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Monthly Variation of Total Petroleum Hydrocarbons Content in the Surface Water and Sediments of Kolo Creek, Engenni Axis, Niger Delta, Nigeria

Edori, E.S.[1], Bekee, D.[2], Okpara, K.E.[3]

[1]Department of Chemistry, Ignatius Ajuru University of Education Rumoulumeni, P.M.B. 5047, Port Harcourt, Rivers State, Nigeria
[2]Department of Chemistry, Rivers State University, P.M.B. 5080, Port Harcourt, Rivers State, Nigeria
[3]Department of Environmental Management, Prince of Songkla University, Hat Yai Songkhla 90112, Thailand
Corresponding Author: enizeedori@yahoo.com

Abstract This research work examined the contamination/pollution of the surface water and sediments of Kolo Creek by total petroleum hydrocarbons in the months of December, February, April and June. The concentration of the total petroleum hydrocarbons in the different months was determined by Gas Chromatography-Flame Ionization Detector with Agilent 7820A model, after the samples have been extracted through the use of Dichloromethane followed by column chromatography clean-up. The concentration of the total petroleum hydrocarbons in the surface water of the Kolo Creek varied in the months as, 10.590±2.113, 10.485±1.985, 15.547±4.590 and 24.762±6.492mg/L in December, February, April and June respectively with an average monthly concentration of 15.346±3.770mg/L while in the sediments the variations observed was 69.3574±24.2836, 25.4603±6.3405, 36.6575±9.7485 and 33.4888±7.6412mg/Kg for the months of December, February, April and June respectively with an average monthly concentration of 41.241±12.0035mg/Kg. the study observed that the pattern of variation of total petroleum hydrocarbons in the creek was majorly anthropogenic. The study also showed that the illegal oil business in the area has led to the contamination of the creek and the presence of total petroleum hydrocarbons in the creek has greatly affected the life pattern of the inhabitants of the area. The results showed that the creek was slightly polluted and therefore require proper regulation and control of activities leading to the presence of total petroleum hydrocarbons in the creek.

Keywords Petroleum hydrocarbon pollution, surface water and sediments contamination, remediation process, Niger Delta, illegal oil bunkering, petroleum hydrocarbon accumulation

Introduction

The process of extraction and production of crude oil (petroleum) and its refining and eventual usage as the major energy source all over the world has culminated into widespread environmental pollution. The invaluableness of oil for energy and production of several other chemicals from the extraction to the consumption with little or no attention to its resultant effect has caused tremendous environmental degradation, deterioration and pollution that has proven difficult and has brought about severe irreparable conditions to the environment [1]. Several million barrels of untreated petroleum products enter into the aquatic environment on yearly basis [2]. The task of contributing such environmental pollution in the water bodies is difficult since the mode of entry and sources of entry are complex in nature. These petroleum hydrocarbons dissolve and mix with water and also sink to the bottom where they accumulate [3-4] and causes resultant changes in the nature of water and its attendant characteristics [5].

Petroleum hydrocarbons are well known organic contaminants and often being dispersed into the aquatic environment through automobile wastes, industrial effluents, urban runoffs, domestic wastes and stormwater [6]. Petroleum hydrocarbons mainly contains normal alkanes, degraded crude oils and combusted fossils [7-8]. These petroleum hydrocarbons have low solubility in water and are easily adsorbed into organic and particulate matters and easily sink to the bottom where they become contaminants and reserviour for hydrophobic substances [9-10]. The contamination of the aquatic environment by petroleum hydrocarbons have become a thing of great concern to several authors due to its carcinogenic, mutagenic and toxicological effects [11-12] on aquatic organisms and humans. The presence of total petroleum hydrocarbons and other associated chemicals that contaminates the aquatic environment from sources has posed great danger to the coastal marine environment [13].

