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Temporal (Monthly) Distribution and Variation of Total Petroleum Hydrocarbons Content in the Water and Sediments from Orashi River, Engenni, Rivers State, Nigeria

Edori E. S. (Corresponding Author)
Department of Chemistry, Ignatius Ajuru University of Education Rumoulumeni, P.M.B. 5047, Port Harcourt, Rivers State, Nigeria
Email: enizeedori@yahoo.com

Edori O. S.
Department of Chemistry, Ignatius Ajuru University of Education Rumoulumeni, P.M.B. 5047, Port Harcourt, Rivers State, Nigeria

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Abstract

The total petroleum hydrocarbons content of the surface water and the sediments from the Orashi River were investigated between December 2019 and June 2020 at two months interval to find out the level of contamination of the river. The total petroleum hydrocarbons were determined by GC-FID using Agilent 5890N, after following due laboratory procedures of sample pretreatment and clean-up. The results recorded from the surface water were December, 5.844±1.231 mg/L; February, 8.767±2.501 mg/L, April, 16.886±3.157 mg/L and June, 7.271±1.110 mg/L. Variation in concentration showed that April > February > June > December. Results recorded in the sediments were December, 39.8427±13.5 mg/Kg; February, 29.5322±5.301 mg/Kg; April, 50.5040±16.813 mg/Kg and June; 16.6545±3.35 mg/Kg. The variation in concentration showed that April > December > February > June. The variations observed in of total petroleum hydrocarbons content in the river indicated that the contamination source was primarily anthropogenic. It is therefore recommended that effective measures and adequate steps be taken by government to mitigate the effect that may result from accumulation of the total petroleum hydrocarbons in the river and on the aquatic inhabitants and man who depends on the river for daily living.

Keywords: Contamination; Environment; Sediments; Water; Petroleum hydrocarbons.

1. Introduction

Over the years, the exploration, exploitation and the eventual production of oil in the Niger Delta Region of Nigeria have created a lot of challenges to individuals and communities of the region. One of such challenges is environmental pollution, which has led to the contamination of water bodies of the area. As a result of the increase in oil production, there is a notable change in the degradation in the environment, such as lakes, creeks, freshwater, estuaries and in the generality of the ecological settings of the oil-rich Niger Delta [1]. The discharge of petroleum products has led severe degradation, environmental pollution and deterioration of our heritage and has brought about other socio-economic problems over the years to the host communities of oil producing companies in the Niger Delta [2, 3].

Apart from agricultural waste and chemicals, petroleum hydrocarbons are the next group of chemicals that are significant in environmental contamination. Petroleum hydrocarbons have widespread use in transportation, heating at homes and industries which has led to the pollution of the environment due to its release [4]. Total petroleum hydrocarbons may find its way into the river by accidental discharge, operational failure and leakages from tanks and pipes. This definitely will lead to the contamination of the river or the environment in question. Studies have verified that petroleum hydrocarbons have neurotoxic effect to man and animals [5-7]. In order for the polluted environment to be restored and properly rehabilitated, there is the need to accurately measure the degree of contamination of the river by total petroleum hydrocarbons.

Major sources of environmental pollution as a result of oil production in the Niger Delta region of Nigeria include, gas flaring, oil spillage, pipeline explosion, disposal of petroleum product into the environment directly by uniform personnel, illegal oil bunkering and refining, sabotage of oil facilities, etc. [8, 9]. The process of oil exploration has led to serious impact which also includes the characteristics of the oil. Natural environmental conditions such as temperature, and weather have effect on the behaviour of oil in the aquatic environment, destroying the very essence of livelihood [10]. Aquatic organisms, most especially invertebrates play significant role in the purification process of natural water. Erosion control of the shoreline are easily affected by petroleum hydrocarbons due to oil spill. This results in grave consequences since they produce food for a variety of aquatic wild life. The negative effect of oil spill on the environment is such that it effects the invertebrate life stage, habitat, feeding habit etc. and the ability to avoid contamination during the spill [11].

