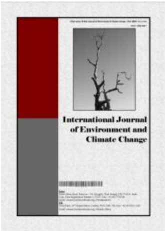

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Distribution of Polycyclic Aromatic Hydrocarbons in Surface Water and Fishes in Bodo/Bonny River Nigeria

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Authors' contributions

This work was carried out in collaboration between both all authors. Both authors read and approved the final manuscript.

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ABSTRACT

Several industrial activities around the Niger Delta region have contributed to the widespread contamination of marine ecosystems with organochlorine compounds (OCs), petroleum products that are a source of polycyclic aromatic hydrocarbons (PAHs). These pollutants tend to be persistent in the environment and are also often highly toxic to the biota. The study was therefore, aimed at determining the concentrations of organic pollutants (PAHS) in the Bodo/Bonny coastal waters and their effect on the marine ecosystems. This is exacerbated by the risks posed by polycyclic aromatic hydrocarbons (PAHs) in oil spilled environments. Surface water, sediment and fish samples were collected from different sampling stations along the river and analyzed using standard analytical methods. Sampling of surface water was done on Link fish pond which served as control. The results of the value of TPH ranged from 0.31 to 40.85 mg/l, PAHs range from 2.06 to 2.73 mg/l and BTEX ranged from 0.043 to 0.081 mg/l. The Total Petroleum hydrocarbon (TPH) values obtained were above DPR permissible Limit of 20 mg/l in all the stations. However, values

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of individual polycyclic aromatic hydrocarbons show that Benzo(a) anthracene had the highest concentration especially in all the surface water stations sampled. Also results showed a presence of carcinogenic PAHs in the fish tissues. This still poses a danger if accumulation was to take place over a long period of time. The values obtained from this study stations also exceeded the WHO quality criteria for drinking, aquatic life support and recreation. This reveals that Bodo/Bonny River is under pollution threat and underscore the need for early remediation if adverse health defects are to be prevented.

Keywords: Water pollution; oil spillage; health defects; fishes; polycyclic aromatic hydrocarbons; Bodo/Bonny river.

1. INTRODUCTION

The surface water resources of Bodo/Bonny communities in Rivers State suffers regular pollution of its ecosystem due to increase in crude oil exploration, refining and activities of other industrial establishments operating within the coastal areas of the Ogoniland of the Niger Delta region of Nigeria. This have resulted in the wide scale contamination of most of its creeks, swamps and rivers with hydrocarbons and dispersant products. Despite community concerns about their health and the damage of their water bodies, water pollution in the Niger Delta is commonly attributed to activities related to oil exploitation; resulting in serious threat to public health and the ecosystems. Oil spill poses a major threat to the environment in Nigeria. Polycyclic aromatic hydrocarbons (PAHs) are relatively stable constituents of petroleum, and from the environmental aspects, they are probably the most important analytes because many of these compounds are potential or proven carcinogens. Unfortunately, their low aqueous solubility, limited volatility and recalcitrance toward degradation allows PAHs to accumulate to levels at which they may exert toxic effects upon the environment [1]. Total Petroleum Hydrocarbons (TPHs) are toxic to the aquatic fauna and flora because they cause severe damage to benthic species, impaired growth and survival, interference with breeding ability, low reproductive ability and destruction of spawning and nursery grounds of most species in the sea [2]. Polycyclic Aromatic Hydrocarbons (PAHs) are a class of diverse organic compounds containing two or more fused benzene rings of Carbon and Hydrocarbon atoms [3]. They are generated primarily during the incomplete combustion of organic materials such as coal, oil, petrol and wood [3,4].

Sources of PAHs in the environment can be natural, pyrolytic, anthropogenic or petrogenic. Pyrolytic and anthropogenic PAHs are formed as a result of incomplete combustion of carbon sources whereas petrogenic PAHs are mainly obtained from crude oil or organic carbon combustion related activities. The International Union of Pure and Applied Chemistry recorded the simplest PAHs as phenanthrene and anthracene. These are isomers of three fused rings ($C_{14}H_{10}$). Other members of the PAHs clade may be made up of four-, five-, six- or seven-member rings with the five to six benzene ring member being the most common.