The resultant effect of petroleum hydrocarbons pollution is grievous and of utmost environmental challenge to the world this moment [14]. It introduces both known and unknown challenges to the ecological system due to the presence of contaminants that are toxic to the environments. The ecosystem experiences complex changes due to oil pollution by altering metabolic processes, violation of the production processes, organic matter destruction, which eventually leads to the reduction in the composition of species diversities and structure of the ecosystem and loss to stability of life within the environment [15]. The inhabitants of an oil polluted environment are usually associated with depression and anxiety, mental health issues, facial expression, smoking lifestyle and general health challenges [16]. The intake and absorption of petroleum hydrocarbons by lower organisms within the ecosystem can pass through the food chain/web system to the higher organisms which may result in loss and reduction of biodiversity as a result of oil pollution [17] and eventual death and elimination of non-resistant organisms.

Negligence on the part of the various levels of government in Nigeria and other relevant agencies in educating and sensitizing the general public on the negative effect of petroleum hydrocarbon pollution has led to the rate at which the environment has been degraded and polluted [18]. The rate at which toxicants/pollutants are being discharged by accidents or by deliberate action into the environment is alarming. Anthropogenic and accidental discharges have brought total petroleum hydrocarbons into the creeks and rivers of the Niger delta region of Nigeria and have left evidences of damages (chronic and acute) to tissues and organs of organisms. Such pollutants include heavy metals and PAHs [19-20]. The exposure to these pollutants has led to serious health challenges because their poisonous and toxic effects to both plants and animals has resulted in changing the population status and density of organisms and possible elimination of certain species within the polluted area [21-22].

The research work aimed at determining the degree of contamination/pollution of Kolo Creek and also examined the degree of variation that existed in the concentration of total petroleum hydrocarbons within the months in which investigation was carried out. The work was limited to four months at bi-monthly intervals with a sampling rate of three times in a month. An average concentration in any particular month was taken as the level of contamination for that month.

Materials and Methods

Description of Study Area and Location

The Kolo Creek is a tributary of the Orashi River. The creek begins from Okarki, in Engenni, in Ahoada West Local Government Area Rivers State, Nigeria. The creek was the major route used the Engennis in Rivers State and the Ogbias in Bayelsa State to access trade and travels prior to the construction of roads by the government. Notable locations in which samples collected in the Engenni axis of the creek were Okarki Baptist Waterfront, Pilopia Waterfront (Okparaki Bridge) and Otegwe (Agbaa) waterfront. These locations were chosen because they give easy access to the creek and other activities such as oil business and bathing take place within these points of the creek. The samples were collected within 4^{o}58’53.1”N, 6^{o}25’52.6”E and 4^{o}57’26.3”N, 6^{o}26’10.3”E geographical positions. The exploration, production and the eventual exploitation of oil within the local governments that make use of the creek has brought about the degradation and deterioration of the creek and its environs. The transportation of petroleum products, oil bunkering business, artisanal refineries has deeply affected the livelihood of the inhabitants due to the effect of pollution on the creek. Normal activities such as fishing, farming, recreations (swimming, canoeing), boat transport and even local sand dredging for building and other construction works have been greatly affected.

Materials and Reagents for the Study

The materials for the research work were; weighing balance, (±0.0001 accuracy), micro-syringes, dessicators, vials, GC-FID, water baths, glass separatory funnels, glass stoppers aluminium foil,hand held van veen grab, fume cupboard,filter paper, measuring cylinder, volumetric flasks, ice pack.

The chemicals/reagents used for the work were of analar grade. They include; anhydrous sodium sulphate (Na_2SO_4), dichloromethane, pentane silica gel, 0.2M H_2SO_4, n-hexane, 1-chlorooctadecane.

Sampling Period and Sample Collection

Samples were collected from the surface water and sediments of Kolo Creek in the month of December 2019 to June 2020 at bi-monthly intervals. In any particular location, water samples were collected with the aid of glass bottles at 20cm deep from three different points and then combined together to form one sample. The collected water sample was then preserved by the addition of 0.2M H_2SO_4 to keep the pH at 2. Sterilized pieces of aluminium foils were used to cover the bottles and then plastic screws were used to tightly hold the aluminium foils unto the bottle in order to avoid contamination.

Sediment samples from the three stations in the Kolo creek were collected at a depth of 1 metre from the top about 5-6 centimetres deep into the Combination of three points samples within a location gave a bulk representative sample. A hand-held van veen grab was used in the collection of the sediment samples and then transferred to an already prepared glass bottle. Both surface water samples and sediment samples were keptin an iced park to keep the temperature at 4^oC before being transported to the laboratory for pre-treatment and analysis [23-24].

