

HEALTH RISK ASSESSMENT OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) IN AQUATIC BIOTICS FROM RIVERS STATE CREEKS

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Abstract

Aquatic biota such as fish, crayfish, and other edible aquatic organisms are susceptible to contamination by hazardous chemicals released from various industrial, agricultural, and domestic sources. These chemicals, particularly Polycyclic Aromatic Hydrocarbons (PAHs), accumulate in the tissues of aquatic organisms, posing a significant threat to human health when consumed. PAHs are organic compounds primarily released into the environment as a result of incomplete combustion, improper disposal, and oil spills from the oil and gas industries. The Rivers State region, with creeks like Isaka and Marine Base, has been severely affected by such pollutants, particularly due to spills and poor waste management practices. PAHs are among the most harmful contaminants, as they are persistent, bioaccumulative, and highly toxic to both aquatic life and humans. These compounds, along with other hydrocarbons such as total hydrocarbons (THC), oil and grease, have significantly polluted the marine ecosystems in this region, leading to adverse effects on biodiversity. The ongoing contamination of these waterways poses a serious environmental and public health challenge. This study assesses the human health risks associated with the consumption of aquatic biota from these polluted creeks, focusing on the levels of PAHs in five selected aquatic species. By analyzing the accumulation of PAHs and other related pollutants, the study aims to evaluate the potential health hazards to humans who rely on these aquatic resources for food. The findings underscore the need for stricter regulation and effective pollution management strategies to mitigate the health risks posed by PAHs and other pollutants in Rivers State's aquatic ecosystems.

Keywords: Polycyclic Aromatic Hydrocarbons (PAHs), Aquatic Biota, Environmental Pollution, Human Health Risk, Rivers State

1. Introduction

Aquatic biotics like fish, crayfish, and other edible aquatic lives are prone to hazardous chemicals released from industrial, agricultural, and public sources. Most of these chemicals accrue in the tissues of these biotics which pose threat to human health when consumed (Wu et al 2012). One of such contaminants is PAHs.

Polycyclic Aromatic Hydrocarbons (PAHs) are organic compounds released into the environment because of the incomplete combustion, theft, and poor handling of oil and gas, crude oil, and industrial wastes (Adeniji et al., 2017). This has specifically led to spill and pollution of the waterways and creeks. Isaka and Marine Base creeks are examples of such creeks in the riverine areas of Rivers State that is affected by spills.

According to Iyama et al., (2017), the diverse bases of marine pollution can be home-made (domestic), industrial or natural. Also, report has revealed that organic dirt is responsible for the worsening and pollution of several creeks in Rivers State (Duru et al., 2018). Compounds of hydrocarbons such as, THC, Oil and Grease, PAHs etc. form the typical organic pollutants from many sources that are present in the marine ecology in Rivers State (Adeniji et al., 2017). These compounds are lethal to man, animals, and plants (e.g., PAHs are very recalcitrant and noxious to the biosystem).

The existence of PAHs have been reported in different environmental media including fish and crustaceans in areas like Bonny (Nkpaa et al, Nwachi and Ntorgbo, 2016). PAHs have received notable attention recently due to their highly carcinogenic possibilities (Wu et al); and as such, it is worthy to note that residual levels of PAHs in fish, lobsters, and other seafoods could lead to adverse health challenges in humans (Ilobet et al, 2006).

Hence the study to evaluate the level of PAHs in five aquatic biotics, mostly consumed by man within Isaka and Marine base areas and the potential risk to human health from consumption.

2. Materials and Methods

2.1 Description of the Study Area

The study was conducted in Isaka and Marine Base creeks in Okrika and Port Harcourt LGAs, Rivers State. Isaka creek is a salty H₂O body located in Isaka, Okrika L.G.A of Rivers State, Nigeria. It is an armlet of the National Ports Authorities highway-sea that lies between Latitude

4°73 North and Longitude 6°99 East. The river is decked by mangrove vegetation by the sides. However, wastes originating from dumped boats, open bathrooms, municipal trashes, and spills emanating from oil thefts have escalated the unhealthy status of the creek.