Owing to their low solubility and high affinity for particulate matters, PAHs are not usually found in stable concentrations and their presence in ground water, streams, soil and sediment is a marker of pollution [5]. Because PAHs do not dissolve easily in water and generally do not burn, they can persist in the environment for months to years, though according to some members can undergo volatilization, photo-oxidation, chemical oxidation, bioaccumulation, adsorption to soil particles, leaching and microbial degradation. Due to their lipophyilic nature, PAHs have a high potential for biomagnifications through trophic transfer [6].

The hydrocarbon contamination of habitats constitutes public health and socioeconomic hazards [6]. The hydrocarbons so discharged may also pose serious aquatic toxicity problems [6]. Hydrocarbons may even affect the physiological process of microorganisms and other aquatic organisms including the fauna and flora. It is speculated that the discharged refinery effluent or waste in the long run will be a potential hazard due to accumulation of mutagenic and carcinogenic aromatic hydrocarbon in soil and water resources through storm water run-off [7]. Seepage during storage is also another major problem because it is responsible for ground water contamination. Further, many researchers have also shown that PAHs may also cause harmful effects on body fluid, skin, and immunity. PAH is also an effective carcinogen and people who are exposed long time to PAHs and other related compounds having developed cancer. Laboratory animals developed lung cancer when exposed to PAHs compounds while breathing, stomach cancer. Additionally, PAHs shows putative estrogenic and anti-estrogenic properties in human body. Human exposed to PAHs by ingestion of contaminated meat or vegetables can cause intestine enterocytes and liver hepatocytes in both mice and humans [8]. Significant developmental, reproductive and immune effects from low-level exposure in numerous animals were studied by Vom Saal and Hughes [9]. Toxicity from hydrocarbon exposure can lead to different syndromes, especially on which organ system is predominantly involved. Organ systems that can be affected by hydrocarbons include the pulmonary especially aspiration which leads severe pneumonitis, neurological, cardiac, gastrointestinal, hepatic, renal, dermatologic, and hematological systems. Therefore, the aim of the study was to determine the presence and distribution in surface waters and fishes in Bodo/Bonny coastal waters.

2. MATERIALS AND METHODS

2.1 Sampling Location

Samples for this study were collected from four different stations in Bodo/Bonny River in Gokana Local Government Area of Rivers State. Control samples were collected from Link fish pond located at a distance of 100 meters away from a location where there was no record of crude oil pollution within the river environment. Surface water, sediment and fish samples were collected from 5 stations (BBW1, BBW2, BBW3, BBW4 and LFPW5) with LFPW5 serving as control (Fig 1; Table 1). The sampling stations were chosen based on an experimental scheme design following ecological settings and human activities in the area. Bodo Creek is characterized by low tidal energy current, making its swamps and canals exceptional breeding grounds for a vast variety of fish and shellfish. It also provides an excellent habitat for periwinkles (*Tympanotonus fuscatus; Tympanotonus fuscatus varradula; Pachymelania aurita; Pachymelania fusca*) [10]. The original diversity of shellfish found in the Creek included bloody cockle (Senilia senillis), oyster (Crassostrea gasar), swimming crab (Callinectis amnicola), razor clam (Tagelus adansonii), land crab (Cardisoma amatum), and mangrove purple hairy crab (Goniopsis pelli) [10]. The Bodo/Bonny River meet several socio-economic needs including aquaculture, fishing, sand dredging and drainage of the various towns and villages bordering it.

2.2 Sample Collection

Samples used for this study were surface water, sediment and fishes from the river and Fish Link pond as control (Plates 1 and 2). Dead floating fishes (Pseudotolithus elongatus) were collected from the seashore during the sampling period. Sampling was done in the mornings between 10 am-12 noon each day for a period of 12 months covering both wet and dry seasons, at an interval of once a month. Sample were collected in duplicates from each location, monthly at floodtide in the river.

2.3 Surface Water

Surface water samples were collected using the method of Adesemoye et al [11]. Sterile 1.5 litre bottles were used to aseptically collect the surface water. The samples were collected at four different points (about 50m apart) in the direction of water flow while the fifth sample served as control from a Link fish pond. To collect the surface water, base of the sterilized sample container was held with one hand, plunged about 30 cm below the water surface with the mouth of the sample container positioned in an opposite direction to water flow [12]. About 500 ml of the sample collected from each station was pooled together to get a composite sample. During the period of sampling, there was fish mortality in the river floating, dead, so the fish samples were collected for analysis (Figs 2 and 3). After collection, the samples were placed in a cooler containing ice blocks and transported immediately to the laboratory for analysis.

