Oil-Gas and Environmental Nexus: Impact of Human Actions on selected Soil Physicochemical Parameters in Port Harcourt and its environment, Nigeria

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ABSTRACT: Impact of oil and gas activities on quality of soil in Port Harcourt and its environments was assessed. The study evaluates levels of physicochemical parameters associated with oil and gas industry in soils of 9 locations in Port Harcourt and its environs in Rivers State in Nigeria using standard methods. Composite samples were collected by random sampling from each of the 9 test locations. 3 control samples were also collected from each of the study areas. Benzene, Toluene, Ethyl-benzene, and Xylene (BTEX) and Total Petroleum Hydrocarbon (TPH) was measured using Gas Chromatography (Hewlett Packard 5890 Series II Gas Chromatograph FID). Total Organic Carbon (TOC) was measured using ASMD standard methods of analysis (ASMD 2579). Refinery Eleme and Aluu recorded the highest mean levels of BTEX (3.21±0.61 and 3.24±0.18 ppm respectively). The mean levels of TPH in soil were within the EPA acceptable optimum of 50 mg/kg except at A2 (Oquwi). There was significant difference among test samples and control samples as determined by one-way ANOVA (F11, 24=3.395; p=0.006). The highest mean levels of TPH was at A2 (56.70±25.77 ppm). ANOVA revealed that there were significant differences in TOC (p=0.023), BTEX (p=<0.001) and TPH (p=<0.001) among the study areas. Industrial areas I1 [Onne], I2 [Agbada] and I3 [Trans-Amadi] recorded highest levels of TOC. Oil and gas activities have negative impact on levels of BTEX. Industrial areas recorded highest levels followed by agricultural areas and lastly urbanized areas. The study concludes that oil and gas activities are a threat to soil health and integrity in Port Harcourt.

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Oil/gas industry also known as petroleum industry and includes processes of exploration, extraction, refining, transportation and marketing of petroleum products. During exploration, seismic surveys produce voluminous sounds that affect commercial species in water bodies and spawning grounds for the fishes. Extraction mud and cuttings are deposited on drilling sites which cause organic enrichment and accumulation of toxins. Further, production water contains naturally occurring heavy metals, radionuclides and oilfield chemicals. Refining of oil releases toxins to the atmosphere and effluent that contaminate soils and water bodies. Transportation and marketing are key processes in the oil and gas industry. During transportation, oil spills may occur along the pipelines, at the flow stations, cars, airplane, boats and trucks. History recalls some of the major oil spills; 1989 Exx Valdez in Alaska and 2010 BP deep water Horizon in Mexico, but hardly is the cumulative small spills from cars, boats, pipelines and airplanes accounted for. Oil is a raw material for many chemical products including pharmaceuticals, solvents, fertilizers, pesticides, synthetic fragments and plastics. Oils spills are common scenario and negatively linked to pollution of the environment and its inhabitants. Contaminants from oil to the environment include; paraffins, alkanes, cycloalkanes, aromatics (BTEX and TPHs), alkynes, alkenes, Sulphur, Nitrogen and heavy metals. Oil spills affect human health directly by penetrating through into the body, in foodstuffs and inhalation of fumes, or indirectly through destruction of recreational and agricultural activities. Oil spills in land penetrate vertically to contaminate underground water. Natural barriers like clay can restrict vertical movement and thus soil type is necessary to forecast possible contamination of underground water in case of a spill.

Nigeria is one of the largest crude oil producing nations in Africa and globally (Gboyega et al., 2011).
Rivers State is one of the centers of Nigerian Oil Industry with a high number of companies including; Shell Petroleum Development Company of Nigeria, AGIP, Texaco, Elf, NRPC, Michelin, West African Glass Industry, Alcan Aluminium, Metaloplastica, Risonpalm, NAFCON, Padbod Breweries among others (Chete et al., 2011). These are possible sources of organic and inorganic wastes that may pollute the environment. Organic wastes are simply biodegradable materials that are produced as agricultural wastes, market wastes, urban solid food wastes and municipal solid wastes (Chete et al., 2011). Organic wastes are among the leading sources of organic pollutants to water bodies, air, and soil. Inorganic wastes include non-biodegradable materials including; plastics, glass, metals, textiles, rubber, e-wastes, ash and other inert materials (World Bank, 2012).