Water quality have significant impact in the distribution and diversity of water organisms. Exposure of aquatic organisms to polluted water have the possibility of altering the structure of the community due to the possible elimination of some of the species that do not have the ability to withstand stress, and also the increase of species that can withstand or tolerate stress [12, 13]. The suitability of water for domestic use and aquatic life is hampered due to the release of total petroleum hydrocarbon into it. Some fractions of total petroleum hydrocarbons float on the water surface while other fractions sink to the bottom sediment and become a problem to bottom sediment organisms [14, 15] and also results in the death of many aquatic plants and animals [16]. Hence the effective use of water which is very essential for agricultural, industrial and domestic use is highly affected.

The purpose of this research work is to determine the concentration of total petroleum hydrocarbons in the surface water and sediments of Orashi Rivers for a period of eight months, taking interval of two months, which may be as a consequence of natural factors, legal and illegal exploitative and explorative activities of crude oil. It will also determine the level of pollution or contamination of the water bodies. The results obtained may be useful in providing information on the contamination or pollution status of the river which may be of great importance to the rural dwellers who are in constant contact with the water. It will further be used as a source of data bank on the content of total petroleum hydrocarbon in the river.

2. Materials and Methods

2.1. Study Area and Location Description

The Orashi River is a tributary of River Niger and runs from Imo State through Ogba Egbema Ndoni Local Government Area to Engenni in Ahoada West Local Government Area of Rivers State. The sample location points in the Engenni axis of the Orashi River were within longitude.

The inhabitants of Engenni are mainly fishermen and farmers. There are numbers of activities that take place within the Engenni Axis of the river which includes, oil exploration and production, illegal oil bunkering, transportation, etc. Oil production facilities that belongs to multi-national oil companies includes flow station, oil fields, gas plants, which has constantly led to the contamination of the river. The environmental impact and pollution due to oil exploitation and production over the years in this region have been a source of worry for the inhabitants. Other activities that are predominate in the river apart from oil production includes fishing, sand miming recreational activities (such as swimming and canoeing).

The study area covers two rivers in Engenni, Ahoada West Local Government Areas of Rivers State, in the Niger Delta area of South South Nigeria. The Orashi River is a tributary of River Niger and runs from Imo State through Ogba Egbema Ndoni Local Government Area to Engenni in Ahoada West Local Government Area of Rivers State. The sample location points in the Engenni axis of the Orashi River were within the geographic positions $4^{o}107.3'$ N, $6^{o}30'6.6''$ E and $4^{o}59'10.1''$ N, $6^{o}27'2.5''$ E. The inhabitants of Engenni are mainly fishermen and farmers. There are numbers of activities that take place within the Engenni Axis of the river which includes, oil exploration and production, illegal oil bunkering, transportation, etc. Oil production facilities that belongs to multi-national oil companies includes flow station, oil fields, gas plants, which has constantly led to the contamination of the river. The environmental impact and pollution due to oil exploitation and production over the years in this region have been a source of worry for the inhabitants. Other activities that are predominate in the river apart from oil production includes fishing, sand miming recreational activities (such as swimming and canoeing).

2.2. Collection of Water Sample

Glass bottles were used in collecting water samples from four different locations in Orashi River. The use of dichloromethane (99.8 %) was applied in rinsing the glass bottles were thoroughly washed so as to ensure no contamination with the collected water samples. At each location, water samples were collected at three different points at a depth of 35cm below the water surface and thoroughly mixed to form one composite sample. The collected water sample was preserved by the addition of 2ml of 0.2M H_2SO_4 (95-99 %), to make up the pH to 2. The glass bottles were then covered with sterilized pieces of aluminum foil to prevent the sample from contamination, then the glass bottles were firmly covered with plastic screw. The glass bottles were then put in an ice packed cooler to maintain the temperature at $4^{o}C$ and then transported to the Science Laboratory of Ignatius Ajuru University of Education for sample pre-treatment and further analysis [17].

2.3. Collection of Sediment Sample

Sediment samples from the Orashi River were collected with the aid of a hand-held van veen grab sampler at four different locations from the top few centimeters (about 5-6cm) deep into the sediment. A bulk representative sample was formed by mixing sediments from three different points within a location together. The samples were collected then transferred to previously washed glass bottles then transported to the laboratory. The collected sediment samples were treated as in the case of water samples before extraction analysis for total petroleum hydrocarbons.