Marine base is situated in Port Harcourt at Latitudes 04° 43' and 04° 57' North of the Equator and between Longitudes 06° 53' and 07° 58' East of the Greenwich Meridian and is bounded by the Dockyard creek, Bonny River and Amadi creek, of the Niger Delta, at an elevation of about 12m above sea level. It is roughly 60km from the crest up stream of the Bonny River. Marine base is in the coastal area of Port Harcourt, where loading and off-loading of crude oil and other petroleum products by ships, cargo oil tankers occur. The marine base surrounding is not free from contaminants from petrol products, effluents from industries and garbage migrating via drains from hinterland. Thus, has a direct impact on the inhabitant H₂O organisms such as mudskippers, periwinkles, oysters, and crabs as well as the consumers.

2.2 Sampling method and procedure.

The sampling method for collection of the biotics in each of the three locations within the creeks is a simple random method, where homogeneous species of each of the biotics (fish, crayfish, crab, periwinkle, and oyster) were selected at three different locations per creek on different days.

The samples were collected in three different basins containing water from each of the creeks. These samples were then taken to the laboratory and a portion of each of the biotics evaluated for PAHs using Gas Chromatography/Flame Ionization Detection Analysis.

2.3 Materials and Reagents

- Weighing balance, 250ml extraction bottle, Glass funnel, Separator funnel, Glass wool, Fume hood, Water bath, 250ml beakers, 100ml measuring cylinders, Pipette, Volumetric flask, GC/FID
- Hexane, Methanol, Potassium Hydroxide, PAH Standard, Anhydrous Sodium Sulphate, Dichloromethane (DCM)

2.3.1 Standards and Reagent Preparation:

MeOH/ KOH solution: weigh 120g of KOH, add 60ml of water and dilute with 900ml Methanol.

MeOH/H₂O Mixture: Measure 400ml methanol into a 500ml volumetric flask and add 100ml distilled water to mark.

2.4 Digestion procedure of the collected samples

The processes involve saponification of the Biota, extraction of the hydrocarbon for PAH analysis and washing of the organic extract.

About 5.00g of the biota sample was taken into an extraction bottle, saponified with MeOH/KOH solution in a H₂O bath for 60 minutes at 60°C. The sample was cooled and extracted with hexane at least twice using separator funnel. The hexane extract was further washed with MeOH/Water to ensure effective saponification. The hexane layer extract was received in beaker and allowed to concentrate to 1ml. The 1ml concentrate was further passed through a column bed packed with oven preheated silica gel; then prior to dryness 10ml of hexane was introduced on the column to elute for Aliphatic components. Immediately the hexane drains down and prior to dryness 10ml of DCM solvent was introduced to elute the PAH components. It was afterward received in a beaker, allowed to concentrate to 1 ml and then packed into 2ml vial ready for GC/FID analysis.

2.5 Data Interpretation

Descriptive statistics was adopted to depict the average, range, mean and standard deviations (SD) of the assayed noxious elements in the aquatic biotics specimens. The test of significance and oneway ANOVA were also employed to depict the causes of PAHs in the studied site, the contamination dynamics and potential linkage among established variables. Statistical analysis was done using IBM SPSS 23 software.

2.6 Health Risk Analysis to Humans

Human wellbeing toxicity was assayed by deducing the probable carcinogenic and noncarcinogenic impact of exposure to PAHs through consumption of contaminated aquatic biotic over a time. The assessment was carried out for adults (70kg) for both non-carcinogenic and carcinogenic health risk.

2.6.1 Chronic daily intake (CDI) indices:

The Chronic daily intake of PAHs through biotics ingestion was deduced from the equation: $CDI_{Ingestion} = CS \times IRS / BW$ (1)

Where:

CDI = chronic day-by-day intake; CS = Concentration of PAHs in the biotics (mg/kg); BW = body weight = 70 kg (USEPA, 1992); IRS = Ingestion Rate = 0.33mg/kg/d for crab and 0.85mg/kg/d for fish and other biotics.

