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Research Article
Water Quality and Hydrocarbon Contaminant Level in Soil and Fishes Around Abuloma Jetty, Port Harcourt, Nigeria

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Abstract

Background and Objectives: A jetty is a structure that projects from the land out into water. Anthropogenic activities in and out of the land and water introduce contamination around such water and that needs evaluation. This study was carried out to assess the water quality and hydrocarbon exposure levels of soil around the river and selected commonly consumed fishes (Tilapia and mullet) and seafood (Crab) collected from Abuloma Jetty river. Okujagu-ama river served as control. **Materials and Methods:** Biological oxygen demand, pH, total suspended solids, total dissolved solids, temperature conductivity, dissolved oxygen, PAHs, TPH and BTEX were measured in water and soil samples while, PAHs bioaccumulation were measured in studied fishes and seafood. **Results:** The physico-chemical parameters of the water showed a high degree of pollution of study water samples indicated by low DO, pH and high TSS. Benzo [b] fluoranthene gave the highest amount (6.91 mg kg^{-1}) among study fishes and seafood. Average concentration of TPH ranged from below detectable limit 0f 0.0001 to 55.81±1.00 mg kg^{-1} in water and 0.0001 to 1166.14±57.57 mg kg^{-1} in soil. Generally, the total TPH and PAHs concentration in Abuloma Jetty were significantly higher ($p<0.05$) than those in Okujagu-ama samples and exceeded set limits by United States Environmental Protection Agency (USEPA) and Department of Petroleum Resources (DPR). Diagnostic ratios applied implicated pyrogenic sources of PAHs in water from Abuloma and petrogenic sources for Okujagu. Highly carcinogenic Benzo{a}pyrene was found in abundance in samples from Abuloma Jetty river and portends danger. **Conclusion:** The consumption of fishes and seafood from this river should be discouraged and immediate remediation is highly recommended.

Key words: Polycyclic aromatic hydrocarbons, bioaccumulation, total petroleum hydrocarbon, petrogenic and pyrogenic sources, physico-chemical parameters, water quality, exposure level

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Majority of substances causing pollution from activities carried out both on land as well as water are being disposed into rivers, wetlands, seas and lakes and this has listed pollution in aquatic environment among the leading forms of pollution. Soil contamination around such river is another form of pollution that needs consideration. These aquatic environment provide water, food, transportation and a variety of economic opportunities for humans and also homes for several aquatic organisms. Aquatic organisms as well as plants provides majority of the proteins present in food taken by humans. Hence, determination of the water quality in aquatic life and the soil is essential due to the various activities carried out around and in the river.

Physico-chemical study is very essential to ascertain the precise thought about the quality of water and results of various physico-chemical parameters can be compared with standard values[1]. El Morhit *et al.*[2] stated that a healthy aquatic ecosystem is largely reliant on the physico-chemical and biological features. The condition of a water body can be influenced by several physico-chemical parameters and biological characteristics and these characteristics can detect a number of conditions for the state of living organisms and recommend necessary plans for environmental protection and management.

Polycyclic aromatic hydrocarbons are pollutants that exist abundantly in the environment which are made from both natural and anthropogenic processes and consumption of these substances can cause health challenges to humans and aquatic organisms[3]. Mohan *et al.*[4] reported that PAHs have been classified as a group of continuous pollutants since, they are toxic, carcinogenic and possess mutagenic properties and have ability to resist several forms of break down by micro-organisms. Since, PAHs are hydrophobic and easily absorbed by particulate matter, therefore coastal and marine sediments tends to absorb these compounds easily[5]. Due to the carcinogenic nature of PAHs, it can cause major impacts on marine organisms such as; benthic, Demersal and Pelagic fishes, crustaceans and shellfish[6]. Despite several successful remediation activities done on PAHs polluted sites, the biodegradation process is constrained by the scarcity of PAHs biodegrading micro-organisms due to lack of nutrients which is as a result of low aqueous solubility and strong adsorptive capacity to soil and sediments[7-9]. PAHs are a group of over hundred different chemicals which exist as complex mixtures that are generated during the incomplete burning of coal, gas, wood, garbage or other organic substances, such as; tobacco and overcooked meat[3]. They are present all around the environment in the air (can be attached to dust particles), water and soil (as solids) in substances such as; crude oil, coal tar pitch, creosote and roofing tar. Mohan *el al.*[4] reported that in medicine or in the manufacturing of dyes, plastics and pesticides PAHs can be used.

Total Petroleum Hydrocarbon (TPH) is a term used to refer to a large group of numerous chemical compounds that are derived from crude oil. However, TPH is actually a combination of chemicals. Almost all these chemicals comprise of hydrogen and carbon, therefore, they are referred to as hydrocarbons. The amount of chemicals contained in crude oils brings about a variety in the petroleum products to be made from the crude oil whereby some are produced as pure or light-coloured liquids which can easily evaporate and others are thick, dark liquids or semi-solids which do not easily evaporate. Most of these products possess gasoline, kerosene or oily odors. Due to the use of petroleum products (like fuel oil, kerosene and gasoline, etc.) by modern society, contamination of the environment by them is possibly extensive. The amount of TPH present in a sample can be used as a common pointer for petroleum contamination at that site, therefore, assessment of TPH level or concentration shows the influence that the petroleum hydrocarbons has on humans, animals and plants. Accidents releases from industries, byproduct from commercial or private uses are means through which TPH can be released into the environment. When there is a direct TPH discharge into water through spills and leaks, several TPH portions will float in water and create light surface films and other heavier portions will be stored in the sediment at the lowest part of the water, which may in turn affect bottom-feeding organisms. According to DEQ (2000), C-4 through C-9 hydrocarbons are generally said to be gasoline and condensate range organics (GRO), C-10 through C-24 are Diesel Range Organics (DRO), while C-5 through C-34 hydrocarbons are generally crude oil. The TPH can go in and exit the body through breathing of air, ingesting it from water, food or soil or touching it. Most TPH components gets into the bloodstream very fast when an individual inhales them as air or mist or swallow them[10]. They are assimilated faster into the body during breathing or when swallowed, but more slow to an extent when touched. Mostly TPH compounds exit the body by means of urine or breathing of air containing the compounds. This study investigated the water quality and hydrocarbon exposure levels of soil around the river and selected commonly consumed fishes and implication given heightened interactions in the environment.