Table 1. GPS coordinates of sampling sites across sampling stations

Sampling Stations (BBW)	Northing	Easting	Sample Type
Point 1	$4^0$36’ 5”N	$7^0$16’34” E	Water and Fish
Point 2	$4^0$35’56”N	$7^0$16’30”E	water
Point 3	$4^0$36’6”N	$7^0$16’25” E	water
Point 4	$4^0$36’27”N	$7^0$15’51”E	water
Point 5 (control)	$4^0$37’10” N	$7^0$16’1” E	Link fish pond ,water

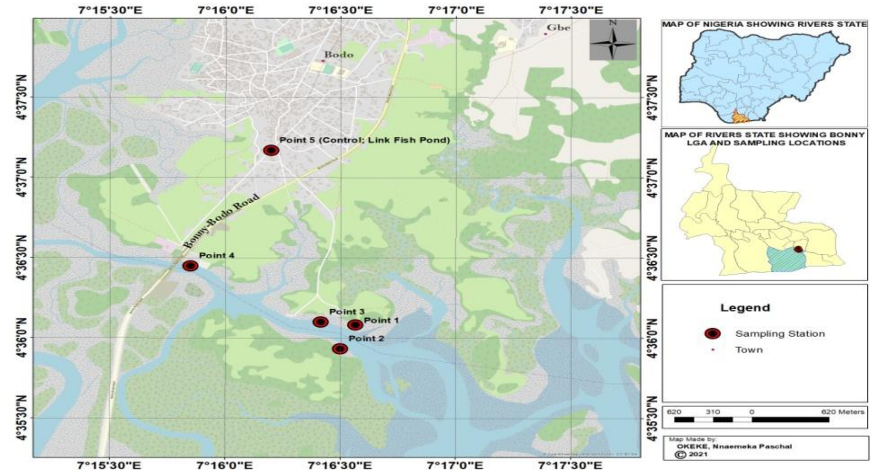

Fig. 1. Map indicating Sampling Stations (Bodo/Bonny River)

Fig. 2. Fish Samples (*Pseudotolithus elongatus*) from the Bodo/Bonny River

Fig. 3. Dead Fishes (*Pseudotolithus elongatus*) in the Bodo/Bonny River due to asphyxiation

2.4 Determination of Total Petroleum Hydrocarbon (TPH) and Polycyclic Aromatic Hydrocarbons

2.4.1 Extraction process

The analyses of TPH and PAHs with water samples was performed using the cold extraction method as adopted by Olufemi et al [13] and Inengite et al [14]. Filtered water samples were subjected to liquid-liquid extraction while sediment samples were air dried, homogenized, sieved and extracted using hot extraction (sohxlet extractor). A gravimetric method was used to determine oil and grease [15,16]. Extracts were subjected to silica gel clean-up to remove polar organic substances leaving non- polar hydrocarbon in the solvent. Gravimetric method was used to determine the TPH. The TPH was analysed for petroleum hydrocarbon using gas chromatography (GC). Analyses was performed with a Perkin model 5890 gas chromatography equipped with Ni 63 electron capture detector. A low polar HP-5 column of 30m length, 0.32 mm i.d and 0.25µm film thickness was used. Nitrogen was used as the carrier gas at a flow rate of 50ml/s. Data were processed using an HP 3396 integrator. The operating parameters were as follows: injector temperature set 240 and 310^{0}C for the detector, the oven temp was programmed at 150^{0}C initially (5 minutes hold) and increased to 310^{0}C at 5^{0}C/minute to give an analysis period of 35 minutes. Mass spectrometer coupled with GC is preferred because it offers robust identification of the analyte compounds both by retention time and mass spectrum, with additional structural information. The fish sample was determined using the method of Fernandez-Tajes et al [17]. Before extraction 5g sample of freeze-dried fish was spiked with 5µL of PAH-LCs standard. The extraction was carried out by MAE with 15 ml of acetone-hexane (1:1 v/v). The extracts were cleaned with a 22g aluminum oxide (10% deactivated) columns and eluted with 90 ml of hexane. Then they were concentrated in a rotary evaporator, dried under nitrogen stream and dissolved in 100µL of hexane.