2.6.2 Hazard quotient (HQ) indices: The HQ for **non-carcinogenic** menace was arrived at by the equation:

$HQ = CDI / RfD$ (2)

Where:

The non-cancer hazard quotient (HQ) is the ratio of exposure to perilous substances, and RfD is the chronic reference dose of the toxicant ($mg \cdot kg^{-1} \cdot d^{-1}$).

According to database (USEPA, 2009b), the oral toxicity indication dose values (RfD) for the PAHs are 0.02mg/kg/day for Naphthalene, 0.04mg/kg/day each for Fluorene, Anthracene, Phenanthrene, Fluoranthene, and Benzo(ghi)perylene.

The exposed population is assumed to be safe when $HQ < 1$ and unsafe when $HQ \geq 1$ (Khan et al., 2008).

2.6.3 Chronic hazard index (HI)

Chronic Hazard Index (CHI / CHQ) = $\sum_k CDI_k / RfD_k$ (3) Where:

CHI is the aggregate of more than one Hazard Quotient for multi-substances or exposure routes, CDI_k is the day-by-day intake of a noxious substance (k) and RfD_k is the chronic indication dose for the noxious substance k. HI values ≥ 1 shows that there is a chance that non-carcinogenic risk may emanate and when $HI < 1$, the turnaround is the case.

2.6.4 Cancer risk (CR)

Cancer risk was deduced from: $Cancer\ risk\ (CR) = CDI \times SF$ (4)

where: Cancer risk (CR) depicts the prospect of self's lifetime health menace from carcinogens. CDI is the chronic day-by-day carcinogens intake ($mg \cdot kg^{-1} \cdot d^{-1}$); SF_k is the slope factor or Toxic Equivalency Factor (TEF) for each congener k ($mg \cdot kg^{-1} \cdot d^{-1}$).

The Carcinogenic potency of individual PAHs was deduced from: $B(A)P_{teq} = PAH_i \times TEF_i$ (5)

where: PAH_i is the concentration of individual PAH congeners; TEF_i , the toxicity equivalency factor for each congener.

The carcinogenic harmfulness of the high molecular mass PAHs seen in the specimens harnessed from the study sites were assayed relative to benzo[a]pyrene (with enough toxicological data) using toxic equivalent quotient (TEQ). The high molecular high mass PAHs are BaA, Chry, BbF, BkF, BaP, DiahA, Inpy, and BghiP; and their corresponding TEF values used in these calculations were 0.1, 0.01, 0.1, 0.1, 1, 1, 0.1, and 0.01 (Van den Berg et al., 2006; CCME 2010; Lerda 2011; Benson et al., 2017; Zhao et al., 2017).

Excess Cancer Risk (ECR) = $\Sigma Q \times B(A)P_{teq} \times IFR \times ED / BW \times AT_n$ (6) where:

3.0. Results and Discussions 3.1 Results of Recovery Analysis

Table 3.1.1 Concentration of PAHs in Aquatic Biotics, Isaka Creek.