2.4. Water Samples Extraction for Total Petroleum Hydrocarbon Determination

The water samples collected were filtered and then subjected to the process of extraction with the use of a separatory funnel. The water samples of different volumes were collected from different locations and were then extracted into a 2 litre glass separatory funnel and then filtered with the help of a glass stopper. 30ml dichloromethane was used as the solvent of extraction. The separatory funnel was vigorously shaken for at least 5 minutes. This enhances the organic layer to thoroughly separate completely from the aqueous layer. About 5g of anhydrous sodium sulphate was then mixed with the extract (lower layer) so that water will be removed. The extracted sample was collected into a beaker with the use of a filter paper. The filtrate was then allowed to evaporate under room temperature conditions in an already prepared fume cupboard. The filtrate was concentrated to 3ml. the procedure of extraction was done three times for each water sample collected [18].

2.5. Sediment Sample Extraction for the Determination Total Petroleum Hydrocarbon

10g of sediment sample was weighed out from the bulk sample with an analytical weighing balance and then transferred into a previously washed amber bottle. About 5g anhydrous sodium sulphate (Na_2SO_4) of purity (96.0 %) was added into the amber glass bottle containing the sediment sample, and then the sediment sample was shaken with vigour for thorough mixing of sample particles. Anhydrous sodium sulphate was added in order is to reduce water content of the sediment sample. After due mixing of the sediment sample, the addition of 30ml dichloromethane was then added to the sediment sample as the solvent of extraction. The amber bottle was closed tightly and was taken to a mechanical shaker immediately the dichloromethane was added, the amber bottle was then very tightly closed and then transferred to a mechanical shaker and thereafter was agitated at room temperature and the agitated sample allowed to settle down for a period of 1hr. The sediment sample was then filtered into a previously prepared clean beaker with a board on it with the aid of a 110mm filter paper. The sediment sample that was filtered was then allowed to evaporate at room temperature and concentrate to 1ml [18].

2.6. Sample Clean Up for Total Petroleum Hydrocarbons Analysis

The preparation of the column was performed by the introduction of glass-wool into a previously washed chromatographic column. Silica gel was put into an already washed beaker and then the addition of slurry into chromatographic column was made for analysis. Anhydrous sodium sulphate was then added into the chromatographic column and thereafter pentane was added also to the column. The sample that was previously concentrated was mixed with cyclohexane in an already cleaned beaker and then the sample was introduced into the chromatographic column already prepared. n-pentane (99.9 %) was used in the elution of the sample which was then collected in an already prepared beaker kept below the chromatographic column. More elution of sample was done out through the addition of more n-pentane, thereafter the column was then properly rinsed with dichloromethane. The sample was then allowed to stand in a firm cupboard after the elution at room temperature conditions in order for evaporation to take place [18].

2.7. Separation and Detection of Sample

The total petroleum hydrocarbons in sediment and water samples of the area investigated was analyzed and detected using Agilent 5890N gas chromatography-flame ionization detector (GC-FID) [19]. A 3ml of the concentrated water and sediment samples was injected into the gas chromatography vial for proper cleaning of the micro-syringe, after then a blank dichloromethane was then injected into the micro-syringe of the gas chromatography. Before any analysis of the sample will take place, the cleaning of the syringe was done three times. Thereafter the micro-syringe was rinsed again with the sample before the sample was then injected into the gas chromatography vial for the actual separation of the components of the sample. After separation of the different components in the sample, the quantity of total petroleum hydrocarbon content resolved at a particular chromatogram was then measured out in mg/L for water sample and mg/Kg for the sediment sample.

All laboratory reagents and chemicals used were of analytical grade (AnalaR grade), manufactured by BDH Chemicals Limited, Parkstone, England.

3. Results and Discussion

Considering Table 1, the level of surface water pollution or contamination for the various months are, December; 5.844±1.231mg/L, February; 8.767±2.501mg/L, April; 16.886±3.157mg/L and June; 7.271±1.110mg/L. The concentration degree due to contamination in the different months revealed that, April > February > June > December. The mean concentration degree of contamination in the months is 9.692±2.00mg/L. The variation of the results obtained from the different months was a proof that the contamination of the river by total petroleum hydrocarbons was as a result of anthropogenic activities. This finding is corroborated by other authors [16, 20]. As the months progressed from December 2019 to June 2020, it was observed that total petroleum hydrocarbons did not accumulate in the Orashi River. The Orashi River is a fresh water body and it is fast flowing especially in the rainy periods when the volume of the river has greatly increased and thus does not aid accumulation of contaminants.