MATERIALS AND METHODS

Study area: This study was done between February and December, 2018 and methods described by Nwaichi and Ntorgbo[11] were deployed.

Samples were collected from Abuloma Jetty and Okujagu-ama river both in Rivers state. Abuloma is surrounded by rivers linking to different communities like Ojimba-ama, Kalio-ama, Okujagu-ama and etc. Since, the Abuloma river is being linked to different communities, many businesses are being carried out there. The Abuloma Jetty is being occupied by several companies. These companies render services like leasing/construction of marine vessels and equipments such as; tugboats, barges, houseboats, dredger, amphibious excavators (swamp buggies), pay loaders, cranes, etc. They also carryout pipeline constructions maintenance, supply of sand, stones, bitumen, transportation of products like diesel, kerosene and wood, etc. At the Jetty most of these vessels bring in products like; diesel, kerosene (not often), sand, stones and wood, etc. During the offloading period most of these products or their fall-outs drop into the river. For example, diesel is being transported by barges which have tanks. During offloading of this product into drums or from one barge to another, some quantity of diesel get spilled into the river either due to leakages or careless acts of workers. Okujagu-ama which served as the control area is a community in Okrika L.G.A. in Rivers state that has the same geographic features as Abuloma like the rainfall and temperature pattern and also their type of soil and vegetation[12]. Okujagu-ama river was used in this study as a control site because the industrial facilities and activities mentioned for Abuloma are not present there.

Samples: Fresh samples of *Oreochromis niloticus* (Tilapia), *Liza falcipinus* (Mullet) and *Callinectes amnicola* (Crab) were collected from Abuloma Jetty (test) and Okujagu-ama (control) river in Port Harcourt and Okrika Local Government areas of Rivers state, Nigeria, respectively. At each site, three individual fish and crabs were collected, cleansed, covered and packed in aluminum foils, then stored in a frozen ice chest which was used to convey the samples for laboratory analysis. Plastic bottles were carefully rinsed with water and left to dry before being used to collect water samples (1L) from the two rivers in triplicates at 0–30 cm from the surface. Soil samples were collected randomly in triplicates from different points at the bank of the Abuloma Jetty and Okujagu-ama rive reach. The soil samples were collected between 10-40 cm depth (about 500 g each) using a clean and dry shovel. After each soil sample was collected, the shovel was washed with water and dried before the next was collected. All the samples were transported to the laboratory in clean polythene bags that were properly labeled for analysis.

Preparation of samples: Fresh fish samples were well cleaned in distilled water to remove any external dirt, dried in the oven at 60-70°C, ground to powder by using a manual grinding machine and then kept in an air tight container ready for extraction. Soil samples were allowed to undergo air drying, grinded and a 2 mm mesh sieve was used for sieving.

Determination of PAHs, BTEX and TPH

Extraction: Two grams of sample was measured into a clean extraction container and about 10 mL of the extraction solvent (DCM) was introduced into the sample where complete mixing was done and allowed to settle. A filter paper fitted into Buchner funnels was used to carefully filter the samples into a clean solvent-rinsed extraction bottle. The extract obtained was concentrated to 2 mL and removed for cleaning.

Cleanup/separation: A moderately packed glass wool of 1cm was positioned at the bottom of 10 mm ID×250 mm Loup chromatographic column. Slurry of 2 g activated silica in 10 mL methylene chloride was made and set into the chromatographic column where 0.5 cm of sodium sulphate was added to the top of the column. Extra 10 mL methylene chloride was used to wash the column and then pre-eluted with 20 mL of dichloromethane. This was left to run through the column for about 2 min until the liquid in the column exceeded the sulphate layer and immediately 1 mL of the extracted samples were transferred into the column. Dichloromethane of about 1 mL was used to rinse the extraction bottle and still added to the column. The stop-cork used for the column was opened and 10 mL graduated cylinder was used to collect the components. Just before the sodium sulphate layer was exposed to air, DCM was introduced into the column in about 1-2 increments. Accurate measurements for volume of 8-10 mL of the eluant was recorded and labeled.

Gas chromatography analysis: Labeled glass vials with rubber clip cap were used to collect the concentrated aliphatic fractions which were collected for gas chromatography analysis. One microlitres of the concentrated sample was injected using a syringe through a rubber septum into the column. At the vapor constituent partition, there was separation at the middle of the gas and liquid phase. As the samples appeared from the column (at constant flow rate) the FID detector automatically detected them and this response is based on the components of the vapor.

Physico-chemical analysis

Determination of temperature: The temperature measurements were made at the site of sampling with the aid of a Wagtech digital field thermometer. The probe of the instrument was dipped into the sample and the readings were taken when a steady temperature was attained.

Determination of pH: Inside a glass beaker, 20 mL of distilled water was added to 20 g of the soil sample forming a mixture which was stirred for about 10 min and allowed to stand for 30 min and stirred again for 2 min. Orion research digital pH meter model 407A was used to get the pH readings after prior calibrations.

Determination of conductivity (µS cm^{-1}): Deluxe conductivity meter was used to determine the electrical conductivity of the samples. The meter's probe was inserted into the sample and a steady reading was obtained which was recorded as the conductivity of the sample in µS cm^{-1}.