Chrysene	0.12±0.00	0.13±0.00	0.1±0.00	0.1±0.00	0.107±0.00
Benzo(b)Fluoranthene	0.01±0.00	0.01±0.00	0.07±0.00	0.07±0.00	0.003±0.00
Benzo(k)Fluoranthene	0.023±0.001	0.04±0.00	0.04±0.00	0.04±0.00	0.019±0.00
Benzo(a) Pyrene	0.06±0.00	0.09±0.00	0.13±0.00	0.13±0.00	0.043±0.00
Benzo(a,h)anthracene	0.03±0.00	0.03±0.00	0.5±0.00	0.5±0.00	1.355±0.00
Indeno(1,2,3-cd) pyrene	0.02±0.01	0.07±0.00	0.034±0.002	0.037±0.00	0.018±0.00
Benzo (ghi) Perylene	1.782±0.02	0.61±0.00	0.03±0.00	0.03±0.00	0.028±0.00
Total PAHs (ΣPAH)	2.79±0.051	1.58±0.00	2.038±0.004	2.044±0.00	1.932±0.00
Total Carcinogenic PAHs (	2.105±0.04	1.02±0.00	0.964±0.002	0.967±0.00	1.618±0.00
Parameters (mg/kg)	Fish	Periwinkle	Crab	Oyster	Prawn
Naphthalene	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00
Acenaphthylene	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00
Acenaphthene	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00	0.01±0.00
Fluorene	0.03±0.00	0.01±0.00	0.034±0.002	0.037±0.00	0.028±0.00
Anthracene	0.1±0.00	0.07±0.00	0.12±0.00	0.12±0.00	0.096±0.00
Phenanthrene	0.173±0.02	0.08±0.00	0.18±0.00	0.18±0.00	0.102±0.00
Fluoranthene	0.3±0.00	0.26±0.00	0.5±0.00	0.5±0.00	0.027±0.00
Pyrene	0.04±0.00	0.11±0.00	0.21±0.00	0.21±0.00	0.031±0.00
Benz(a)anthracene	0.06±0.00	0.04±0.00	0.06±0.00	0.06±0.00	0.045±0.00

Σ cPAH)

The result of Table 3.1.1 are the concentrations of PAHs in the various aquatic biotics in Isaka creek. A total of sixteen PAHs were examined; and the result of the experimentation shows that

Naphthalene, Acenaphthylene, and Acenaphthene are of the same concentration of $0.01 \pm 0.00 \text{ mg/kg}$ in the five biotics. The concentration of Fluorine was found to be $0.03 \pm 0.00 \text{ mg/kg}$ in fish, $0.034 \pm 0.00 \text{ mg/kg}$ in crab, $0.028 \pm 0.00 \text{ mg/kg}$ in prawn, $0.01 \pm 0.00 \text{ mg/kg}$ in periwinkle and $0.037 \pm 0.00 \text{ mg/kg}$ in oyster. For that of Anthracene, mean value of $0.1 \pm 0.00 \text{ mg/kg}$ was observed in both fish and prawn, $0.12 \pm 0.00 \text{ mg/kg}$ in both crab and oyster, and $0.07 \pm 0.00 \text{ mg/kg}$ in periwinkle. Phenanthrene on the other hand was examined to be of a conc. of $0.18 \pm 0.00 \text{ mg/kg}$ in crab and oyster, $0.173 \pm 0.02 \text{ mg/kg}$ in fish, $0.08 \pm 0.00 \text{ mg/kg}$ in periwinkle and $0.102 \pm 0.00 \text{ mg/kg}$ in prawn. The concentration of Fluoranthene, was found to be $0.5 \pm 0.00 \text{ mg/kg}$ in crab and oyster, $0.3 \pm 0.00 \text{ mg/kg}$ in fish, $0.027 \pm 0.00 \text{ mg/kg}$ in prawn, and $0.26 \pm 0.00 \text{ mg/kg}$ in periwinkle. Pyrene content was found to be, $0.04 \pm 0.00 \text{ mg/kg}$ in fish and $0.03 \pm 0.00 \text{ mg/kg}$ in prawn, $0.21 \pm 0.00 \text{ mg/kg}$ for crab and oyster and $0.11 \pm 0.00 \text{ mg/kg}$ for periwinkle. The mean concentration of Benz(a)anthracene was found to be $0.06 \pm 0.00 \text{ mg/kg}$ in fish, crab, and oyster, and $0.045 \pm 0.00 \text{ mg/kg}$ in prawn, and $0.04 \pm 0.00 \text{ mg/kg}$ in periwinkle. For Chrysene, $0.12 \pm 0.00 \text{ mg/kg}$ concentration was found in fish, $0.107 \pm 0.00 \text{ mg/kg}$ in prawn, $0.10 \pm 0.00 \text{ mg/kg}$ in crab and oyster and $0.13 \pm 0.00 \text{ mg/kg}$ in periwinkle. Benzo(b)Fluoranthene has concentrations of $0.01 \pm 0.00 \text{ mg/kg}$ in fish and periwinkle, $0.003 \pm 0.00 \text{ mg/kg}$ in prawn, and $0.07 \pm 0.00 \text{ mg/kg}$ in both crab and oyster. The concentration of Benzo(k)Fluoranthene was found to be $0.04 \pm 0.00 \text{ mg/kg}$ in periwinkle, crab, and oyster, and $0.019 \pm 0.00 \text{ mg/kg}$ and $0.023 \pm 0.01 \text{ mg/kg}$ in prawn and fish individually. Benzo(a) Pyrene concentration was found to be $0.06 \pm 0.00 \text{ mg/kg}$ in fish, $0.043 \pm 0.00 \text{ mg/kg}$ in prawn, $0.09 \pm 0.00 \text{ mg/kg}$ in periwinkle and $0.13 \pm 0.00 \text{ mg/kg}$ in crab and oyster. For Benzo(a,h)anthracene, concentrations of $0.03 \pm 0.00 \text{ mg/kg}$ was found in fish and periwinkle, $0.5 \pm 0.00 \text{ mg/kg}$ in crab and oyster; and concentration of $1.355 \pm 0.00 \text{ mg/kg}$ in prawn. The conc. of Indeno (1,2,3-cd) pyrene was $0.02 \pm 0.001 \text{ mg/kg}$ in fish; $0.07 \pm 0.00 \text{ mg/kg}$, $0.034 \pm 0.002 \text{ mg/kg}$, $0.037 \pm 0.00 \text{ mg/kg}$, and $0.018 \pm 0.00 \text{ mg/kg}$ in periwinkle, crab, oyster, and prawn correspondingly. For Benzo(ghi) Perylene, a conc. of $0.03 \pm 0.00 \text{ mg/kg}$ was observed in crab and oyster, $1.78 \pm 0.002 \text{ mg/kg}$ in fish, $0.61 \pm 0.00 \text{ mg/kg}$ in periwinkle, and $0.028 \pm 0.00 \text{ mg/kg}$ in prawn.