The presence of total petroleum hydrocarbons in the river was mainly due to the illegal oil bunkering activities and other associated businesses that take place within the region. The total petroleum hydrocarbons discharged into the river were not subjected to treatment. This was the same observation made by Aziz [21], in which sewage discharged into the river were not subjected to treatment. The petroleum hydrocarbons reached the river through drainages occasioned by rainfall or by direct discharge from the illegal refinery operators and their allied business partners. This assertion is also close to that observed by Talal [22] that most wastes reach the river through drifting during rainfall and the melting of snow.

During December, the high temperature associated with the harmattan breeze gave the low content of the total petroleum hydrocarbons in the river. This may also be as a result high volatility rate of the lower fractions and hence its low availability. The observation here is similar to that made by Edori, *et al.* [23] in the Edagberi River were total petroleum hydrocarbons concentration was as low as 2.47858±0.119mg/l in the month of December. The evaporation of the lighter fractions of total petroleum hydrocarbons is the fastest known physical process that influences the reduction of such in the aquatic environment [24]. The rate at which this occur can also be influenced by such factors as wind velocity, temperature, surface water characteristics of the river also known as turbulence [25], which are prevalent environmental conditions during the month of December in the area. The period also encourages effective metabolic activities of micro-organisms which helps in the degradation of hydrocarbons in any environment [26], since the required temperature for effective life metabolism for optimum use is achievable during this period. The increase in concentration of total petroleum hydrocarbon fractions in the month of February was due to the early rains of the year associated with the month which drifts petroleum hydrocarbons fraction from the adjoining farmlands to the river, and thereby increasing the hydrocarbon content [20].

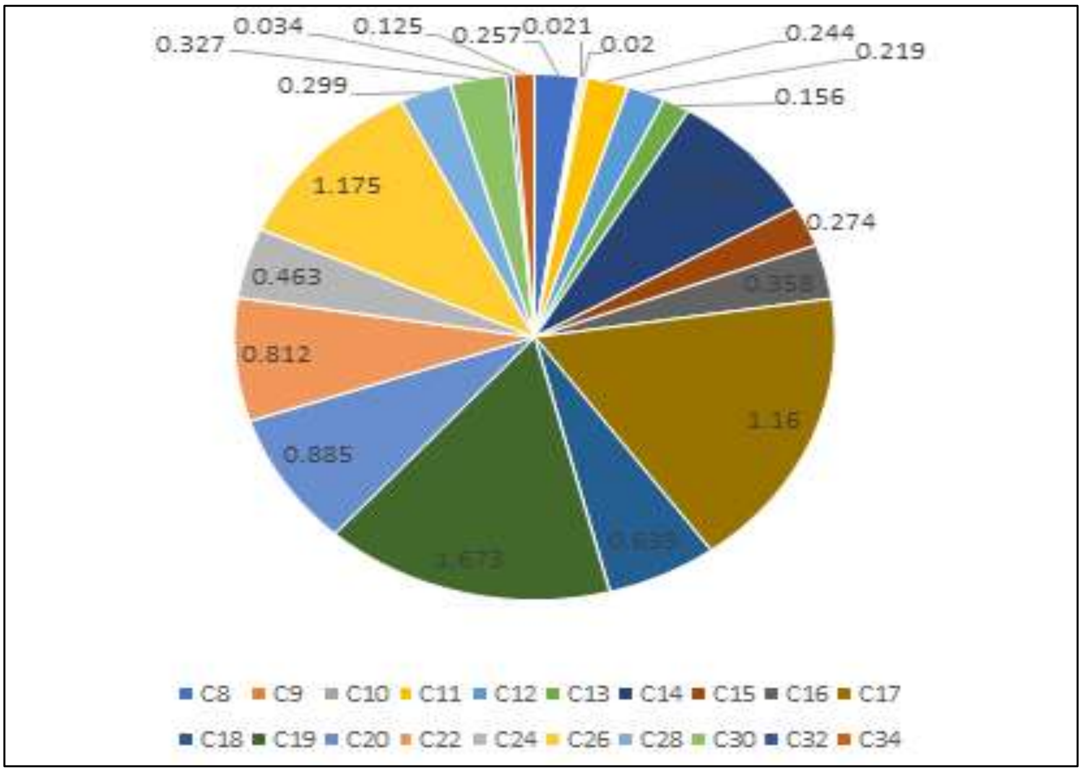
The further increase observed in the total petroleum hydrocarbons content in April was due to the increase in the amount of rainfall within the area. The total petroleum hydrocarbons in the surface soils of the adjoining farmlands and artisanal refineries were further washed into the river into the river and the increase in the water waves and the resuspension of the fractions from the sediments that resurfaced in the surfaced in the surface water must have led to the noticeable increase [20, 27]. The low result obtained in June was as a result of the increased volume of water due to that occasioned by the annual flooding of the river. This result agreed with Ololade, *et al.* [28], who noted that total petroleum hydrocarbons decreases with increase in water depth. During this period, as the suspended and dissolved particles enter into the river, which has large volume of water it is immediately dissolved and dispersed into the larger volume and its effect is suppressed and not felt as expected [29]. The total petroleum hydrocarbons contaminants are quickly dispersed and emulsified and remain as small droplets within the river system as it diffuses through the water body [30].

