



Total Petroleum Hydrocarbon Concentration in Surface Water of Cross River Estuary, Niger Delta, Nigeria

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Authors' contributions

This work was carried out in collaboration between both authors. Author IED designed the study, wrote the protocol and the first draft of the manuscript. Author PJN managed the literature searches, experimental process and the statistical analysis of the study. Both authors read and approved the final manuscript.

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ABSTRACT

An increase of petroleum hydrocarbons levels in the aquatic environments has become worrisome because of their deleterious impact not only on marine organisms but also its effect on human health. In view of the devastating effect of total petroleum hydrocarbons, this study was carried out in order to ascertain the level of contamination of Cross River estuary. Total petroleum hydrocarbon levels were determined in surface water samples collected from various sampling points along Cross River estuary using Gas Chromatography – Flame Ionization Detector (GC-FID) instrument. Results obtained indicated that the surface water was contaminated with total petroleum hydrocarbon with mean concentration ranging from 13.16181 ± 1.485 to 24.85462 ± 8.058 mg/l compared to the control sample with concentration of 9.68200 ± 0.233 mg/l. Moderately high molecular weight hydrocarbons were dominant in all the sampling stations except in station 3. It was further observed that the level of total petroleum hydrocarbon in this study was high relative to Nigerian permissible limit of 10 mg/l in water. This poses a serious risk to the survival of aquatic organisms and also affects the quality of water used for various purposes.

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1. INTRODUCTION

Water is very essential for agriculture, industries and other human activities, and are normally obtained from two major natural sources which are surface water (water bodies) such as rivers, streams, fresh water lakes and ground water (geological water) such as borehole and well water [1,2]. Despite the numerous benefits of water, there is a very serious concern for the accessibility of this free and indispensable product due to the high rate of contamination of most water bodies. A wide range of contaminants are continuously introduced into the aquatic environment mainly due to increased industrialization, technological development, growing human population, oil exploration and exploitation, agricultural and domestic wastes run-off, and may contribute greatly, to the poor quality of river water [3,4,5]. Also, many aquatic animals such as fishes and shellfishes either die or become polluted with trace metals and bio-contaminants often associated with petroleum and municipal wastes [6,7,8]. Among the various sources of pollution, the petroleum industry is considered the greatest source of water pollution in Nigeria [9]. The increase in crude oil exportation and exploitation has resulted in remarkable increase in environmental degradation of terrestrial and aquatic ecosystems, and the degree of contamination may be significant where frequent spillages occur [10]. Organic chemicals such as hydrocarbons are major constituents of petroleum and can enter the aquatic environment through natural and anthropogenic sources. However, only small amounts of natural hydrocarbons are present in the environment that originates from geochemical and biosynthetic cycle [11]. Man made activities that contribute to the pollution of water bodies may be intentional or accidental and these include gas flaring, oil spill, discarding of used lubrication oils, tank cleaning, leakages from marine vessels and off shore oil production, direct ocean dumping, coastal, municipal and industrial wastes, runoff from crude oil polluted lands, seepage, refinery effluents etc [12,13], [14,15]. Among the hydrocarbons which are components of crude oil are total petroleum hydrocarbons. Total petroleum hydrocarbon (TPH) is a mixture of measurable amount of petroleum-based hydrocarbons found in crude oil in an environmental media [16]. Some of the chemicals found in TPH are hexane, benzene toluene, xylene, naphthalene as well as other