Mean concentrations for total carcinogenic PAHs (sum of BaA, Chr, BkFL, Bap, BbFL, Ind, DBA, BP) accounted for **84%, 75%, 65%, 47%, and 47%** respectively in prawn, fish, periwinkle, crab, and oyster of the total PAHs. Total mean carcinogenic PAHs (ΣcPAH) and Total mean PAH (ΣPAH) concentrations were higher in fish than other biotics, with values of 2.105 mg/kg and 2.79 mg/kg respectively.

For individual concentration of PAHs, Benzo(ghi)Perylene was the most dominant congener in fish and periwinkle samples, and the concentrations are significantly higher than the other congeners, with mean concentrations of $1.782 \pm 0.02 \text{ mg/kg}$ and $0.61 \pm 0.00 \text{ mg/kg}$, accounting for 64% and 39% of the total PAHs in fish and periwinkle respectively. Fluoranthene and Benzo(a,h)anthracene were the most dominant congeners in crab

and oyster with the same mean concentration of 0.05 ± 0.0 mg/kg and percentage contribution of 24.5%. For prawn, Benzo(a,h)anthracene happen to be the most predominant congener with a mean concentration of 1.355 ± 0.00 mg/kg amounting to 70% of the total level of PAHs in prawn. Benzo(a,h)anthracene concentration in prawn was significantly higher than the other congeners.

The PAH composition shape by ring type showed a considerable predominance of the three-ring, four-ring type, and five-ring type PAHs. The mean percentage concentration of the higher molecular weight PAHs (HMWPAHs) (four to six rings) was higher than the lower molecular weight PAHs (LMWPAHs) (two to three rings) in all the aquatic biotics with fish and periwinkle accounting for 88% each, prawn 87%, and crab and oyster 82% each in Isaka creek.