Table-1. Mean (X ± SD) Concentrations (mg/L) of Total Petroleum Hydrocarbon in Surface Water of Orashi River within the Sampled Months

Carbon Length	Months			
	December	February	April	June
C8	0.295±0.10	-	0.732±0-092	-
C9	0.085±0.003	-	-	-
C10	0.065±0.002	-	-	-
C11	0.446±0.014	-	0.528±0.074	-
C12	0.296±0.006	-	0.579±0.076	-
C13	0.445±0.013	-	0.178±0.008	-
C14	0.084±0.002	3.190±1.010	0.248±0.01	-
C15	-	-	0.616±0.06	0.479±0.04
C16	-	0.123±0.011	1.291±0.19	0.016±0.002
C17	2.348±0.800	0.514±0.073	1.776±0.071	-
C18	-	1.636±0.27	0.712±0.07	0.208±0.013
C19	1.643±0.28	1.326±0.21	3.117±1.20	0.604±0.054
C20	-	1.626±0.26	1.295±0.201	0.620±0.06
C21	-	-	-	-
C22	-	-	1.423±0.210	1.823±0.32
C23	-	-	-	-
C24	-	-	0.800±0.068	1.052±0.23
C25	-	-	-	-
C26	0.137±0.011	0.352±0.010	2.938±0.58	1.272±0.25
C27	-	-	-	-
C28	-	-	0.598±0.082	0.598±0.072
C29	-	-	-	-
C30	-	-	0.790±0.089	0.519±0.061
C31	-	-	-	-
C32	-	-	0.103±0.0009	0.034±0.003
C33	-	-	-	-
C34	-	-	0.453±0.057	0.046±0.005
C35	-	-	-	-
C36	-	-	-	-
C37	-	-	-	-
C38	-	-	-	-
C39	-	-	-	-
C40	-	-	-	-
Total	5.844±1.231	8.767±2.501	16.886±3.157	7.271±1.110

Figure 1 showed the results for the average concentration for the different total petroleum hydrocarbons components in the surface water of Orashi River. The mean value ranged from not detected to 1.673mg/L in water column of the river. The results indicated the absence of C21, C23, C25, C27, C29 and C31 and also higher fractions of C35-C40 during the months investigated. The highest concentration of 1.673mg/L was obtained in the C19 fraction.

Figure-1. Mean concentrations for the various total petroleum hydrocarbon fractions in the surface water of Orashi River within the months under study



Considering Table 2, the contamination degree or level of total petroleum hydrocarbons in the sampled months are; December; 39.8427±13.5mg/Kg, February; 29.5322±5.301mg/Kg, April; 50.5040±16.813mg/Kg and June; 16.6545±3.35mg/Kg. The results of the concentration levels of total petroleum hydrocarbons in the months under review showed that April > December > February > June. The mean concentration level for the months is 34.1334±9.741mg/Kg. Table 2 showed that there existed variation on the occurrence of total petroleum hydrocarbons in the sediments of the Orashi River during the months under consideration. The results revealed that there was no progressive accumulation of total petroleum hydrocarbons in the sediments of the river and the total petroleum hydrocarbons content varied from month to month.