2.4.2 Measurement of polycyclic aromatic hydrocarbons

The Polycyclic Aromatic Hydrocarbon content of sample were determined through the principle of Gas Chromatography by flame ionization detection as sample extracts are being forced through an immobile, inert stationary phase (1,3-dimethyl siloxane) and components of low solubility take a longer elution time leading to the differential mobilities of the fractional components of the polycyclic aromatic hydrocarbon (PAHs). Samples were automatically detected as they emerge from the column (at a constant flow rate) by the FID detector whose response was dependent upon the composition of the respective constituent fractions.

2.4.3 Determination of TPH and PAHs concentration in the extract

Gas chromatography with flame ionization detection was the method used for the analysis. Helium gas which flows at the rate of 14.81psi was steamed as mobile phase, while an agilet HP5 gas chromatographic capillary column, with 100% 1, 3 dimethylsiloxane stationary phase material with a temperature of range of -60^{0}C to 325^{0}C was used for the separation of vapour constituents of different hydrocarbon fractions. Hydrogen and air at flow rate of 30psi served ignition gases [18].

3. RESULTS

3.1 Total Petroleum Hydrocarbon Content of Bodo/ Bonny Surface River water and the Link Fish Pond

The results of Total Petroleum hydrocarbon (TPH), Polycyclic Aromatic Hydrocarbons (PAHs) and BTEX (Benzene, Toulene, Ethyl Benzene and Xylene) obtained from the various sampling points of the River water and the Link fish pond are presented in Table 2. The value of TPH ranged from 0.31 to 40.85 mg/l, PAHs range from 2.06 to 2.73 mg/l and BTEX ranged from 0.043 to 0.081 mg/l. The Total Petroleum hydrocarbon (TPH) values obtained were above DPR permissible limit of 20 mg/l in all the stations (BBW1 – BBW4), except in the Link fish pond water which was within limit. However, values of individual Polycyclic Aromatic Hydrocarbons show that Benzo(a) anthracene had the highest concentration especially in the various stations BBW1, BBW3 and BBW4 (Fig. 4) while BTEX (Benzene, Toulene, Ethyl Benzene and Xylene) were highest in station BBW3, followed by BBW4 and lowest in BBW2 (Fig. 5).

The results of Total Petroleum Hydrocarbon (TPH), Polycyclic Aromatic Hydrocarbons (PAHs) and BTEX obtained from both fresh and smoked fish samples of the surface River water and the Link fish pond are presented in Table 2. The value of TPH for Fresh fish had 0.232 mg/kg while the dry fish recorded 0.294 mg/kg. PAHs had similar pattern with fresh fish having 0.004 mg/kg and dry fish samples had 0.005 mg/kg. BTEX values had insignificant values for both fish samples.

Table 2. Hydrocarbon content of River water and the Link fish Pond

Hydrocarbon Content	Stations					
	BBW1	BBW2	BBW3	BBW4	LFPW5	DPR Limit for Brackish/ Salinewater
TPH (mg/L)	40.753	39.045	40.850	36.653	0.31	20.00
PAHs (mg/L)	2.729	2.312	2.152	2.06	<0.00	-
BTEX (mg/L)	0.0501	0.0434	0.0807	0.0639	<0.00	-