petroleum products and gasoline components. However, it is likely that samples of TPH will contain only some, or a mixture, of these chemicals [17,18]. There are many sources of TPH contaminants in our environment which include petroleum extraction, transportation, refining and consumption [19]. The amount and types of compounds in petroleum hydrocarbon release differ widely depending on the product spilled and how it weathered. It has been reported by several researchers that some of the total petroleum hydrocarbon (TPH) compounds can affect human central nervous system, cause asphyxiation of the organisms in the water bodies by oil coating thereby causing death, produce carcinogenic and mutagenic effect in humans, impairs the growth and development of marine organisms, causes fish, crustaceans and molluscs to acquire objectionable odour or flavour which reduces their market value and acceptability, ultimately it leads to death of both flora and fauna [20,17,21,22,23,24]. The Cross River system traverses from freshwater swamp ecology upstream to the mangrove swamp forest at the estuary (most of the estuary). The Cross River basin has an estimated area of 54,000 km², of which 39,000 km² (72.2%) lies in Nigeria while the remaining 14,000 km² (25.9%) lies in Cameroon [25]. The river discharge upstream (Itu) is 879 and 2533 m³ s⁻¹ for the dry and wet seasons, respectively [26]. The width of the Cross River estuary at the mouth is approximately 25 km, and the river is more than 440 km long with a tidal flushing of 1.83 billion cm³ per day [25,27].

This research work is aimed at determining the total petroleum hydrocarbons in surface water from Cross River estuaries in order to establish the baseline record necessary for any future monitoring programme.

2. SAMPLE LOCATION

The study was conducted in the Cross River Estuary in Itu Local Government Area of Akwa Ibom State. The Cross River Estuary lies between 7°55' E and 8°15' E and 4°32' N and 6°15' N. It has an area of about 1500 km² [28].

Four sampling stations were chosen along the shore of the estuary for the purpose of this study. Station 1 at was located Ikot Offiong upstream, (control station) far away from industrial activities except local fishing. Station 2 (downstream from

station 1) was approximately 100 m from Calabar Itu Head Bridge in Ayadehe village. This is a commercial station with a large market located at the River side, which serves as a landing site for fishermen and farmers. The area also serves as a major and general transportation channel in and out of the hinterlands. There is also a bus/heavy duty truck terminal within the vicinity. Domestic wastes from human households as well as a water and erosion runoffs are being emptied into this section of the River. Station 3 was about 20 km downstream from station 2 in Oku Iboku, There was also an intense surface water runoff from the adjoining farmlands and forest, while station 4 is situated at Ikot Nya which is about 1 km downstream of station 3.

2.1 Sample Collection

Surface water samples were collected from the different stations under study using glass bottles previously washed with dichloromethane. The water samples were preserved with 2 ml of 0.2 M H_2SO_4 to bring the pH to about 2. A piece of sterile aluminium foil was used immediately to cover each bottle so as to prevent any sort of contamination. No space was allowed between the foil and the sample bottle. The bottles were thereafter tightly covered with plastic screw cover. These were cooled to 4°C and kept in ice-packed cooler and transferred to laboratory for pre-treatment and analysis [29].

2.2 Extraction of Samples for TPH Determination

Filtered water sample were subjected to separatory funnel extraction procedure. One litre of water sample collected from different stations was extracted in a two litre (2 L) glass separatory funnel fitted with a glass stopper using 30 ml dichloromethane (DCM) as the extracting solvent. The separatory funnel was shaken vigorously for at least 3 minutes and pressure released at intervals. The flask was allowed to stand for a minimum of 5 minutes, thus allowing the organic layer to separate clearly from the aqueous phase. The lower layer (extract) was mixed with 5 g of anhydrous sodium sulphate to remove water and the sample collected into a beaker through a filter paper. The filtrate was concentrated to 3 ml by allowing it to evaporate at room temperature in a fume cupboard [30]. The extraction was repeated thrice for each sample.

2.3 Sample Clean – Up and Detection

Water extracts were subjected to silica gel clean – up to remove polar organic substances in the solvents. For the determination of total petroleum hydrocarbons, a gas chromatograph with flame ionization detector (GC FID) was employed as described by [31]. An Agilent 6890 N gas chromatograph with a flame ionization detector (FID) was used. The volume of the sample injected was 1 μ l while the carrier gas was nitrogen at flow rate of 1.5ml/min. Samples were injected in split less mode, The column used was DB-1, length 30 meters i d 0.25 mm and film thickness 0.5 μ m. Column temperature was programmed with initial temperature 60°C followed by an increase at the rate of 8°C per minute up to the final temperature of 275°C. The detector temperature was set at 300°C. The sum of all aliphatic and aromatic hydrocarbons measured by GC-FID provides a measure of total hydrocarbon concentration. Also, blanks were analysed as part of the quality control measures. A recovery test of the procedure was carried out and the % recovery was also determined.