In December, due to the low level (shallow depth) of the river during that period and the slow nature of the flow of the river, petroleum hydrocarbon content tends to be high to the level obtained in the result. There is also the issue of less disturbance as most activities like fishing, and manual sand dredging are always minimal during that time of the year. The shallowness of the river at this time gave rise to the quantity of total petroleum hydrocarbons content during the month of December. Despite the fact that the river is flowing in one direction, the shallowness of the river during this period readily allowed and favoured sedimentation. This agreed with other authors that shallow water or rivers are always associated with and full of suspended particles of solid materials and therefore encourages sedimentation [28, 31]. The observed reduction in the concentration of total petroleum hydrocarbons in February in the river can be attributed to the fact that there was reduction in the activities of illegal refining and bunkering. At this period, there is higher concentration of farming activities and fishing in lakes and ponds in the forest, therefore less attention was given to the illegal and self-sustaining oil business. The constant one directional flow of the river does not allow sedimentation of the total petroleum hydrocarbons as observed by Edori and Marcus [32]. The rate of volatility of the total petroleum hydrocarbons due to the temperature of the area at this period is high. Increase in temperature leads to faster photochemical reactions and hence decomposition of petroleum hydrocarbons which is responsible for the reduction of the contamination level in this period [33]. The rapid evaporation of the lower fractions of total petroleum hydrocarbons from the surface of the sediments makes it difficult for the hydrocarbon fractions to persist in the water and sediment columns [15, 16].

The sharp rise in the petroleum hydrocarbon content in April was due to the increase in rainfall, whereby total petroleum hydrocarbons were swept into the river from the adjoining farmlands. This observation agreed with Kim, *et al.* [34] and Shirodkar, *et al.* [35], where temporal distribution of petroleum hydrocarbons in sediments were in the order April > January > May > March > February. The concentration of total petroleum hydrocarbons was higher in autumn than summer due to higher rainfall. Despite the increase in rainfall, the contaminants in the sediments were not being re-suspended by being released again into the water column, for the contaminants were still trapped within the sediment column and cannot be remobilized into the water phase. This was contrary to that observed by Lick

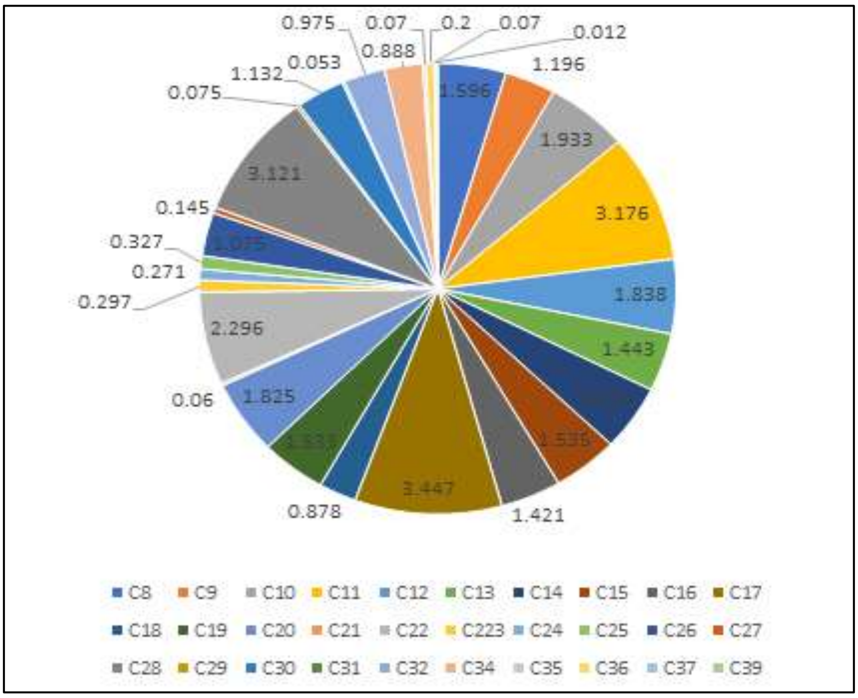
[36], for at this time the flow rate, turbulence and re-suspension of contaminants in the river was still very low. A further variation in the level of contamination of total petroleum hydrocarbons was observed in June with a sharp decline in the concentration. The increase in the volume of water due to flooding causes increase in dispersion, spreading, evaporation occasioned by turbulent flows quickly emulsifies the contamination and push it away from the very source of contamination [37]. The flow of the river pushed the contaminants into the body of trees and the adjoining farmlands due to the overflow of the river during this period [32]. The openness of the river allowed for unlimited connections and therefore high capacity for self-purification. This finding disagrees with Deppe [38], in the Khowr-re Musa Bay, which is semi closed and therefore its connection to the Persian Gulf is limited, hence high contamination of suspended solids and low tendency to self-purification. The sediments contained low total petroleum hydrocarbons content because the flow quickly drives the pollutants away into the sea due to the faster flow rate and turbulence during the flooding period. The observation of Udoh and Akpan [39], agreed to this fact in the Bonny estuary where turbulence, strong wind current, speed of the water quickly pushed away any crude oil that entered the creek to the Bonny estuary.