Determination of Total Dissolved Solids (TDS) and Total Suspended Solids (TSS): An evaporating dish was washed, oven dried at 108°C until a constant weight was obtained. A 100 mL of the water sample was poured into the dish and placed on a steam bath with holes. The water was evaporated to dryness, placed in the desiccators to cool to room temperature. The dried evaporating dish and solids were then weighed and the weight difference recorded as the total dissolved solid. The TSS was measured by thoroughly shaking the water sample (50 mL) and then a pre-weighed filter was used to filter sample. On the filter, the residue left was dried in an oven at about 105°C and then left for a period of time to cool. The weight of the filter was measured until a constant weight was attained. The total suspended solids level is being indicted by the increase in weight of the filter.

Determination of Dissolved Oxygen (DO): The water sample was carefully introduced into a 300 mL glass BOD stopper bottle and allowed to fill the brim. Immediately 2 mL of $MnSO_4$ solution was mixed and then 2 mL of alkaline potassium iodide solution was added. The bottle was closed to entrap no air bubbles before mixing gently. A 2 mL of concentrated sulphuric acid was introduced into the sample and closed. It was mixed gently by tilting the bottle to dissolve all the previously formed precipitate. An aliquot of 200 mL of this solution was titrated against freshly prepared sodium thiosulphate solution (N/80) using starch indicator. The titer value is equal to the dissolved oxygen in mg L^{-1}.

Determination of Biological Oxygen Demand (BOD)

Principle: The BOD tests are generally carried out by measuring the quantity of dissolved oxygen available in the sample before and after the incubation in the dark for 5 days at 20°C.

Procedure: Dilution water was the diluent for the sample. This was prepared by adding 1 mL of the reagents (phosphate buffer solution, magnesium sulphate solution, calcium chloride solution and ferric chloride solution) into good quality distilled water at the rate of 1 mL L^{-1} in a 150 mL BOD bottle. The stoppers were inserted living no air bubbles in the bottles. An initial determination of DO was taken before the dilution from one of the duplicate bottles. The samples were then incubated including a blank for 5 days in the dark at 20°C in an air cooled incubator. On the 5th day, the DO was measured in the incubated samples and the blank. The BOD after 5 days is computed as follows (Q10841-INFO):

$$\text{BOD} = \frac{\text{IDO} - \text{FDO}}{\text{VS / VB}}$$

Where:

IDO : Initial DO of diluted sample (mg L^{-1})
FDO : Final DO of diluted sample (mg L^{-1})
VS : Volume of sample (mL)
VB : Volume of bottle (mL)

Statistical analysis: Data obtained were subjected to student's t-test using the statistical package SPSS (version 21) while test of significance was done at 95% confidence level.

RESULTS AND DISCUSSION

Physico-chemical parameters of water samples studied: The Biological Oxygen Demand (BOD) levels in the two sites were not above permissible limit[13] of 2 mg L^{-1} and were not significant at $p<0.05$. Dissolved Oxygen (DO) of water sample from Abuloma water was markedly lower than recommended level (Table 1) while that of Okujagu-ama was within the permissible limit. This low level of DO in Abuloma water may be associated with the oil spillage occurrences and other

Table 1: Physico-chemical parameters of study water samples

Parameters	Abuloma jetty	Okujagu ama (Control)	WHO standards
BOD (mg L^{-1})	1.22±0.01	0.63±0.03	2*
DO (mg L^{-1})	2.79±0.02^a	7.55±0.17^a	8-10
TSS (mg L^{-1})	34.36±0.03^a	7.52±0.67^a	25**
TDS (mg L^{-1})	97.58±0.24^a	16.98±0.84^a	1000
Conductivity (µS cm^{-1})	296.16±5.80^a	126.66±2.27^a	1000**
pH	5.62±0.01	6.57±0.02	6.5-8.5
Temperature (°C)	32.0±0.00	30.3±0.33	35°C

*FEPA: Federal Environmental Protection Agency, **NDWQS: National Drinking Water Quality Standard, BOD: Biological oxygen demand, DO: Dissolved oxygen, TSS: Total suspended solids, TDS: Total dissolved solids, Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$)

Table 2: Physico-chemical parameters of soil samples

Parameters	Abuloma jetty	Okujagu ama	FEPA standards
TSS (mg kg^{-1})	429.77±29.34^a	81.85±2.85^a	60*
TDS (mg kg^{-1})	182.28±21.81^a	45.25±1.64^a	-
Conductivity (µS cm^{-1})	306.18±36.26^a	75.47±2.74^a	1000
pH	5.55±0.00	4.56±0.03	6.0-8.5
Temperature (°C)	32.73±0.37	33.33±0.67	35°C

*DPR: Department of Petroleum Resources, Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), TSS: Total suspended solids, TDS: Total dissolved solids

industrial activities being carried out. Bavinck *et al.*[14] reported similar result and noted that the presence of crude oil spilt could inhibit O_2 penetration into the water where fishes live thus bringing about O_2 tension. The DO values from this study were also below the report of other similar researchers[15,16]. Total Suspended Solids (TSS) showed a high value in Abuloma river, when compared to the control site which may be due to the frequent washing of particles into the river from the refuse dump at the river bank. The TSS standards were adopted for fisheries and water supply having a formulated standard concentration of 50 and 25 mg L^{-1}, respectively[17]. The mean concentration of TSS from the values gotten in this study in Abuloma (34.36 mg L^{-1}) water was slightly higher while that of Okujagu-ama (7.52 mg L^{-1}) was within the permissible limit[18] of 25 mg L^{-1}. High TSS brings about absorption of heat from sunlight by suspended particles leading to an increase in temperature of the water. Mitchell and Stapp[19] also reported that a high TSS can lead to further reduction in the dissolved oxygen levels (because warmer waters can hold less DO) and thereby, affecting aquatic life in several ways. The concentration of Total Dissolved Solids (TDS) from the two water samples fell within the FEPA tolerance limits of 500 mg L^{-1}, but were significantly different. This is evident in documented low DO levels for Abuloma river. On the other hand, extremely high or low TDS concentrations can affect the growth of many aquatic lives and can even lead to death. Measured conductivity values for both water samples fell within the permissible limit[19] of 1000 µS cm^{-1}, but were markedly different. The increased mean value of conductivity in Abuloma water may be associated with more decomposition and mineralization of organic materials in the river[20]. The pH reading for water sample from Abuloma was acidic and fell outside the permissible range, while that of Okujagu-ama was within WHO permissible limit of 6.5-8.5. This acidity level observed in the Abuloma Jetty river may be attributed to the high presence of organic materials in the river. Baker and Schofield[21] reported that previous studies have shown that low pH is toxic to fish and other aquatic lives. The temperature values were within WHO allowable limit of 35°C. The mean value of temperature in this work is above the report of Agbaire *et al.*[22] in Abaraka Delta state, Nigeria, but in tandem with the work of Taiwo *et al.*[23] in the study on Nigerian surface waters.

