EVALUATION OF TOTAL PETROLEUM HYDROCARBONS AND SOME HEAVY METALS IN THE SOIL OF NNPC DEPOT IN ABA, NIGERIA

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Abstract: The study attempt to evaluate the presence of some heavy metals and Total petroleum Hydrocarbons in NNPC Aba soils. Heavy metals concentrations in soil samples were determined using Atomic Absorption Spectrophotometer (UNICAM SOLAR 32). Gas chromatography-flame ionization detector instrument was used in the detection and evaluation of levels of TPH while measurements of total PAHs concentration were performed with the aid of gas chromatography coupled to mass spectrometry (GC-MS). Pollution indices such as contamination factor (CF) and geo- accumulation index (I_{geo}) as well as established guidelines of some selected countries and organizations on site contamination were used to compare results. Soil samples were obtained in triplicates and in soil depths of 0–10 cm, 10–20 cm and 20–30 cm. The concentrations of Zn, Cu, As, Pb, Cd and Hg in soils of NNPC depot ranged from 112.30 to 202.10 mg/kg, 0.90 to 5.80 mg/kg, 0.08 to 2.50 mg/kg, 16.10 to 32.3 mg/kg, 1.90 to 11.78 mg/kg and 0.30 to 1.66 mg/kg with mean concentrations of 157.0, 3.84, 1.40, 22.30, 7.02 and 0.89 mg/kg respectively. Results of analysis of PAHs and TPH ranged from 6.30 to 7.40 mg/kg and 5120.50 to 24902.23 mg/kg, respectively. Values of geo-accumulation index revealed that the study site was contaminated with Pb, Hg and Cd. Established guidelines on assessment of site contamination soils from several countries used to compare this result demonstrated high levels of contamination of soils from NNPC depot by Hg, Pb, Cd, PAHs and TPH compounds. Heavy metals in the study site are presented in increasing order of concentration as follows: Hg < As < Cu < Cd < Pb < Zn.

Keywords: Geo-accumulation index, total petroleum hydrocarbon, soil contamination, heavy metal

Introduction

Crude oil has continued to be the main income earner for Nigeria since the change of attention from the agricultural sector to the oil sector. Nigeria crude oil is predominantly the light grade type with low sulphur content, high concentration of naphthenic hydrocarbons, dye additives, antioxidants, alkanes, alkenes, alkynes and heavy metals Guy *et al.*, 2012).
Refined petroleum products are more toxic compared to crude oil due to alteration of metal speciation as well as addition of new metal in the matrix during refining process. Petroleum products are transported by the Nigeria National Petroleum Cooperation (NNPC) through pipes lines to several oil depots distributed all over the country. The petroleum products are in turn transported from these industrial oil and petroleum storage facilities to the end users through mobile tankers. Several sources of oil contamination of our environment have been identified; during exploration of crude oil, willful damage of pipes and pipelines for political and economic reasons as well as during loading and offloading of petroleum products in these depots (Alinnor, 2013). Others are deliberate discharge by refineries and petrochemical plants of effluent containing some residual hydrocarbon products without pretreatment, accidental spills from tankers during delivery as a result of collision and ground in gas well as leakages from corroded and poorly maintained oil pipes.
Polycyclic aromatic hydrocarbons (PAHs) refer to a class of hydrocarbon molecules that have multiple carbon rings and are mostly made up of carcinogenic substances and environmental pollutants (Hussein *et al*., 2016). Polycyclic aromatic hydrocarbons consist of over 100 different chemicals that are usually released from the incomplete combustion of substances such as coal, oil, gasoline, trash, tobacco, wood and charcoal – broiled meat (Brigit, 2017). Polycyclic aromatic hydrocarbons are also released into the environment from forest fires and volcanoes. Polycyclic aromatic hydrocarbons are also manufactured and are mostly colourless white or pale yellow solids. Inhalation of polluted air, wood smoke and eating of contaminated food are the primary source of exposure to man (Hussein *et al*., 2016). Polycyclic aromatic concentrations fluctuate meaningfully in various rural and urban environments and are primarily influenced by vehicular and domestic emissions.