Total Petroleum Hydrocarbons Extraction from Water and Sediment Samples

A surrogate standard of 1ml 8g/ml 1–chlorooctadecane was used in spiking 400ml of surface water sample and then extracted 3 times using a separatory funnel with 16ml of n-hexane. The extracts were dried with anhydrous sodium sulphate after being pooled together and then concentrated to 2ml with rotary evaporator [25-26].

The sediment samples were allowed to air dry for 5 days [9-10] then anhydrous sodium sulphate (Na_2SO_4) was added to 10g of the sample eliminate moisture. The sediment samples were extracted using a Soxhlet apparatus (extractor) with the use of dichloromethane for a period of 24 hours after it has been spiked with 1 – chloroctadecane. A glass funnel containing Na_2SO_4 was used to collect the extracts and a rotary evaporator was then used in concentrating the extract to 2ml [25, 27]. Both surface water and sediment samples were now ready for analysis after due clean up and separation.

Sample Clean Up and Separation

The extracted water and sediment samples were transferred into a chromatographic column with dimension 10mm ID x 30cm. The column was packed with 10g of silica gel and slurry with 2cm thickness of anhydrous sulphate on the top. In other to obtain the total petroleum hydrocarbon fractions, 20ml n-hexane was used in eluting the column [28]. With the use of a rotary evaporator the eluted solvent was effectively concentrated to 2ml and then exchanged to n-hexane. A control sample was also prepared as in the case of the actual sample for quality control and assurance [25, 29].

Sample Detection Using Gas Chromatography-Flame Ionization Detector

After due clean up and separation, the extracted samples were analyzed with Agilent 7820A gas chromatography-flame ionization detector. The carrier gas used in the chromatogram is helium and it operates at a flow rate of 1.75ml/min. with an average velocity of 29.47cm/sec [9-10]. About 1µl of the sample extract was injected into the chromatogram's splitless mode at a temperature of 300 ^oC with the aid of a micro-syringe at a temperature of 40 ^oC. The temperature of the detector was kept at 300 ^oC [13]. The detected amount of total petroleum hydrocarbons by the chromatogram is measured in mg/L and mg/Kg for water and sediment samples respectively.

Results and Discussion

In Table 1, the degree of surface water contamination/pollution in the different months were; December; 10.590±2.1129mg/L, February; 10.485±1.985mg/L, April; 15.547±4.59mg/L and June; 24.762±6.492mg/L. The results for the various months revealed that in the months; June > April > December > February. The mean monthly concentration of total petroleum hydrocarbons in Kolo Creek in the surface water was 15.346±3.77mg/L. Individual petroleum hydrocarbons fractions also showed variations within the months investigated, for example C_{17}fraction varied between 0.034±0.002, 0.146±0.008, 0.022±0,020 and 2.754±0.810mg/L for the months of December, February, April and June respectively. Also, the C_{26} fraction varied from 1.042±0.090, 0.007±0.001, 0.422±0.210 and 2.237±0.590mg/L for the months of December, February, April and June respectively. The variations observed in the concentration of total petroleum hydrocarbons within the months and even in the individual hydrocarbons fractions was an indication that the presence of the total petroleum hydrocarbons in the creek was due to anthropogenic input.

The low concentration of total hydrocarbons value of 10.590±2.1129mg/L observed in December in the water column of the Kolo Creek probably arose due the rapid evaporation occasioned by winds and waves of the harmattan period in the area. This assertion is corroborated by Hanson *et al*., [30], that lighter petroleum hydrocarbon fractions undergo rapid evaporation during periods that high winds and waves are prevalent. The low amount of rainfall in December and the absence of runoffs from farmlands and non the precipitation of flared components of hydrocarbons fractions probably led to the low concentration of total petroleum hydrocarbons during December. This finding agreed with that obtained by Edori and Kpee [31] in the Taylor Creek. The semi-still nature of the creek also enhanced the rate at which the hydrocarbon fractions were evaporated, since the spilled crude was not allowed to flow away from the creek as in the case of a fast-flowing river or creek. A further lower value of 10.485±1.985mg/L was observed in February due to similar reasons given above. At this period the level of the creek has become even lower and the prevalent winds and waves in the area due to the harmattan season caused the further reduction in the concentration of total petroleum hydrocarbons value observed. Some of the petroleum hydrocarbon fractions further evaporated away into the air and others may still settle down to the bottom of the creek while others stray away. This observation agreed with Okoye and Okunrobo [32] and USEPA [33]. The more volatile components of the total petroleum hydrocarbons such as gasoline and light fuel oils were easily lost to the air during this period.