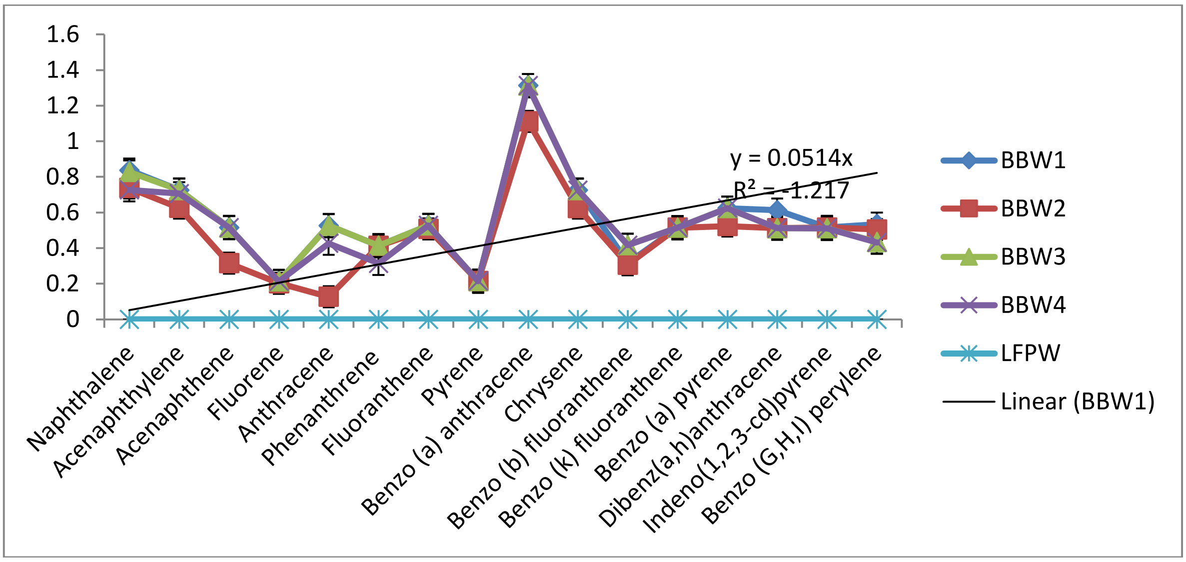

Fig. 4. Concentration of polycyclic aromatic hydrocarbons (PAHs) from various points of river water and the Link fish Pond

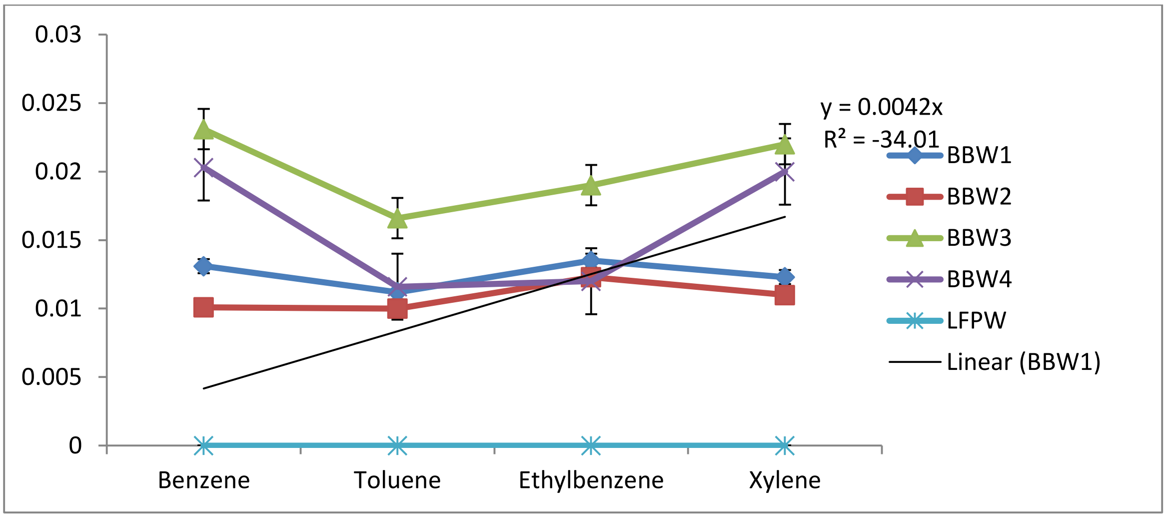

Fig. 5. Concentration of BTEX from various points of the river water and the Link fish Pond

Table 3. Total petroleum content in fish samples from river water and the link fish pond

Sample	TPH (mg/kg)	PAHs (mg/kg)	BTEX (mg/kg)
River Water Fresh Fish	0.232	0.004	<0.001
River Water Dry Fish	0.294	0.005	<0.001
Link Fish pond Fresh Fish	0.002	<0.002	<0.001
Link Fish pond Dry Fish	0.003	<0.002	<0.001