Table-2. Mean (X ± SD) Concentrations (mg/Kg) of Total Petroleum Hydrocarbon in Sediment of Orashi River within the Sampled Months

Carbon Length	Months			
	December	**February**	**April**	**June**
C8	5.2485±2.030	-	1.1357±0.012	-
C9	2.8636±0.525	1.9194±0.116	-	-
C10	4.8402±1.906	2.8905±0.124	-	-
C11	9.0004±3.601	3.7055±1.320	-	-
C12	6.4897±2.160	0.3750±0.061	0.4883±0.200	-
C13	1.2660±0.031	2.6250±0.250	0.7743±0.310	1.1064±0.101
C14	3.1244±1.112	1.8562±0.130	0.9712±0.430	0.5455±0.062
C15	0.0001±0.000	-	4.6982±1.760	1.4411±0.200
C16	-	01604±0.007	5.5220±2.050	-
C17	7.7660±2.010	0.7061±0.030	1.4772±0.500	3.8377±1.030
C18	-	1.2562±0.112	0.4321±0.032	1.8224±0.320
C19	-	1.3969±0.140	1.0122±0.056	3.7213±1.000
C20	-	3.0030±1.320	3.4089±1.192	0.8861±0.063
C21	-	0.0805±0.006	-	0.1577±0.008
C22	-	1.7462±0.090	6.8998±2.410	0.5364±0.040
C23	-	0.9606±0.009	-	0.2277±0.013
C24	0.0020±0.001	0.0165±0.001	0.5677±0.050	0.4996±0.260
C25	-	1.0820±0.019	-	0.2246±0.008
C26	0.0178±0.010	2.1655±0.911	1.2614±0.630	0.8552±0.071
C27	0.0069±0.002	0.5720±0.052	-	-
C28	0.0111±0.003	0.3130±0.040	11.9709±4.010	0.1899±0.071
C29	-	0.3011±0.036	-	-
C30	0.0155±0.009	1.0480±0.500	2.8615±1.012	0.6034±0.103
C31	0.0188±0.010	0.1933±0.002	-	-
C32	0.0204±0.010	1.0690±0.020	2.8101±1.001	-
C33	-	-	-	-
C34	0.0248±0.010	0.0903±0.006	3.4371±1.260	-
C35	0.0263±0.110	-	-	-
C36	0.0247±0.100	-	0.7754±0.032	-
C37	0.0278±0.014	-	-	-
C38	-	-	-	-
C39	0.0477±0.031	-	-	-
C40	-	-	-	-
Total	39.8427±13.500	29.5322±5.301	50.5040±16.813	16.6545±3.350

Figure 2 illustrated the mean concentration value of total petroleum hydrocarbons fractions in the sediments of Orashi River. The values ranged from not detected in the C33, C38 and C40 fractions to 3.447mg/Kg in the C17 fraction in the months investigations were carried out.

Figure-2. Mean concentrations for the various total petroleum hydrocarbon fractions in the sediments of Orashi River within the months under study



4. Conclusion

The level of total petroleum hydrocarbons available in the surface water and sediments of the Orashi River within the months investigated showed that the river was contaminated. The contamination brought about environmental degradation, deterioration and imbalance in the ecological system. Such ecological effect may pose reasonable effect on the aquatic plants and animals within the river system and also humans that rely on them for daily food. The petroleum hydrocarbons in the river were mainly due to the activities of man which include illegal oil bunkering, artisanal refining and transportation businesses and the high level of contamination may lead to a remarkable decrease of the diverse species that inhabit the river. Adequate steps should therefore be taken by relevant agencies of government to restore the river by remediation processes to put the river back to its former useful state and restore the ecological system of the Orashi River environment.

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