A slight increase in the level of total petroleum hydrocarbons value of 15.547±4.590mg/L was observed in April. The increase in concentration possibly was obtained due to increased rainfall in the area which was able to precipitate flared components and also dissolve fractions attached to soil particles and then drain them through adjoining valleys and agricultural farmlands into the creek [31, 34]. A further increase to 24.762±6.492mg/L observed was observed in June in the level of contamination by total petroleum hydrocarbons in the creek. This observation may be due to fact of increased volume of water and the turbulent nature of the flow which provided a medium for the resuspension and resurfacing of total petroleum hydrocarbons in the sediment column of the creek [31, 35]. The inflow of hydrocarbon contaminants from the Orashi River, which is the parent river to the Kolo Creek during this period may also be a reason of such an increase in the level of contamination of the creek by total petroleum hydrocarbons.

Table 1: Mean (X ± SD) Concentrations (mg/L) of Total Petroleum Hydrocarbon in Surface Water of Kolo Creek within the Sampled Months

Carbon Length	**Months**			
	December	**February**	**April**	**June**
C8	0.288±0.013	-	-	-
C9	0.410±0.029	1.531±0.160	-	-
C10	0.423±0.032	-	-	-
C11	0.179±0.006	-	-	-
C12	1.360±0.140	-	-	1.335±0.450
C13	1.226±0.130	-	-	0.181±0.013
C14	0.088±0.003	-	-	0.082±0.006
C15	0.118±0.011	0.378±0.015	-	1.645±0.370
C16	0.024±0.002	-	1.183±0.310	0.896±0.051
C17	0.034±0.002	0.146±0.008	0.022±0,020	2.754±0.810
C18	0.039±0.002	0.733±0.026	-	2.247±0.700
C19	3.058±1.020	2.016±0.800	1.273±0.390	2.042±0.450
C20	2.121±0.630	0.827±0.030	3.914±1.26	1.367±0.410
C21	0.002±0.000	-	0.440±0.15	-
C22	0.010±0.001	0.160±0.006	0.150±0.060	0.840±0.052
C23	0.002±0.000	-	-	-
C24	0.005±0.001	1.194±0.190	2.146±0.600	2.162±0.630
C25	-	-	1.115±0.380	-
C26	1.042±0.090	0.007±0.001	0.422±0.210	2.237±0.590
C27	0.005±0.000	-	-	-
C28	0.008±0.000	1.322±0.410	0.934±0.310	3.425±1.230
C29	0.001±0.000	-	-	-
C30	0.005±0.000	0.658±0.070	2.616±0.810	1.582±0.320
C31	0.008±0.000	-	-	-
C32	0.137±0.009	0.866±0.220	1.332±0.090	0.390±0.050
C33	0.005±0.000	-	-	-
C34	0.006±0.000	0.647±0.049	-	1.577±0.360
C35	0.006±0.000	-	-	-
C36	0.040±0.003	-	-	-
C37	-	-	-	-
C38	-	-	-	-
C39	-	-	-	-
C40	-	-	-	-
Total	10.590±2.113	10.485±1.985	15.547±4.590	24.762±6.492

Figure 1 is an illustration of the mean values of total petroleum hydrocarbons fractions in the water of the Kolo Creek. The mean values ranged from not detected to 2.097mg/L in the months investigated. Total petroleum hydrocarbons of C37 to C40were not detected in the course of investigation in the months. The highest mean concentration value was obtained in the C19 fraction in the study.