2.4 Statistical Analysis

Data obtained were subjected to Simple student t-test and ANOVA (single factor) to test whether there is significant differences between the mean values of the control and the different sampling stations.

3. RESULTS AND DISCUSSION

Results of the average individual concentrations of hydrocarbons in water samples from the four locations studied are presented in Table 1. Average individual concentrations of TPHs ranged from below detection limit to 3.36848 \pm 0.010 mg/l in station 1 (control), with the highest concentration recorded for C- 26. For station 2, the level of individual hydrocarbons varied from below detection limit to 3.96863 \pm 0.012 mg/l, with the C – 26 having the highest concentration. For station 3, the range was not detectible – 12 .03553 \pm 0.03 mg/l with maximum concentration reported for C – 15. In Station 4, the total petroleum hydrocarbon levels recorded a range of non -detectable limit to 17. 40931 \pm 0.004 mg/l with C – 32 having the highest concentration. A comparison of the mean concentration of the entire individual total petroleum hydrocarbon in the four sampling stations using analysis of variance indicated that the differences in the

concentration of the individual hydrocarbons in the four sampling stations were not statistically significant. Evaluation of the mean concentration of TPH in the study areas with those of control showed that the mean concentrations of TPH in the study areas were higher than that of the control. However, when the concentrations of TPH were subjected to a paired test (student T Test), the results obtained indicated that the differences were not significant at 95% confident level. According to [32], gasoline and condensate range organics (GRO) generally include C4 through C9 hydrocarbons; diesel range organics (DRO) generally include C10 through C24 while Lube oil range >C28-C35. In station 2, it was observed that the surface water was mostly contaminated with hydrocarbons whose ranges fell into the category of mineral oil (lubricating oil) and diesel range organics. This is so because this sample station constantly receives high concentration of these hydrocarbons from runoffs from soil within the vicinity of heavy duty truck/ bus terminal. Generally, the lighter fraction aromatic hydrocarbons evaporate rapidly, particularly during periods of high wind and wave activity, [33] and this may also account for the absence of this range of hydrocarbons in most of the sampling stations studied. For station 3, the results obtained indicated that Gasoline Range Organic (GRO), which is between n-C6 to n-C12 and Diesel Range Organic (DRO) n-C12-n-C28 were dominant. The abundance of low molecular weight hydrocarbons (<n-C23) suggested that the contamination of this water sample may have been recent. These may be attributed to run offs from adjoining farmlands, domestic wastes and discharge of sewage. Also, petroleum product spills, outboard and inboard engine boats may have been a contributing factor. At station 4, diesel organic range (DRO) and lubricating oil range were dominant, with C – 32 being dominant. The observed levels of total petroleum hydrocarbon in surface water in all sampling stations in this study was higher than the maximum permissible limit of 10 mg/l for inland waters set by the [34,35,36] except in the control station. The variation in the pattern of the total hydrocarbon within the study area implied that most of the hydrocarbons in water samples were of anthropogenic origin. On comparing the mean concentration of total petroleum hydrocarbon obtained in this study with results obtained for other rivers in Nigeria and other countries, it was observed some of the results were comparable while others were higher than the concentration reported in this study. For instance, the mean

level of total petroleum in this study was within the range (23.6 + 4.3 mg/l) reported by [37], who evaluated the total hydrocarbon levels in some aquatic media in an oil polluted mangrove wetland in the Niger Delta, Nigeria. The results obtained for groundwater samples in some communities in Rivers State, Nigeria by [38], indicated that the mean concentration was higher (33076.00 µg/l) than that reported in this study. [39] carried out a research to ascertain the level of total petroleum hydrocarbon in soil and groundwater of crude oil impacted area of five communities in Niger Delta region of Nigeria. The result this study shows that water samples obtained from all the stations were contaminated with TPH with the mean TPH concentrations at Stations I, II, III, IV and V of water sample given as 8186.67, 12110.00, 1351.67, 4137.00 and 9020.67 µg/l, respectively. This was high compared to what was obtained in this study.