Physico-chemical parameters of soil samples studied: The markedly high level of TSS in the soil sample from Abuloma may be due to the presence of the refuse dump depositing solid waste on the surface of the soil. This introduced pollutants which may be released from the surface of the land into the water body by flowing water. Similarly, Total Dissolved Solids (TDS) level of Abuloma (182.28±21.81 mg kg^{-1}) was significantly higher than those from Okujagu-ama (45.25±1.64 mg kg^{-1}). The conductivity levels in the soil from Abuloma was markedly higher than Okujagu. This may be due to the breakdown of organic constituents of waste by the micro-organisms present in the dumpsite close to the Abuloma Jetty river which gives rise to more-organic carbon (matter) in the soil. These levels of conductivity in the two sites fell within permissible limits by FEPA standard (Table 2). The acidic pH values obtained from the two sites were implicative of non-point culprits. There was no significant difference in the temperature of soil samples around Abuloma river (32.73±0.37) and Okujagu-ama river (33.33±0.67). The temperatures of the two sites were within the tolerable limit by FEPA.

Polycyclic Aromatic Hydrocarbons (PAHs) in water samples studied: A total of 16 PAHs were analyzed (Table 3) for in water samples from Abuloma and Okujagu-ama rivers, 14 PAHs except Anthracene and Fluoranthene were detected in the water samples from Abuloma while, 3 PAHs

(Acenaphthene, Phenanthrene and Benzo[b]Fluoranthene) only were identified in the water samples from Okujagu-ama. The average concentrations of individual PAHs ranged from below detection limits of 0.0001 to 33.89±3.28 mg L^{-1} in Abuloma and Okujagu-ama rivers, respectively. The total concentration of PAHs in the water samples from Abuloma and Okujagu-ama rivers was 244.03±40.84 and 6.41±1.4 mg L^{-1}, respectively. The two sites varied significantly in the values for total mean concentrations of PAHs in the water samples. These results suggested that the high concentration of PAHs obtained could come from the various industrial activities carried out at the Abuloma Jetty such as; the offloading and loading of petroleum products which can introduce PAHs into the water due to careless acts that can cause leakages and spills. The level of PAHs in the water sample from Okujagu-ama river may be likely from transportation by engine boats through the seepage of diesel or the careless removal of engine oil from the boat[24]. Tronczyński *et al.*[25] reported that water in which its total PAHs level is greater than 10 µg L^{-1} is said to have a high amount of PAHs pollutants, so Okujagu may not be termed high PAHs polluted water. The mean concentrations of seven carcinogenic PAHs with high molecular weight (Benz (a)anthracene, Benzo(b) Fluoranthene, Benzo(k) Fluoranthene, Benzo(a)Pyrene, Chrysene, Dibenz (a,h) Anthracene and Indeno (1,2,3,-cd)Pyrene) in water from Abuloma river ranged from 8.55±1.54 to 33.89±3.28 mg L^{-1} and were higher than the safe limits of 0.020 µg L^{-1} established by EPA National Recommended Water Quality Criteria for the safety of aquatic life and human health[26]. A combination of several PAHs has been identified to initiate skin irritation and inflammation. Direct skin irritation can be caused by anthracene, benzo(a)pyrene and naphthalene while, anthracene and benzo(a)pyrene have been shown to be skin sensitizers in animals and humans, that is they can result to a sensitive skin reaction[27]. This indicated that the residents around the study area are at risk of having cancer and other serious ailments. Inhalation, ingestion and dermal contact are the primary routes of exposure for PAHs to humans. Nasher *et al.*[24] reported a lower value (32-46 µg L^{-1}) for PAHs concentration on Langkawi Island, Malaysia when compared to this study.

Table 3: Mean concentrations of PAHs in water samples from Abuloma Jetty and Okujagu-ama rivers

PAHs (mg L^{-1})	Abuloma	Okujagu-ama
Naphthalene	8.29±0.22	BDL
Acenaphthylene	20.72±1.09	BDL
Acenaphthene	28.79±3.42^a	1.48±0.39^a
Fluorene	25.10±4.39	BDL
Anthracene	BDL	BDL
Phenanthrene	18.40±7.8^a	2.36±0.99^a
Fluoranthene	BDL	BDL
Pyrene	14.03±1.82	BDL
Benz[a]anthracene	8.55±1.54	BDL
Chrysene	15.95±2.79	BDL
Benzo[b]Fluoranthene	14.26±1.76^a	2.57±0.05^a
Benzo[k]Fluoranthene	33.89±3.28	BDL
Benzo[a]pyrene	10.35±2.27	BDL
Indeno[1,2,3-cd]Pyrene	22.81±1.79	BDL
Benzo[g, h, i]perylene	6.38±0.89	BDL
Dibenz[a, h]anthracene	16.51±3.39	BDL
Total PAHs	244.03±40.84^a	6.41±1.43^a
LMW-PAH/HMW-PAH ratio	0.71	1.49
BaA/(BaA+Chry) ratio	0.35	0.00

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg L^{-1}

Table 4: Mean concentrations of PAHs in soil samples around Abuloma Jetty and Okujagu-ama rivers