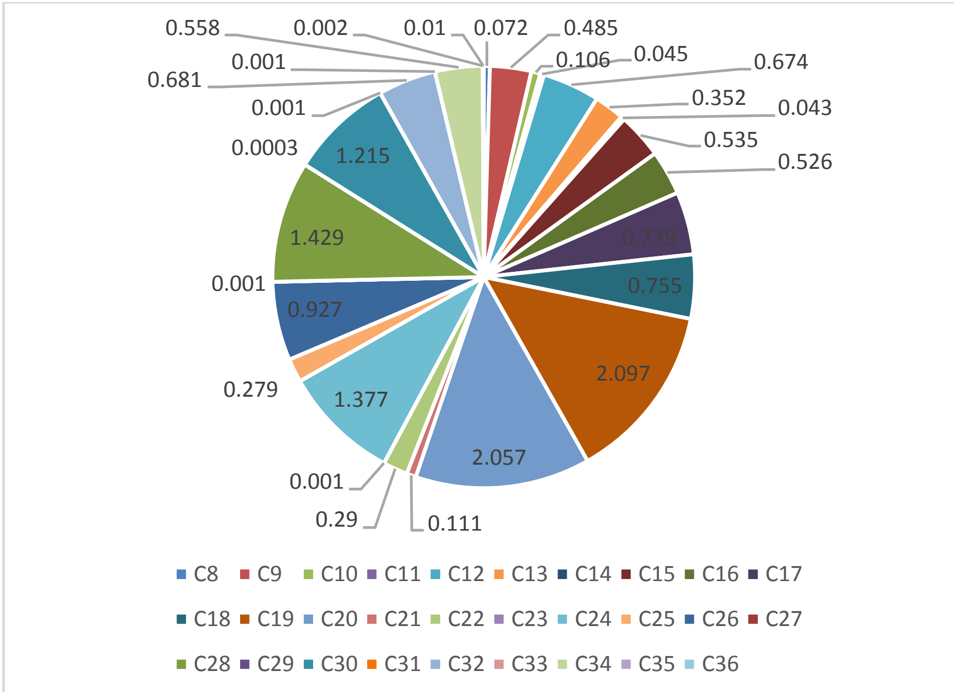

Figure 1: Mean concentration values for the various total petroleum hydrocarbon fractions in the surface water of Kolo Creek within the months under study

The total values of concentration of total petroleum hydrocarbons in the sediments for the various months as shown in Table 2 were; December; 69.3574±24.2836mg/Kg, February; 25.4603±6.3405mg/Kg, April; 36.6575±9.7485mg/Kg and June; 33.4888±7.6412mg/Kg. The results observed revealed that December > April > June > February. The mean monthly concentration of total petroleum hydrocarbons for Kolo Creek was 41.241±0035mg/Kg. The individual total petroleum hydrocarbons fractions found in the sediments of the creek also showed great variations within the months investigated. The C_{10} fraction varied from 8.2759±3.010, 2.2904±0.811, 0.8061±0.290mg/L and not detected for the months of December, February, April and June respectively while the C_{15} fraction varied from 6.9080±2.910, 3.7181±1.125, 1.3775±0.110 and 1.4688±0.104mg/Kg in December, February, April and June respectively. The results from the sediments proved that the source of total petroleum hydrocarbons in the Kolo Creek was due mainly to the activities of man.

The total petroleum hydrocarbons value obtained in December might have arisen due to the intensity of the illegal bunkering activities that took place within the creek at the period. This has similarity to that of Ezekwe and Utong [36] in the Oturuba Creek were the intensity of oil spill led to high pollution rate of total petroleum hydrocarbons in the sediments. The value obtained might have been due to the slow nature of the moving water of the creek coupled with the shallow nature of the creek, for studies have shown that shallow water and slow-moving rivers encourages petroleum hydrocarbons accumulation in the sediments [37-38]. The Kolo Creek is being fed by the Orashi River and during this period the Orashi River usually flow into the Kolo Creek at a very slow rate thereby increasing the total petroleum hydrocarbons content of the creek. The sharp decrease experienced in February may be due to the fact that the water level in the creek has decreased greatly and has exposed the sediments to the sun, thus evaporation of the hydrocarbon fractions was made easy. The total petroleum hydrocarbons contaminants might also have been broken down by micro-organisms [39]. The large quantity of contaminants found in December in the creek were drastically reduced in February due to the activities of bacteria and other micro-organisms and hence reduced the scope or level of pollution in the creek. This finding disagreed with Malmasi *et al*., [40], who observed that wastewater when discharged into the tidal river causes major pollution and the frequent to and fro movement of the tidal water greatly influence and affect the scope and expand the contamination and pollution level.