Total petroleum hydrocarbon (TPH) is a term used to describe a wide variety of derived petroleum compounds and it's by-products. The parameter (TPH) measures the gross quantity of these petroleum hydrocarbon products present in the environment instead of seeking to measure individual component separately which may be tedious and non-practicable. (Cheng- Di *et al.*, 2012).
The aim of this work is to evaluate the levels and the related health hazards of TPH and trace elements in soils of NNPC depot Aba and a reference site at the outskirts of Aba metropolis

Materials and Methods

Sample collection and preparation

Soil samples were collected with auger at different soil depths in the range 0-10 cm, 10-20 cm and 20 - 30 cm from soils within the vicinity of NNPC depot Aba and a reference site located at the outskirts of the city. The soil samples were homogenized in previously washed containers. After homogenization, portion of soil samples were kept in a properly washed amber bottle until use.

Heavy metal determination

One gram of sieved dried soil sample was weighed into a 100 mL beaker and 10 mL of nitric acid was added to the beaker. The content of the beaker was heated until dryness. 10 mL HNO_3 and $HClO_4$ was also added and the content of the beaker was heated until intense fuming was observed. The solution was then allowed to cool and filtered. The residue was treated with hot 6M HCl and the filterate made up to 50 mL. The filterate was analyzed using Atomic Absorption Spectrophotometer (UNICAM SOLAR 32).

Extraction procedure for TPH

Anhydrous sodium sulphate was added into an extracting bottle containing 10 g of previously

Homogenized soil samples and the mixture was stirred vigorously. 300 µg/mL of 1-chlorooctadecane (surrogate standard) was added to the sample. 30 mL of dichloromethane was added in the extracting bottle containing the sample and the bottle tightly corked before transferring it to a mechanical shaker. The sample was allowed to settle for 1 hour after agitation for 5 hour at room temperature in the mechanical shaker. The sample was filtered and the filtrate was allowed to concentrate to 1 mL by evaporation overnight in a fume chamber (LAWI, 2011; Alinnor and Nwachukwu, 2013).

Analysis of TPH

The separation and detection of compounds in the soil were carried out using Agilent 6890N Gas Chromatography- Flame Ionization Detector instrument. 3 µl of concentrated sample that was previously eluted from column was injected into Gas chromatograph. The micro-syringe of GC was first rinsed with dichloromethane (blank) and further rinsed with the sample prior to sample analysis. Separation of sample into constituent compounds was achieved by injection of sample into the chromatographic column. The compounds after separation were passed through a flame ionization detector for detection and the amount of TPH was resolved at a particular chromatogram in mg/kg (LAWI, 2011; Cortes *et* al., 2012; Alinnor and Nwachukwu,
2011).

Extraction procedure and analysis of total PAHs

The PAHs (total) was extracted and analyzed according to the method described by Nor *et al*. (2013). 500 mg of soil sample was dissolved in 25 mL n-hexane and acetone 7:3 (v/v).The extractions were done with the pressurized microwave extractions under control pressure, and over a period of about 40 minutes. The equipment was allowed to cool down at room temperature after extraction which was followed by filtration using whatman glass fiber filters and kept in 25 mL universal bottles. The samples were concentrated by use of rotary evaporator to 1 mL. Measurement of PAHs concentrations was done by the use of Gas chromatography - mass spectrometer equipped (Perkin Elmer Clarus) with elite column and the use of deuterated PAH internal standards (naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12}) and surrogate standards (2-fluorobiphenyl and 4-terphenyl-d_{14}) at 4000 mg/L and 2000 mg/L respectively (Cheng- Di *et* al, 2012). The carrier gas was helium and a constant flow rate of 1 mL/min was maintained and corrections were made of total PAHs concentrations using the surrogate standard recoveries.

Pollution indices

The degree of contamination with respect to measured back ground values of a geologically similar and uncontaminated area (or the average crustal composition of the metal) is appropriately described as Contamination Factor (CF).