PAHS (mg kg^{-1})	Abuloma jetty	Okujagu ama
Naphthalene	87.76±38.70	BDL
Acenaphthylene	38.94±9.06	BDL
Acenaphthene	61.06±1.84	BDL
Fluorene	46.75±6.58	BDL
Anthracene	17.63±3.03	BDL
Phenanthrene	51.46±16.20	BDL
Fluoranthene	BDL	3.18±0.22
Pyrene	73.81±21.61	BDL
Benz[a]anthracene	41.96±10.20^a	1.59±0.17^a
Chrysene	82.80±26.58^a	2.24±0.03^a
Benzo[b]Fluoranthene	74.83±22.27^a	3.04±0.37^a
Benzo[k]Fluoranthene	105.45±38.85^a	1.94±0.13^a
Benzo[a]pyrene	26.65±3.76^a	2.07±0.25^a
Indeno[1,2,3-cd]Pyrene	62.57±12.44	BDL
Dibenz[a, h]anthracene	82.79±4.74	BDL
Benzo[g, h, i]perylene	61.29±21.73	BDL
Total PAHs	915.75±237.59^a	14.06±1.17^a
LMW-PAH/HMW-PAH ratio	0.50	0.00
BaA/(BaA+Chry) ratio	0.34	0.42

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg kg^{-1}

Polycyclic Aromatic Hydrocarbons (PAHs) in soil samples studied: A total of 16 PAHs were detected for in soil samples from Abuloma and Okujagu-ama. From this study findings, 15 PAHs except fluoranthene were detected in the soil samples from Abuloma while 6 PAHs (Fluoranthene, Chrysene, Benzo[b]Fluoranthene, Benzo[k] Fluoranthene, Benzo[a]pyrene and Indeno [1,2,3-cd]Pyrene) were detected in the soil samples from Okujagu-ama. The average concentrations of PAHs in the soil ranged from below detection limits of 0.0001 to 105.45±38.85 mg kg^{-1} from Abuloma and Okujagu-ama rivers (Table 4). The highest average concentration of 105.45 mg kg^{-1} was recorded for Benzo[k]Fluoranthene in soil sample around Abuloma river.

US. DEQ[28] has classified Benzo[k]fluoranthene as group B2 probable human carcinogen upon no human data and sufficient data from animal assays and has also been reported to be a constituent of combinations that have been linked with human cancer. Some PAH components with the highest risk mostly in times of excessive exposure such as; benzo[a]anthracene, chrysene, benzo[b]fluoranthene, indeno[1,3-c,d]pyrene, dibenz[a,h]anthracene and benzo[k]fluoranthene[29] were detected in soil samples in this study. Cal-EPA identified that out of these six high risk PAHs detected in the soil samples, most of them (which are benzo[a]anthracene, benzo[b]fluoranthene, chrysene and benzo[k]fluoranthene) are associated with carcinogenesis[30]. The total concentration of PAHs in soil samples around Abuloma and Okujagu-ama rivers were 915.75±237.59 and 14.06±1.17 mg kg^{-1}, respectively. The total mean concentration of PAHs in soil samples around Abuloma and Okujagu-ama rivers varied significantly ($p<0.05$) where soil samples from Abuloma recorded a very high PAHs concentration, this may be due to dense industrial activities, careless deposition of petroleum products and other commercial products on the soil and also, the scavenging activities around the refuse dumpsite located close to the river bank. The soil from Okujagu-ama showed the presence of a relative amount of PAHs concentration, even though the soil in this area was not exposed to the high levels of pollution as observed at Abuloma Jetty, this may be from deposition from sewage, flowing irrigation water, some waste products from compost, carbonization of plant material or unspecified diffused sources in soil[31]. The total mean PAHs in soil samples from Abuloma and Okujagu-ama exceeded DPR's permissible limits[32] of 1000 μg kg^{-1}. Nwaichi *et al.*[33] reported that the total mean concentration of PAHs in soil samples from two oil impacted sites in Delta state, Nigeria (Ekore and Uduvwoku) was 1261.806 and 4613.020 mg kg^{-1} DW, respectively, these values were higher than that the values in this study. Abbas and Brack[34] studied PAH levels in soils of a Niger Delta community and found dominant PAHs viz. pyrene, naphthalene and Benzo(k)fluoranthene which concentration ranged between 182±112 to 433±236 μg kg^{-1} which was lower than the value obtained in this study.

The values of the LWM-PAH/HMW-PAH ratio in soil samples from the two sites indicated that the number of HMW-PAHs obtained was higher than the LMW-PAHs with ratios of 0.50 and 0.00 from Abuloma and Okujagu-ama, respectively. In the soil sample from Okujagu-ama, none of the LMW-PAHs was detected. This may be attributed to the fact that LMW-PAHs are differently degraded as PAHs are transported and submerged into sediments[35]. The LMW-PAH/HMW-PAH ratios in the soil samples from the both sites were <1 which indicated that the PAHs contamination in these samples are mostly from pyrogenic sources.

The BaA/(BaA+Chry) ratios in soil samples from Abuloma and Okujagu-ama were 0.34 and 0.42, respectively. The BaA/(BaA+Chry) ratio obtained in soil samples from Abuloma Jetty was between 0.2-0.35 which indicated a mixed petrogenic and pyrogenic origin. This suggested that the PAHs contamination in the soil samples from Abuloma Jetty may be as a result of the emission of by-products from the incomplete combustion of fuel from sea vessels at the river bank and also from the direct discharge of petrogenic sources like spilled petroleum products during off-loading. BaA/(BaA+Chry) ratio of soil samples from Okujagu-ama was >0.35 indicated a pyrogenic origin, this suggested that it may be from deposition from sewage, irrigation water, particulate waste or products from compost.