In April there was an increase in the concentration of total petroleum hydrocarbons in the sediments. Due to the increase in rainfall, petroleum hydrocarbons found their way into the creek from run-offs from the adjoining agricultural lands and self-sustaining refineries (artisanal refining sites) [41] which sank to the bottom of the creek to increase the hydrocarbons content. For petroleum hydrocarbons can easily find their way into the rivers and creeks through drifts during rainfall [42]. Report by Iturbe *et al*., [43] showed that rivers could get contaminated through soil that have been contaminated by total petroleum hydrocarbons through oil spill or from untreated crude oil from storage tanks during rainfall. A decrease was observed in the level of total petroleum hydrocarbons contamination in the month of June. This was primarily due to the increase in the amount of rainfall which increased the turbulence of the creek. This led to increase in the physical chemical and biological activities in the creek, thus resulting in the reduction in the level of contamination of the sediments by total petroleum hydrocarbons. Such activities like resuspension, emulsification drifting and metabolic activities of micro organisms help in reducing pollution of the sediments [44-45].

Table 2: Mean (X ± SD) Concentrations (mg/Kg) of Total Petroleum Hydrocarbon in Sediments of Kolo Creek within the Sampled Months

Carbon Length	Months			
	December	February	April	June
C8	-	-	0.1932±0.003	0.2377±0.010
C9	-	-	0.5955±0.056	-
C10	8.2759±3.010	2.2904±0.811	0.8061±0.290	-
C11	-	2.6424±0.901	1.0963±0.100	0.7040±0.300
C12	-	0.2244±0.010	0.8838±0.321	0.7715±0.310
C13	0.0626±0.005	2.2193±0.732	0.2150±0.051	0.2379±0.012
C14	-	2.3176±0.841	2.1820±0.831	0.9924±0.038
C15	6.9080±2.910	3.7181±1.125	1.3775±0.110	1.4688±0.104
C16	8.9886±3.060	0.0220±0.001	1.1566±0.100	2.0310±0.105
C17	1.0746±0.102	1.8430±0.176	2.8648±0.910	1.6036±0.069
C18	-	3.7319±1.201	0.8382±0.322	3.0444±1.100
C19	5.6404±2.070	1.9970±0.190	6.9611±2.934	1.4361±0.058
C20	0.3014±0.031	0.9644±0.010	3.1102±1.000	5.3570±2.040
C21	10.5495±4.013	0.1965±0.002	0.2496±0.004	-
C22	5.0210±2.010	1.9526±0.180	0.9459±0.001	0.9636±0.009
C23	0.6680±0.069	1.3407±0,160	-	-
C24	-	-	2.7981±0.910	1.4146±0.102
C25	1.0705±0.151	-	-	-
C26	9.9159±3.500	-	2.9600±0.921	1.4305±0.103
C27	-	-	-	-
C28	1.0884±0.100	-	0.8259±0.273	4.3211±1.460
C29	9.6801±3.251	-	-	-
C30	0.0006±0.000	-	1.4886±0.123	3.0602±1.020
C31	0.1119±0.001	-	-	-
C32	-	-	1.5060±0.141	1.0069±0.100
C33	-	-	-	-
C34	-	-	1.7332±0.173	2.3769±0.600
C35	-	-	-	-
C36	-	-	1.9311±0.180	1.0306±0.101
C37	-	-	-	-
C38	-	-	-	-
C39	-	-	-	-
C40	-	-	-	-
Total	69.3574±24.2836	25.4603±6.3405	36.6575±9.7485	33.4888±7.6412

Figure 2 indicated the mean concentration of total petroleum hydrocarbons fractions in the sediment column of the Kolo Creek during the months of investigation. The mean concentration values ranged from not detected to 4.009mg/Kg during the period investigation was carried out. Total petroleum hydrocarbons fractions of C27, C35 and C37-C40 were not detected while the highest mean concentration of 4.009mg/Kg was observed in the C19 fraction in the months investigations were carried out.