Soil particle size plays a critical role as it influences the capacity of soil to hold water and nutrients (MIRA, 2012) and also can influence capacity of pollutants to penetrate vertically or horizontally to contaminate underground waters. Naturally, nitrogen exists in soil typically bound to organic matter or mineral soil material as either nitrate or nitrite, which are salts that can completely dissolve in water. Nitrates and nitrates can naturally be found in igneous and volcanic rocks. Animal wastes and nitrogen-containing fertilizers can increase concentration of nitrate in the soil. Bacteria can convert nitrite to nitrate hence affecting its availability and distribution in the environment. Human consumption of huge concentrations of nitrates can lead to decreased capacity of blood to transport oxygen (methemoglobinemia), low blood pressure, tachycardia, headaches, abdominal cramps and occasionally is fetal. The maximum contaminant level for nitrate is 1 mg/L and for nitrite is 3.3 mg/L (United States Environmental Protection Agency [U.S. EPA]). Phosphates occur naturally in soils and found in elemental form and free phosphate ion in solution also called inorganic phosphate. Benzene, toluene, ethylbenzene and xylene (BTEX) are highly volatile, with high mobility and are detectable in oil contaminated environments and are considered toxic and carcinogenic (Pinedo et al., 2013). Benzene is the most hazardous due to its carcinogenic and toxic characteristics (Ma, Martínez-pagán et al., 2014), where recommended limit is a maximum level of 5 μg/kg (U.S. EPA). Electrical conductivity is a measure of salts in soil and is an important soil health indicator. Electrical conductivity affects nutrient accessibility, and Microbial activities in the soil which influence key soil processes (Smita & Ingole, 2015). Soil pH is important as it affects all other parameters (Mccauley et al., 2017). A pH of less than 6 is acidic while a pH above 8.5 is alkaline soil (Mccauley et al., 2017). Potassium does not form major component of plant but is involved in many plant metabolic processes ranging from formation of cellular components to regulation of photosynthesis (Smita & Ingole, 2015). Potassium is found in mineral form and affects cell division in plants, carbohydrate formation, translocation of sugars, enzyme action and disease resistance (Prajapati & Modi, 2012). Total organic carbon (TOC) is carbon stored in soil organic matter and is derived through decomposition of living and dead organisms. In soils, Sulphur exists in organic and inorganic form and is cycled between the two forms by means of mobilization, immobilization, oxidation and reduction and mineralization. Sulphur is essential building block for proteins in plants and it can only be taken up from the soil solution as sulphate (Edwards, 1998). Of the 17 essential elements for plant growth, Sulphur (S) is considered the fourth important after Nitrogen (N), Phosphorous (P) and Potassium (K) in terms of plant requirement (Hopkins, 1999). Sulfur is found in commercial fungicides; Cosan, Crisazufre, Hexasul, Sulflox, Tiolene, and Thiolux, among others. Sulphur is known of low or little toxicity to human and animal health but can affect populations of microorganisms in soil. It is available in soil to plant and microbes mostly as sulphate. Sulphate in soils comes through soil minerals, fertilizers and pesticides and may be lost through reduction from inorganic Sulphur to sulphate by microorganisms; heterotrophic bacteria and fungi (Amponsah & Nagai, 2014). Sulphate can also be lost through reduction to Hydrogen Sulphide and through anaerobic respiration (Amponsah et al., 2014). Total petroleum hydrocarbons (TPH) refers to a large family of several hundred chemical compounds; hexane, jet fuels, mineral oils, benzene, toluene, xylenes, naphthalene, and fluorene, that originally come from crude oil and are all made from hydrogen and carbon (Tod et al., 1999). Total Petroleum Hydrocarbon (TPH) released to the soil may move through the soil and dissolve to the groundwater and move away to different areas from the release areas whereas some will evaporate into air. Some compounds will be attached to soil particles and be broken down by organisms (Tod et al.,1999).
MATERIALS AND METHODS

Description of the Study Site: This study was conducted in 9 selected areas of Port Harcourt, the Capital of Rivers State, Nigeria (Figure 1). The areas of interest were grouped into 3; urban, industrial, and agricultural areas. The urban areas of interest included; GRA phase 2, Diobu-Mile 1 and Mguoba. Agricultural areas of interest included; Aluu, Oquwi- Eleme, Eemuoha-Eu, while the industrialized areas were; Eleme which hosts the NNPC Refinery, Agbada-SPDC flow station in a rural setting and Trans-Amadi. The characteristics and main activities (Table 1) conducted in the study areas include; drilling and mining, fishing, fish farming, horticulture, dairy farming and crop farming, industrial processing. The study areas were assigned codes as in Table 1.

Sampling: Composite samples were collected by random sampling from each of the three areas; urbanized, industrialized and agricultural in the wet season (April to October 2018). Five (5) individual samples were collected following a random pattern around each test field. The five individual samples were thoroughly mixed by coning and quartering in a sterile container to attain a homogenous composite mixture. A total of 12 composite samples; A1, A2, A3, I1, I2, I3 U1, U2 and U3 as test samples, and CA, CI and CU as control samples (Table 1), were collected from the topsoil within a depth of 0 to 15 cm using a standard auger 3 times in the rainy season. Homogenized composite samples (400 gm) were then packed in polyethylene bags using a sterile wooden shovel. Samples for microbial analysis were collected using pre-sterilized materials to prevent contamination of the samples. Locations of the sampling sites were identified using a GPS and the GPS readings recorded. Samples were transported to the laboratory for analysis.

