

Conclusion

The study was carried out to ascertain the levels and effect of treatment with NaOH and heat on polycyclic aromatic hydrocarbon in spent engine oil contaminated soil. It is evident from the study that Wukari mechanic village is contaminated with polycyclic aromatic hydrocarbons arising from indiscriminate disposal of spent engine oil. Furthermore, the study revealed that treatment with NaOH seems to be a more effective means of remediation of PAHs relative to treatment with heat. Even the lower concentration of others PAHs recorded in this study should not be taken for granted since there is no threshold concentration which carcinogenic effect of PAHs does not occur.

Conflict of interest: The authors declare no conflict of interest.

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