4. DISCUSSION

Total Petroleum Hydrocarbons (TPH) and Polycyclic aromatic hydrocarbons (PAHs) concentrations can be potentially toxic and dangerous when they cross the food chain into the biological system. PAHs are classified as environmentally hazardous pollutants due to their known hydrophobic, mutagenic, and carcinogenic characteristics [19]. However, oil production activities generally pollute both surface and groundwater with benzene, toulene, ethylbenezene and xylene (BTEX) as well as other toxic chemicals including toxic PAHs [10]. This could account for the presence of BTEX (Benzene) and PAHs especially Benzo (a) anthracene in the surface water samples in high amounts in this study. Water pollution remains one of the major environmental public health problems arising from extensive oil operations causing severe impairment due to surface and underground water contamination [20]. The environmental health consequences are far reaching, particularly in most areas of the Niger Delta where residents are dependent on very shallow wells connecting aquifers [21]. This implies that many host communities in the Niger Delta including Ogoni, will continue to suffer diverse range of illnesses arising from exposure to oil pollution, such as blood disorders, cancers of different organs, negative reproductive conditions, impaired disease immunity and growth; respiratory diseases, and undesirable physiological responses [22]. Gobo [23] also reported that the prevalence of diarrhea may be severe because of the consumption of fish and other animals contaminated by hydrocarbons contained in the spilled oil environment. This may result from the bioaccumulation of hydrocarbons and heavy metals, which may occur in toxic amounts in major high protein content sea foods, such as periwinkle (*Tympanotonus fuscatus*) and the Mudskipper (*Periophtalus papillio*) [24]. In this study TPH and PAHs values were recorded for both fresh and smoked fishes from the river and such values may predispose those who depend on such food products as sources of proteins and other essential nutrients to public health risk. Benzene is a “known carcinogen” and can cause cancer to humans if they are contained in such fish products [25]. Water contaminated by oil leaks containing Benzene can poison streams serving as drinking water sources and cause significant detriment to marine/aquatic life [25]. Fishes are continuously exposed to pollutants in the water, due to continuous water flow through gills and food and it has been discovered that fish are able to accumulate several folds higher concentration of pollutants than the surrounding water [26]. Therefore the impact of pollutants on aquatic organisms is due to the movement of these substances from various point sources which give rise to coincidental mixtures in the ecosystem [26] thus posing a serious threat to aquatic organisms especially to fishes which constitute one of the major sources of protein-rich food for mankind. In this study, the concentrations of PAHs; TPH and BTEX however, are reflective that pollution could affect aquatic lives via bioaccumulation in oil polluted areas. This condition could be attributed to mass death of fishes ((*Pseudotolithus elongatus*) in the Bodo/Bonny River due to smothering or asphyxiation of the organisms in water by oil coating, thereby causing death. These results agree with the report of Ikue et al [27] on the accumulation of PAHs in tissues of catfish *Chrysichthys grodidatatus* from crude oil tainted water of Ogoni, River State, Nigeria. Similar reports have been documented elsewhere in Ubeji River, Warri, Nigeria [13]. However, pollution of water bodies by PAHs is usually caused by petroleum spills, and other man-made activities such as discharges and seepages, industrial and municipal waste water, urban and suburban surface runoffs, and atmospheric deposition [28]. This pollution could be point source or non-point source, but is very prominent in point source especially when it occurs around such environments. This could be responsible for the high values recorded in the fishes, surface water and sediments in this study. The results show a high level of PAHs in the fish samples that are consumed by the population. Several authors have reported that ingestion of seafood contaminated with pyrene and total PAHs increased incidence of tumours of the lung, skin and possibly bladder [29,30]. This suggests a significant risk when such contaminated seafood like fish represent a significant component of the diet. The accumulation of toxic substances to hazardous levels in aquatic biota has become a problem of increasing concern. This is because it has a direct and almost non escapable effect on man, either by drinking the water, using it as irrigation or by consuming the fishes from such sources.

5. CONCLUSION

The high level of petroleum hydrocarbon extracted from the surface water and fishes has provided evidence of severe crude oil contamination of the river. Such condition generally implies massive environmental degradation and ecological imbalance resulting in severe health defects like leukemia, brain damage, mental deficiency, retarded growth especially amongst children, anaemia, lungs and kidney cancer, liver necrosis, nephritis and even death, among indigenes residing in the area.

As a result of the increased industrial and anthropogenic activities around the area, there is a consequent release of millions of toxic and recalcitrant chemicals into the surface water environment. These activities however, release Polycyclic Aromatic Hydrocarbons and heavy metals (among many others) thus causing serious health problems to humans, marine/ aquatic animals and also pollute the environment due to their recalcitrant and bio-accumulative nature. Therefore, it's recommended that epidemiological studies should be carried out to ascertain the health impact of the spill on the indigenes of the area.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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