Polycyclic Aromatic Hydrocarbons (PAHs) in Tilapia, Mullet fish and Crab samples: A number of carcinogenic PAHs have also been noticed in water and food samples[36]. Ajiboye *et al.*[37] also reported that the presence of these substance in the environment has turned into an issue which concerns the whole world because the main route through which man is exposed to this contamination is by consumption of affected food, water and marine products such as fish.

The total mean concentrations of PAHs in all the fish and seafood samples from Abuloma were significantly higher than samples from Okujagu-ama (Control). This may be as a result of the high industrial activities at the Abuloma Jetty which introduces PAHs into the water where the fish and seafood live. Tilapia fish collected from Abuloma river recorded the highest (Table 5) total PAHs concentration accumulated compared to Mullet fish (Table 6) and Crab samples (Table 7) from the two sites. Differences in feeding preferences and general behavior can be said to be the reason for variations in PAH bioaccumulation among these species[38] and even the way the various species feed[39]. The highest average individual concentration of 6.91±0.54 mg kg^{-1} was recorded for carcinogenic Benzo[b]Fluoranthene in Tilapia fish from Abuloma river. Indeno[1,2,3-cd]pyrene levels in the three species from the two sites did not exceed the European Union (EU) limit of 5 μg g^{-1} wet wt.[40] Benzo[a]Pyrene known to be seriously carcinogenic was observed in Tilapia and Mullet fish samples from Abuloma river, but was not found in all the samples from Okujagu-ama. The values of the PAHs concentration of the three species were lower than the amount of 70.440 μg g^{-1} wet wt. which was obtained among five species of fish samples collected along the harbor line,

Table 5: Mean concentrations of PAHs in Tilapia fish (*Oreochromis niloticus)* from Abuloma Jetty and Okujagu-ama rivers

PAHs (mg kg^{-1})	Abuloma jetty	Okujagu ama
Naphthalene	2.33±0.21	BDL
Acenaphthylene	4.18±0.21	BDL
Acenaphthene	1.88±0.31	BDL
Fluorene	5.31±0.05	BDL
Anthracene	BDL	BDL
Phenanthrene	2.17±0.21	BDL
Fluoranthene	BDL	0.93±0.12
Pyrene	2.33±1.13	BDL
Benz[a]anthracene	1.52±0.14	BDL
Chrysene	1.60±0.26	BDL
Benzo[b]Fluoranthene	2.03±0.37	BDL
Benzo[k]Fluoranthene	6.91±0.54[a]	1.60±0.29[a]
Benzo[a]pyrene	3.17±0.06	BDL
Indeno[1,2,3-cd]Pyrene	2.24±0.26	BDL
Dibenz[a, h]anthracene	BDL	BDL
Benzo[g, h, i]perylene	BDL	1.50±0.26
Total PAHs	35.67±3.75[a]	4.03±0.67[a]
LMW-PAH/HMW-PAH ratio	0.80	0.00
BaA/(BaA+Chry) ratio	0.49	0.00

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg kg^{-1}

Table 6: Mean concentrations of PAHs in Mullet fish (*Liza falcipinus*) samples from Abuloma Jetty and Okujagu-ama rivers

PAHs (mg kg^{-1})	Abuloma jetty	Okujagu ama
Naphthalene	BDL	BDL
Acenaphthylene	4.23±0.52	BDL
Acenaphthene	BDL	1.50±0.06
Fluorene	3.38±0.29	BDL
Anthracene	BDL	BDL
Phenanthrene	BDL	BDL
Fluoranthene	BDL	1.67±0.20
Pyrene	1.82±0.35	BDL
Benz[a]anthracene	2.51±0.26	BDL
Chrysene	5.68±0.34	BDL
Benzo[b]Fluoranthene	1.61±0.07	BDL
Benzo[k]Fluoranthene	5.28±0.44	BDL
Benzo[a]pyrene	2.84±0.19	BDL
Indeno[1,2,3-cd]Pyrene	4.29±0.36	BDL
Dibenz[a, h]anthracene	3.39±0.27	BDL
Benzo[g, h, i]perylene	BDL	BDL
Total PAHs	35.03±3.09[a]	3.17±0.26[a]
LMW-PAH/HMW-PAH ratio	0.28	0.9O
BaA/(BaA+Chry) ratio	0.31	0.00

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg kg^{-1}

Mumbai[41] and greater than 3.365 ppm obtained in edible fishes of the Gomti river, Lucknow, India[42]. Nwaichi and Ntorgbo[11] reported that PAHs concentration in fish and sea foods from 3 coastal rivers in Niger Delta region (Sime, Kporghor and Iko) ranged from 3.670±0.050 to 171.900±0.450 µg g^{-1}. These values were within the same range for values obtained in this study.

Table 7: Mean concentrations of PAHs in Crab (*Callinectes amnicola)* samples from Abuloma Jetty and Okujagu-ama rivers

PAHs (mg kg^{-1})	Abuloma jetty	Okujagu ama
Naphthalene	5.11±0.37	BDL
Acenaphthylene	3.63±0.07	BDL
Acenaphthene	3.09±0.06[a]	1.52±0.08[a]
Fluorene	2.72±0.51	BDL
Anthracene	BDL	BDL
Phenanthrene	3.45±0.27	BDL
Fluoranthene	BDL	1.97±0.07
Pyrene	BDL	BDL
Benz[a]anthracene	4.84±0.19	BDL
Chrysene	3.67±0.79	BDL
Benzo[b]Fluoranthene	4.45±0.32	BDL
Benzo[k]Fluoranthene	BDL	BDL
Benzo[a]pyrene	BDL	BDL
Indeno[1,2,3-cd]Pyrene	BDL	1.62±0.23
Dibenz[a, h]anthracene	2.31±0.12	BDL
Benzo[g, h, i]perylene	BDL	BDL
Total PAHs	33.27±2.70[a]	5.11±0.38[a]
LMW-PAH/HMW-PAH ratio	1.18	0.42
BaA/(BaA+Chry) ratio	0.57	0.00

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg kg^{-1}

BaA/(BaA+Chry) ratio and LMW-PAH/HMW-PAH ratio were adopted to analyze the possible sources of PAHs in the study samples. This approach has been used to characterize the sources of PAHs in the fish and seafood analyzed.