CF is therefore expressed, $CF=C_m/B_m$ (1)

Where CF refers to the contamination factor, C_m is metal concentration and B_m is the background value of the metal.

Geo- accumulation Index (I_{geo}) is a quantitative tool which expresses the extent of metal accumulation in sediments. It was proposed by Mueller (1979) and has been very useful in evaluating the degree of metal contamination in both aquatic and terrestrial environment (Elias *et al*., 2011; Tijani *et al*., 2004).

$I_{geo}=In\ [(C_m)/1.5\ x\ (B_m)]$ (2)

Where, C_m and B_m is as defined above, 1.5 is a correction factor due to variation in the background concentration as a result of lithologic differences. Geo-accumulation index scale has seven descriptive classes (0 to 6) ranging from practically unpolluted to very strongly polluted.

Results and Discussion

The results of heavy metal content of soil of NNPC depot and the references is represented in Tables 1 and 2, respectively.

Table 1 revealed a stepwise decrease in lead content across soil profile studied. Lead concentration ranged between 16.1 mg/kg to 32.3 mg/kg with a mean concentration of 22.3 mg/kg. These values were higher than those obtained in Table 2, for control samples and also higher than 15.1 mg/kg reported by Adelekan and Abegunde (2011) as well as 14.13 mg/kg reported by Babatunde *et al.* (2014). The Pb values in the study area were however lower than value (76.92 mg/kg) of Okunola *et al.* (2007), 47.8 mg/kg of Sitkol *et al.* (2004) as well as USEPA (2008) and NEPCA (2010) relative relaxed criteria of 400 mg/kg and 300 mg/kg, respectively.

The values of calculated geo-accumulation index (**I_{geo}**) were presented in Table 3. These values showed as light contamination of lead in soils of NNPC depot at depth of 0-10 cm whereas depths of 10-20 cm and 20-30 cm remained uncontaminated. Inhalation and ingestion are the two main routes of Pb exposure to humans and other vertebrates. Lead finds its way and accumulates in the brain and could lead to death, gastrointestinal tract, kidney as well as central nervous system disorder. Other health risks associated with lead include impaired growth, loss of memory, nausea, insomnia and anorexia (NSC, 2009).

Table 1 also revealed a gradual decrease in the concentrations of Zn, Cu and As across soil layers of the profile. Concentrations of Zn, Cu and As ranged between 112.30 to 202.10 mg/kg, 0.90 to 5.80 mg/kg and 0.08 to 2.5 mg/kg with mean concentrations of 157.00, 3.84 and 1.40 mg/kg, respectively in soils of NNPC depot. These values were much greater than those recorded for control samples as shown in Table 2, indicating some level of accumulation of these trace metals on soils within the vicinity of NNPC depot Aba. The levels of Cu determined in the study area were higher than value (2.78 mg/kg) of Bai *et al.* (2008) but lower than 22.14 mg/kg and 47.0 mg/kg reported by Babatunde *et al.* (2014) and Fisseha *et al.* (2008), respectively. The mean value of Zn in this present study was well above 0.03 to 5.5 mg/kg and 25.06 mg/kg reported by Mitsios *et al.* (2005) and Srinivas *et al.* (2009), respectively.

Table 1: Heavy metal concentration (mg/kg) in soil at NNPC Depot, Aba

Depth (cm)	Pb	Cd	As	Hg	Cu	Zn
0-10	32.30 ±11.20	11.78± 3.03	2.50 ± 1.18	1.66 ± 0.43	5.80 ± 0.92	202.10± 27.56
10-20	18.60 ± 2.46	7.39 ± 2.76	1.63 ± 0.49	0.71 ± 0.21	4.82 ± 1.24	156.50± 17.10
20-30	16.10 ± 4.16	1.90 ± 0.48	0.08 ± 0.02	0.30 ± 0.19	0.90 ± 0.30	112.30 ± 8.23