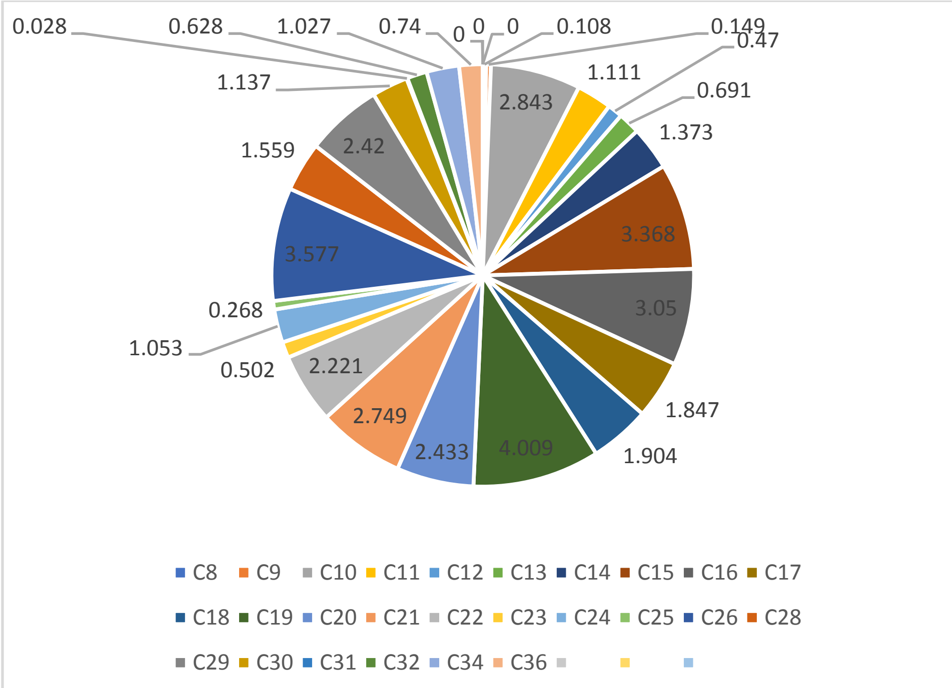

Figure 2: Mean concentration values for the various total petroleum hydrocarbon fractions in the sediments of Kolo Creek within the months under study

The findings from the results of the monthly variation in the surface water and sediments in this work disagrees with Kurylenko and Izosimova [46], who observed that due to the durability of oil, it has long-term exposure and hence tendency to accumulate overtime. The finding of this work agreed with Edori and Kpee [31], whose work revealed that fresh water rivers and creeks undergo flooding annually and hence eliminate or reduce drastically the pollution levels occasioned by total petroleum hydrocarbons and other contaminants no matter the quantity put to the system. From the point of discharge, total petroleum hydrocarbons decrease as it goes along the course of the river or creek of any fresh water system. This happens due to the river dynamics, turbulence, wind and wave [38], which is even aggravated or greatly increased during the months that the river or creek is flooded.

Conclusion

The concentration of total petroleum hydrocarbons in the surface water and sediments of Kolo Creek was determined by using GC-FID and it revealed that the creek was contaminated by total petroleum hydrocarbons. The investigation revealed that the main source of total petroleum hydrocarbons contamination in the creek was anthropogenic. This was due mainly to the illegal activities of oil bunkering and artisanal refining sites found within the creek and its adjoining farmlands. A further proof of the anthropogenicity of the contamination was shown in the variation pattern of the total petroleum hydrocarbons in the surface water and sediments of the creek within the months investigated. As a result of this, management plans should be put in place to dissuade the people from involving in illegal bunkering of crude oil in order to avoid any danger and health issues that may arise from pollution on the environment (creek) since it is the major source of water for the inhabitants.

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