The LMW-PAH/HMW-PAH ratio of all the samples except crab from Abuloma river were <1 showing that the sources of PAHs contamination were mainly pyrogenic, but the LMW-PAH/HMW-PAH ratio for crab samples from Abuloma Jetty was >1, which indicated a petrogenic source of contamination. The crab sample showed a different source of contamination which may be due to the differences in the PAHs accumulation, feeding preference and general behaviour of different organisms. This high concentration of HMW PAHs indicated a predominant pyrolytic origin for the PAH pollution suggested a high level of incomplete combustion of fuel from sea vessels and boats. Nwaichi and Ntorgbo[11] reported that the LMW-PAH/HMW-PAH ratio for all the fish and seafoods harvested from 3 coastal rivers in Niger Delta region were <1 indicated anthropogenic origin of PAHs. Nkpaa *et al.*[43] also reported that the LMW-PAH/HMW-PAH ratio obtained across all the species collected in the Ogonil and coastal environment was <1 indicated anthropogenic origin of PAHs. The observed BaA/(BaA+Chry) ratio for Tilapia fish and Crab from Abuloma river were >0.35 indicated a pyrogenic origin. BaA/(BaA+Chry) ratios in all the samples from Okujagu-ama were <0.20 indicated a petrogenic origin of PAHs contamination. BaA/(BaA+Chry) ratio in Mullet sample from Abuloma river was between 0.20-0.35 indicated a mixed petrogenic and pyrogenic source of PAHs contamination.

Table 8: Total Petroleum Hydrocarbon (TPH) concentrations in water samples from Abuloma and Okujagu-ama (control) rivers

TPH components (mg L^{-1})	Abuloma jetty	Okujagu ama
C-8	37.18±4.38[a]	3.65±0.06[a]
C-9	BDL	12.84±0.33
C-10	30.75±5.92	BDL
C-11	BDL	BDL
C-12	45.30±1.60	BDL
C-13	BDL	BDL
C-14	45.68±7.45[a]	8.76±0.11[a]
C-15	BDL	BDL
C-16	55.81±1.00	BDL
C-17	BDL	16.75±0.45
C-18	26.53±2.27[a]	10.13±0.40[a]
C-19	BDL	BDL
C-20	33.52±1.68[a]	10.15±0.27[a]
C-21	BDL	BDL
C-22	26.51±4.03[a]	6.35±0.40[a]
C-23	BDL	BDL
C-24	15.92±0.77	BDL
C-25	BDL	BDL
C-26	16.70±1.07	BDL
C-27	BDL	BDL
C-28	53.21±4.45	BDL
C-29	BDL	BDL
C-30	BDL	BDL
C-31	BDL	BDL
C-32	BDL	BDL
C-33	BDL	BDL
C-34	BDL	BDL
C-35	BDL	BDL
C-36	BDL	BDL
C-37	BDL	BDL
C-38	BDL	BDL
Total	387.11±34.62[a]	68.63±2.02[a]

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates. Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other ($p<0.05$), BDL: Below detection limits of 0.0001 mg L^{-1}

Total Petroleum Hydrocarbon (TPH) in water samples studied: A total of 11 and 7 TPH were detected in the water from Abuloma and Okujagu-ama, respectively (Table 8). Average concentrations of TPH ranged from below detection limits of 0.0001 to 53.21±4.45 mg L^{-1}. The highest average concentration from Abuloma was 53.21±4.45 mg L^{-1} (C-28) and 16.75±0.45 mg L^{-1} (C-17) from Okujagu-ama. The total TPH concentrations in water were 387.11±34.62 mg L^{-1} and 68.63±2.02 mg L^{-1} for Abuloma and Okujagu-ama, respectively. The TPH content was greater in water from Abuloma than in Okujagu-ama and varied significantly ($p<0.05$). These levels of TPH showed that the water was contaminated by petroleum products. The high value of TPH at Abuloma Jetty may be attributed to washing of petroleum products from transporting vessel tanks into the river, leaked petroleum products into the river during loading and offloading and also other industrial activities. This can result to decline in the amount of oxygen because there will be a reduction in gaseous diffusion through the surface covered with oil and thereby affecting aquatic organisms in the affected area[44]. From this study, the highest value of TPH concentration was observed to be between C-10 to C-24 showed that the water contains much of diesel range organics and a little quantity of some other crude oil components. When there is a high level of TPH in a water body in the aquatic environment, the organisms such as; fish and crabs present tends to ingest and absorb it and later transfer (containing all its health complications) it to higher predators in the food chain such as man. Therefore, the environmental regulators and oil industry players should put into place practical steps to save the aquatic biota as well as maintain the sustainability of the entire environment. Ibezue[45] recorded a mean TPH concentration of 272.68 and 48.51 mg L^{-1} for marine waters of Biarra and Bodo at Gokana area of the Niger Delta, respectively, which were contaminated by oil spillage. These values were lower than the average TPH concentration obtained in this study. Adewuyi and Olowu[46] also reported TPH values ranging from 20.34±1.79 to 27.40±5.32 mg L^{-1} in surface water from a the vicinity of NNPC oil depot in Apata, Ibadan and these values were far lower than the values obtained in this study.