Table 2: Heavy metal concentration (mg/kg) in soil at control site, Aba

Depth (cm)	Pb	Cd	As	Hg	Cu	Zn
0-10	0.020 ± 0.01	0.005 ± 0.002	0.002 ± 0.001	ND	0.900 ± 0.20	106.23 ± 19.83
10-20	0.007 ± 0.001	0.002 ± 0.001	ND	ND	0.059 ± 0.011	123.50 ± 8.047
20-30	0.004 ± 0.001	0.001 ± 0.000	ND	ND	0.030 ± 0.010	54.23 ± 9.96

ND = not detected

Other values recorded by Okunola *et al.* (2007) and Sitkol *et al.* (2004) were 237.96 mg/kg and 761 mg/kg respectively, which were considerably higher than those values obtained in this study. The calculated values of geo-accumulation index do not indicate any contamination of samples from NNPC depot by Zn, Cu and As. Arsenic is associated with skin damage, increase risk of cancer, and problem with circulatory system (Scragg, 2006). Copper and zinc are essential macro nutrient required for both plants and animal health. High doses of Cu are associated with anaemia, liver and kidney damage as well as irritation of both stomach and intestine (Bjuhr, 2007). Excess doses of Zn in the soil retard the breakdown of organic matter by influencing the activity of microorganism and earth worm (Greany, 2005).

The values of Cadmium recorded in the study area ranged between 1.90 to 11.78 mg/kg with a mean concentration of 7.02 mg/kg. Cadmium concentrations at the control site ranged between 0.001 to 0.005 mg/kg and were well below the values obtained from samples at the vicinity of NNPC depot. The mean Cd value in the study area was relatively close to 9.11 mg/kg recorded from soils within the vicinity of NNPC depot Jos, Nigeria by Babatunde *et al.* (2014) but significantly higher than 0.60 mg/kg obtained at automobile mechanic site in Benue, north-central Nigeria by Pam *et al.* (2013). The calculated values of CF and geo-accumulation index in Table 3 shown very strongly contamination of samples from the NNPC depot by Cadmium. Bioaccumulation of Cd in the Kidneys causes kidney dysfunction.

Table 3: **Contamination factor and geo-accumulation index of metals in soil**

Parameter	Depth (cm)	NNPC Depot		Control Site	
		CF	I_{geo}	CF	I_{geo}
Pb	0—10	2.019	0.297	0.0013	-7.0901
	10—20	1.163	-0.255	0.0004	-8.1399
	20—30	1.138	-0.399	0.0003	-8.6995
Cd	0—10	78.53	5.855	0.0333	-1.9095
	10—20	49.27	5.388	0.0133	-2.8260
	20—30	12.67	4.031	0.0067	-3.5190
As	0—10	0.500	-2.708	0.0004	-9.8390
	10—20	0.326	-3.135	ND	ND
	20—30	0.016	-6.150	ND	ND
Hg	0—10	20.75	2.627	ND	ND
	10—20	8.875	1.777	ND	ND
	20—30	3.750	0.916	ND	ND
Cu	0—10	0.083	-2.897	0.0129	-4.7593
	10—20	0.069	-3.081	0.0008	-7.4842
	20—30	0.013	-4.862	0.0004	-8.1605
Zn	0—10	1.531	-4.862	0.8048	-5.5055
	10—20	1.186	-5.118	0.9356	-5.3548
	20—30	0.851	-5.450	0.4108	-6.1778

Mercury was found to be present in all the soil samples from NNPC depot with highest concentration of 1.66 mg/kg and lowest concentration of 0.30 mg/kg. The result revealed a steady downward decrease in concentration of mercury across depths studied. Samples from NNPC depot recorded higher values of Hg concentration than samples from reference site. The value of Hg obtained at depth of 0 to 10 cm was above the NEPC, (1999) recommended limit of 1.0 mg/kg indicating site contamination by Hg. The geo-accumulation index also clearly indicated that NNPC depot soils were moderately contaminated with mercury.