[29] investigated the total petroleum hydrocarbons in plants, surface river water and fish samples from and around River Ethiopie, Oghara community in Delta State, Nigeria. The results of the analysis revealed that the levels of TPHs in water ranged between (0.004 + 0.003 and 0.008 + 0.008) mg/L, which was far below the level obtained in this study. [15] determined the level of total hydrocarbon content (THC) in subsurface seawater sample and surface sediments obtained from the Kua/Kinabere Creek, in Ogoni land – an estuary of the Bonny River. The results showed that concentration of THC in water and sediment varied from 15.6±1.86 – 23.4±2.55 mg/l and 1,403±80.61 – 3,755±113.14 mg/kg respectively. [40] carried out an assessment of oil, grease, total petroleum hydrocarbons (TPH) and some heavy metals in surface and ground water within and around the Nigeria National Petroleum Corporation (NNPC) depot, in Apata, Ibadan metropolis in order to assess the pollution status of the water. Results obtained showed that values of TPH for both surface and groundwater in the studied area ranged from 20.34±1.79 to 27.40±5.32 and 2.67±0.80 to 13.03±2.21 mg/l respectively as against the control of 13.18±2.41 and 1.58±0.22 mg/l. A study was carried out by [41] in the Dungun River basin, southern South China Sea coastal area to determine the distribution of total petrogenic hydrocarbon (TPH) on the surface river water. The results showed that the concentration of TPH was in the range of 2.0-40.8 µg/l (mean 12.9 ± 8.8 µg/l). This was within the range reported in this study. The concentration of total petroleum hydrocarbons

Table 1. Total petroleum hydrocarbons values in mg/l for water samples in the different locations

Components	Station 1	Station 2	Station 3	Station 4
C8	nd	nd	nd	nd
C9	nd	nd	6.71960±0.001	nd
C10	nd	nd	nd	nd
C11	nd	nd	nd	nd
C12	nd	nd	2.08105±0.005	nd
C13	nd	nd	nd	nd
C14	nd	nd	nd	nd
C15	nd	nd	12.03553±0.03	nd
C16	nd	nd	nd	nd
C17	nd	nd	nd	nd
C18	nd	nd	nd	nd
C19	nd	nd	nd	5.30266±0.002
C20	nd	1.84437± 0.001	0.44134±0.002	2.14265±0.01
C22	nd	0.03813±0.003	nd	nd
C24	3.35548 ±0.004	2.76189±0.004	nd	nd
C26	3.36848± 0.010	3.96863± 0.012	nd	nd
C28	nd	0.61092±0.25	nd	nd
C30	2.95804±0.023	0.72928±0.007	nd	nd
C32	nd	3.20859±0.006	nd	17.40931±0.004
C34	nd	nd	nd	nd
C36	nd	nd	nd	nd
C38	nd	nd	nd	nd
C40	nd	nd	nd	nd
Total (mg/l)	9.68200±0.233	13.16181±1.485	21.27952±5.207	24.85462±8.058

nd = Not detected

and heavy metals in surface water and sediments of Ubeji river, Warri, Nigeria was determined by [20]. The mean value reported for total petroleum hydrocarbon in surface water was 73.5 ± 4.8 mg/l, which was higher than what was obtained in this study.

Supplementary chart describing the extraction procedures for total petroleum hydrocarbon in surface water is attached.

4. CONCLUSION

This study revealed that there was low to moderate total petroleum hydrocarbon levels in water samples obtained from all the sampling stations. When the levels obtained in this study was compared with different standards for drinking and other purposes, it was observed that the concentration of total petroleum hydrocarbon was higher than the stipulated limits set by these agencies. Furthermore, there is need for regular monitoring of surface water especially in rural areas with high industrial activities, and efforts should be made to reduce the presence of these hydrocarbons in the aquatic environment to avoid its bio-accumulation and consequently its toxic

effects in aquatic organisms and subsequently human beings.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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