RESULTS AND DISCUSSION

Physicochemical Parameters: Soil pH: The soil pH of samples from in the 9 study areas varied from 4.5 to 10.30 (Table 2) over the study period, where some values were below or above the required EPA 816-F-09-004 limits of 6.5-8.5 (Table 2). The pH showed a decreasing trend from April to September (Figure 2).

Other Physicochemical Parameters:

Particle size
There was no significant difference in particle size in soils among test samples and controls as determined (p=1.000) [Figure 3]. Refinery Eleme (I1) recorded the highest mean level of particle size (45.13±27.43).

Electrical conductivity
Electrical conductivity recorded mean values within the WHO
acceptable optimum of 20 µS/m. There was no significant difference among test samples and control samples as determined (p=0.098). GRA phase 2 (U1) had the highest fluctuation with a minimum of 0.13 µS/m in April and a maximum of 0.57 µS/m in September. The highest mean value of electrical conductivity (EC) was recorded at A2; 0.29±0.24 µS/m (Table 3).

Table 2: Levels of pH in soil samples

| Study area | Study location | pH April | pH July | pH September |
|------------|---------------|----------|---------|--------------|
| Industrial | CI            | 10.10    | 8.40    | 7.20         |
|            | I1            | 10.14    | 7.40    | 6.50         |
|            | I2            | 8.62     | 7.30    | 4.50         |
|            | I3            | 9.24     | 8.50    | 7.50         |
| Agricultural | CA          | 10.50    | 7.80    | 4.50         |
|             | A1            | 8.94     | 7.10    | 5.00         |
|             | A2            | 9.13     | 7.40    | 6.00         |
|             | A3            | 9.34     | 7.40    | 7.00         |
| Urbanized  | CU            | 10.09    | 8.10    | 7.00         |
|            | U1            | 6.99     | 7.10    | 7.50         |
|            | U2            | 10.26    | 7.30    | 8.00         |
|            | U3            | 9.74     | 8.20    | 7.00         |

Fig 2: Variation in pH over the study areas in the wet season

Nitrate: The mean levels of nitrate in soil were above the EPA 816-F-09-004 acceptable optimum value of 100 to 500 mg/100g (Table 3). There was no significant difference among test samples and control samples (p=0.172). Ogali (CI) 848.99±95.96 mg/100g and GRA U1 (738.84±63.43) recorded the highest levels of nitrates.

Nitrite: The mean levels of nitrites (NO₂⁻) in soil samples were above the EPA 816-F-09-004 acceptable optimum range of 40 mg/100g. There was no significant difference among test samples and control samples (p=0.170). The highest values of NO₂⁻ were recorded in CI; 630.61±122.85 U1; 558.48±82.46 and U3; 433.52±362.01 (Table 3).

Phosphate: The mean levels of phosphate in soil were within the WHO acceptable optimum of 20,000 mg/100g. There was no significant difference among test samples and control samples (p=0.170). Eleme [I1] (757.00±81.61 mg/100g), Emohua [CA] (746.30±71.29) and U2 (920.93±124.53 mg/100g) recorded the highest levels of phosphate.

Sulphates: The mean levels of sulphates in soil were within the WHO acceptable optimum range of from 200 to 300 mg/100g. There was no significant difference among test samples and control samples (p=0.431) [Figure 3]. Oguwi (A2) area recorded the highest levels of sulphate (139.34±110.89 mg/100g).

Sulphur: The mean levels of Sulphur in soil were within the WHO acceptable optimum of 500 mg/g. There was no significant difference among test samples and control samples (p=0.175). Oguwi (A2) area recorded the highest levels of Sulphur (51.94±35.06 mg/kg).

TPH: The mean levels of TPH in soil were within the EPA acceptable optimum of 50 mg/kg. There was significant difference among test samples and control samples (p=0.006). The highest mean levels of TPH was at A2 (56.70±25.77 ppm) [Table 4].

BTEX: The mean levels of BTEX in soil in some areas was above the US acceptable maximum value 0.05 mg/kg (ppm) [US EPA 5021]. There was significant difference among test samples and control samples (p=0.004). Eleme (I1) and A1 recorded the highest levels of BTEX (3.21±0.61 and 3.24±0.18 ppm respectively).