Mercury is mainly toxic in its soluble and volatile alkylated forms and if it accumulates in the soil or agricultural crops above 0.006 ppm could cause permanent damage to the brain, lungs and kidneys (Scragg, 2006).

The results obtained from the analysis of Total Petroleum Hydrocarbons (TPH)

Hydrocarbons (PAH) in soils within the vicinity of NNPC depot and control site were presented in Tables 4.

Table 4: Total petroleum hydrocarbons (TPH) and polycyclic aromatic hydrocarbons (PAH) in soils within the vicinity of NNPC depot and at the control site

Depth (cm)	NNPC Depot		Control Station	
	PAH (mg/kg)	TPH (mg/kg)	PAH (mg/kg)	TPH (mg/kg)
0-10	7.40 ± 0. 89	24902.23 ± 99.89	0.050 ± 0.009	0.105 ± 0.015
10-20	6.47 ± 0.85	11349.00 ± 26.46	0.003 ± 0.001	0.012 ± 0.003
20-30	6.30 ± 0.60	5190.60 ± 70.51	0.090 ± 0.010	0.110 ± 0.020

The results of total polycyclic aromatic hydrocarbon in the study site recorded elevated values which ranged between 6.3 to 7.4 mg/kg as compared with 0.003 to 0.09 mg/kg obtained at the control site. Total PAHs concentration obtained in this study were higher than the recommended levels of 1000, 1500 µg/kg and 5 mg/kg imposed by soil clean-up guidelines from Denmark, Netherlands and Australia respectively (DEPA, 2002; MHSPEN, 2000; ANZECC, 1992). The high PAHs content of soil samples indicates contamination of the study site.

Inhalation, ingestion and dermal contact are the primary routes of exposure of PAHs to humans. PAHs are extremely toxic with excellent capability to stimulate health effects such as nausea, vomiting, eye irritation, diarrhea and confusion (short term effects). Other health effects (long term) include immune function suppression, cataracts, kidney and liver damage, skin inflammation, asthma amongst others. Generally, mixtures of PAHs are known to cause carcinogenic, genotoxic, teratogenic effects and are potential immunosuppressant. The mean TPH concentrations of soil samples obtained within the vicinity of NNPC depot are 24902.23, 11349 and 5120.5 mg/kg at depths of 0-10, 10-20 and 20-30 cm, respectively. The TPH concentrations were well above 0.09, 0.012 and 0.11 mg/kg obtain at control site across depths of 0-10, 10-20 and 20-30 cm, respectively and also higher than DPR (2002) and NSW (2009) recommended maximum permissible limit of 1000 mg/kg. Indicating high level contamination of soil samples from study site. Alinnor and Nwachukwu, (2013) reported that soil samples in Rivers State, Nigeria were contaminated with TPH concentrations of 1534.7, 1438.0 and 1651.0 mg/kg at depths of 0.0 to 0.5 m, 0.5 to 1.0 m and 1.0 to 2.0 m respectively, which are much lower than values obtained in this study. According to Iturbe

et al. (2004), the soil of coastal Mexican refinery was heavily contaminated with hydrocarbons with detectable TPH concentration of 130000 mg/kg. This value was higher than those recorded at soils from the study site (NNPC depot) in this work. TPH compounds are generally carcinogenic, Genotoxic, teratogenic and immunotoxin in nature.

Conclusion and Recommendation

The results of this study revealed elevated values of Pb, Hg, Cd, PAHs and TPH in the soil samples of NNPC depot when compared with the control samples. Values of geo-accumulation index indicated contamination of the study site by Pb, Hg and Cd (toxic metals). Established guidelines of several countries used to compare these results demonstrated high levels contamination of soils by these toxicants. Other elements such as As, Zn and Cu were also detected in the soil samples at levels which does not pose treat to health. Remediation and clean up measures should be adopted and periodically carried out on soils contaminated by toxicants in order to reduce or circumvent associated health hazards. Besides, reclamation of these soils from toxicants would maximize the land resources for agricultural operations and ultimately guarantee food safety.

Conflict of Interest

The author declared that there is no conflict of interest.

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