Total Petroleum Hydrocarbon (TPH) in soil samples studied: A total of 18 and 7 TPH were detected in the soil samples analyzed from Abuloma and Okujagu-ama, respectively (Table 9). Average concentrations of TPH ranged from below detection limits of 0.0001 to 828.52±19.78 mg kg^{-1}. The highest average concentration from Abuloma was 828.52±19.78 (C-32) and 22.45±0.67 mg kg^{-1} (C-14) from Okujagu-ama. The total TPH concentrations in the soil were 9913.68±560.74 and 71.90±4.87 mg kg^{-1} for Abuloma and Okujagu-ama samples, respectively. The TPH level in soil from Abuloma was greater than Okujagu-ama and there was a statistically significant difference ($p<0.05$) between the total TPH for the two sites. The high value of TPH at Abuloma Jetty may be due to the spillage of petroleum products on the soil during transportation of these products on daily basis. The TPH values obtained in this study exceeded 1000 mg kg^{-1} set by the Environmental Guidelines and standards for the Petroleum Industry in Nigeria (EGASPIN) intervention value for mineral oil (petroleum) recommended by Department of Petroleum Resources[32] which indicated that the soil around Abuloma Jetty was contaminated with mineral oil (petroleum). From the values obtained in the study, when compared to the intervention value of EGASPIN[32], it showed that the soil has been contaminated and can lead to health impairment in

Table 9: Total Petroleum Hydrocarbon (TPH) concentrations in soil samples from Abuloma and Okujagu-ama (control) rivers

TPH components (mg kg^{-1})	Abuloma jetty	Okujagu ama
C-8	585.44±1.22[a]	14.52±0.41[a]
C-9	390.59±30.48	BDL
C-10	660.37±57.91[a]	6.37±0.80[a]
C-11	BDL	6.54±1.34
C-12	205.26±27.53[a]	3.55±0.18[a]
C-13	119.51±2.65	BDL
C-14	BDL	22.45±0.67
C-15	87.78±1.34	BDL
C-16	522.95±29.95[a]	12.43±1.02[a]
C-17	BDL	BDL
C-18	445.74±28.67	BDL
C-19	BDL	BDL
C-20	348.56±22.34[a]	6.04±0.45[a]
C-21	576.57±64.69	BDL
C-22	1166.14±57.57	BDL
C-23	BDL	BDL
C-24	226.25±7.57	BDL
C-25	BDL	BDL
C-26	678.91±23.30	BDL
C-27	1447.98±8.56	BDL
C-28	361.78±30.13	BDL
C-29	BDL	BDL
C-30	732.50±27.40	BDL
C-31	BDL	BDL
C-32	828.52±19.78	BDL
C-33	BDL	BDL
C-34	528.83±61.74	BDL
C-35	BDL	BDL
C-36	BDL	BDL
C-37	BDL	BDL
C-38	BDL	BDL
Total	9913.68±560.74[a]	71.90±4.87[a]

Values are expressed as Mean±Standard Error of Mean (SEM) of three replicates (n = 3), Mean values across rows with the same superscript letter (a) are statistically significant while those without superscripts are statistically not significant when compared to each other (p<0.05), BDL: Below detection limits of 0.0001 mg kg^{-1}

humans and other harmful circumstances in plant and animals. Presence of TPH compounds can affect the blood, immune system, lungs, skin and eyes[10]. High amount of TPH in Abuloma soil sample showed that the soil is contaminated and so most of these contaminants can be washed into the river by flowing water during the rains. The TPH compounds can be transmitted to man through the food chain and thereby bringing about the harmful effects which can be detrimental to health. Alinnor and Nwachukwu[47] reported that soil samples from five communities in Rivers state, Nigeria, were contaminated with TPH concentrations of 1534.7, 1438.0 and 1651.0 mg kg^{-1} at depths of 0.0- 0.5 , 0.5-1.0 and 1.0-2.0 m, respectively, which were much lower than values obtained in this study. Ibezue[45] recorded high concentrations of TPH in soil samples contaminated by oil spillage in Gokana area of Niger Delta region in Rivers state of Nigeria. The soil samples recorded high mean TPH concentrations of 13949.42 and 8279.35 mg kg^{-1} for soil from Biara and Bodo all in Gokana, respectively. These values fall in the same range with the concentration of TPH in soil samples obtained in this study. The average TPH concentration in this study was found to be lower than that obtained in soil that is close to Benin river in lubricating oil producing factory area in Koko (38700±12000 mg kg^{-1}) by Akporido and Asagba[48]. Nwaichi *et al.*[49] recorded a TPH concentration of 3830±19.6 mg kg^{-1} for soil samples from oil polluted agricultural zone in Gokana, Rivers state and this was lower than TPH concentration obtained from soil samples in this study.

CONCLUSION

Water, soil and three frequently consumed fishes and seafood samples were gotten from high industrial activity area (Abuloma Jetty) and non-industrial activity area (Okujagu-ama) and analyzed for this study. Physico-chemical parameters (which included biological oxygen demand, dissolved oxygen, total suspended solids, total dissolved solids, conductivity, pH and temperature), TPH, PAHs and BTEX levels were determined in water and soil samples while, PAHs bioaccumulation only was determined in studied fishes and seafood. Revealed in this study also are some of the physico-chemical parameters like; BOD, TDS, conductivity and temperature which fell within guideline limits except Dissolved Oxygen (DO), TSS and pH that were below the guideline limits used. However, a larger number of values obtained for the physico-chemical parameters of samples from Abuloma Jetty were comparatively greater than that of Okujagu-ama. PAHs and TPH concentrations found in soil, water, Tilapia fish, Mullet fish and Crab were comparatively higher for samples from Abuloma Jetty than Okujagu-ama which can lead to exposure of the population that feeds on these aquatic organisms to several health challenges.

SIGNIFICANCE STATEMENT

An indication from the diagnostic PAH ratios used showed that the PAHs contamination were majorly from pyrogenic (pyrolytic) sources and a smaller portion from petrogenic sources. The industrial activities carried out at the Jetty pose a great threat to the water body, soil around the river and also the aquatic life. The companies involved in these industrial activities should carry out some clean up processes in other to reduce the level of PAHs and TPH contamination. The government should monitor the activities of the companies operating at this Jetty to regulate the rate at which they cause pollution to the environment.

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