TOC: The mean levels of % TOC in soil were above the U.S. EPA priority compounds acceptable optimum range of TOC 3% by weight. There was significant difference among test samples and control samples (p=0.023) [Figure 3]. Eleme (I1), I2 and I3 recorded the highest levels of % TOC, 8.32±0.86, 8.31±2.33 and 9.54±1.94 respectively (Table 4).
The pH levels of CI, I1, CA, CU and U2 > 9.0 in the month of April that were alkaline and were above the DPR recommended limits of 6.5-9.0 (Table 2/Figure 2). Some pH levels were below the same but were within the recommended limits of 6.5-9.0/ EPA 816-F-09-004 limits of 6.5-8.5. Industrial area (I2) [Aghada] and agricultural control area (CA) [Emohua] recorded a pH of 4.5 in the month of September. Fluctuation of pH values can be associated with acid water drainage or from surface runoffs. Runoffs from metal-rich areas usually have acidic pH and can affect pH levels in soils. Plants that grow in acidic pH and alkaline pH experience a spectrum of challenges including ion toxicity, shortage of nutrient and nutrient imbalances. This is an indication of presence of high levels of either calcium, magnesium or sodium (McCauley et al., 2017). Organisms that inhabit soils with pH values that are outside the recommended limits are exposed to low nutrient availability and high levels of toxicity (European Commission, 2013). There was a significant drop in pH in these areas in the month of September where they were within DPR limits of 6.5-9.0/ EPA 816-F-09-004 limits of 6.5-8.5. This can be attributed to the runoffs that occurred over the period as rain is acidic or neutral. Acid rains can be both beneficial and harmful, as they can neutralize acidic or alkaline soils for benefit of ecosystem integrity. The solubility of Mn$^{2+}$ and Al$^{3+}$ increases in acidic soils leading to toxicity to plants. High pH values were recorded in the month of April (Table 2) which decreased in the months of July and September (Table 2). Availability of Boron (B), Cu, Mn, and Fe depend on pH levels and tend to decrease with increase in pH, therefore level of pH of soils remains concern for farmers. Electrical conductivity mean values were within the WHO acceptable optimum of 20 µS/m. Elevated levels of EC is an indication of increasing level of salts in the soils. Soils that are affected by Ca, Mg, Cl, SO$_4$ or other salts show increased levels of EC. High values of nitrates were recorded in CI; 848.99±166.20; I2; 504.84±343.03 and U1; 738.84±109.86 (Table 3). Total Nitrogen includes nitrate, nitrite and ammonium. This study focused on the levels of nitrates and level of nitrite. The mean levels of nitrate in soil were within the EPA 816-F-09-004 acceptable optimum value of 100 to 500 mg/100g except for some areas which recorded values above the recommended limits, U1 recorded 848.99±166.20.
The mean levels of TPH in the study were above the US EPA acceptable maximum value 0.05 mg/kg [US EPA 5021]. This can be attributed to the varied sources of the contaminant and environmental degradation. BTEX are periodically and variably found in contaminated soils around many industries, storage tanks, accidental spills and pipeline leakages. Eleme (I1) and Aluu (A1) recorded the highest levels of BTEX (3.21±0.61 and 3.24±0.18 ppm) respectively [Table 4]. In the study areas, BTEX is influenced by anthropogenic activities. The findings are in agreement with those of Cheng, 2016, where proximity to sources of BTEX can influence contamination levels in the environment, which can be distributed in the environment vertically or horizontally in solvents (Cheng et al., 2016). Total organic carbon (TOC) is derived from putrefying organisms and metabolic activities of living organisms and also from synthetic sources (detergents, fertilizers, chlorinated organics, herbicides and industrial chemicals). Total organic carbon has effects on soil color, nutrient levels (anion and cation exchange), water retention capacity and aeration. Mean level of % TOC in soil of the study area was 6.28±0.46 (Table 4), which was above the USEPA priority compounds acceptable optimum range of TOC is 3% by weight or 30,000 mg/kg. The apprehension with high levels of TOC in soils is the potential to generate gas and thus should be maintained at low levels. Sustaining low TOC controls the development of biofilms through control of microorganism population. The findings are in agreement with the findings of (Kuang et al., 2018), where TOC were quantified in the study areas. The findings can also be related to findings of Blanchet et al., 2016, where TOC can vary according to activity and soil type (Blanchet et al., 2016), however there was no significant difference in soil types in the study areas.

**Conclusion:** This study reveals that physicochemical characteristics in the study area should be monitored periodically because there were isolated high trends in levels of TPH. High levels of TPH were in A2 (Oquwi), U1 (GRA) and I3 (Trans Amadi) which calls for close attention on monitoring. Industrial areas (I1 [Onne], I2 [Agbada] and I3 [Trans-Amadi]) recorded highest levels of TOC. Activities in industrial areas in Port Harcourt have highest impact on levels of TOC in soils as compared to agricultural and urbanized areas.
Oil and gas activities have negative impact on levels of BTEX. Industrial areas recorded highest levels followed by agricultural areas and lastly urbanized areas. The study concludes that oil and gas activities are a threat to soil health in Industrial and agricultural areas in Port Harcourt.

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