Table 3.1.2 Concentration of PAHs in Aquatic Biotics, Marine Base Creek

Parameters	Fish	Periwinkle	Crab	Oyster	Prawn
Naphthalene	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
Acenaphthylene	0.03 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
Acenaphthene	0.06 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
Fluorene	0.08 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	0.04 ± 0.00	0.01 ± 0.00
Anthracene	0.13 ± 0.00	0.012 ± 0.00	0.01 ± 0.00	0.12 ± 0.00	0.05 ± 0.00
Phenanthrene	0.11 ± 0.00	0.013 ± 0.00	0.02 ± 0.00	0.18 ± 0.00	0.06 ± 0.00
Fluoranthene	0.38 ± 0.00	0.119 ± 0.00	0.01 ± 0.00	0.5 ± 0.00	0.17 ± 0.00
Pyrene	0.47 ± 0.00	0.064 ± 0.00	0.13 ± 0.00	0.21 ± 0.00	0.35 ± 0.00
Benz(a)anthracene	0.41 ± 0.00	0.017 ± 0.00	0.1 ± 0.00	0.06 ± 0.00	0.16 ± 0.00
Chrysene	0.35 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.1 ± 0.00	0.14 ± 0.00
Benzo(b)Fluoranthene	0.33 ± 0.00	0.017 ± 0.00	0.05 ± 0.00	0.07 ± 0.00	0.07 ± 0.00
Benzo(k)Fluoranthene	0.02 ± 0.00	0.044 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	0.04 ± 0.00
Benzo(a) Pyrene	0.59 ± 0.00	0.089 ± 0.00	0.07 ± 0.00	0.15 ± 0.00	0.98 ± 0.00
Benzo(a,h)anthracene	0.01 ± 0.00	0.752 ± 0.00	0.05 ± 0.00	0.6 ± 0.00	0.47 ± 0.00
Indeno(1,2,3-cd) pyrene	0.03 ± 0.00	0.054 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	0.08 ± 0.00

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Benzo (ghi) Perylene	5.63±0.00	0.097±0.00	1.91±0.00	0.03±0.00	3.36±0.00	Total
PAHs (ΣPAH)	8.64±0.00	1.388±0.00	2.55±0.00	2.17±0.00	5.97±0.00	Total
Carcinogenic PAHs (ΣcPAH)		7.37±0.00	1.14±0.00	2.34±0.00	1.09±0.00	
5.30±0.00						

The results of PAHs analysis in the various aquatic biotics in Marine Base Creek are shown in Table 3.1.2. The result of the experimentation has it that Naphthalene is of the same concentration of $0.01 \pm 0.00 \text{ mg/kg}$ in the five aquatic biotics.

Similarly, it was found out that Acenaphthylene and Acenaphthene are of the same concentration of $0.01 \pm 0.00 \text{ mg/kg}$ in four of the other biotics except the fish with $0.03 \pm 0.00 \text{ mg/kg}$ and $0.06 \pm 0.00 \text{ mg/kg}$ correspondingly.

For Fluorine, concentrations of 0.08 mg/kg was found in fish, $0.01 \pm 0.00 \text{ mg/kg}$ in crab, periwinkle, and prawn and $0.04 \pm 0.00 \text{ mg/kg}$ in oyster. The concentration of Anthracene was found to be $0.13 \pm 0.00 \text{ mg/kg}$ in fish, $0.012 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.12 \pm 0.00 \text{ mgkg}^{-1}$ in crab and $0.05 \pm 0.00 \text{ mg/kg}$ in prawn, while that of Phenanthrene was found to be $0.11 \pm 0.00 \text{ mg/kg}$ in fish, $0.013 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, $0.02 \pm 0.00 \text{ mg/kg}$ in crab, $0.18 \pm 0.00 \text{ mg/kg}$ in oyster, and $0.06 \pm 0.00 \text{ mg/kg}$ in prawn. For Fluoranthene, concs. of $0.38 \pm 0.00 \text{ mg/kg}$ were found in fish, $0.119 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, $0.01 \pm 0.00 \text{ mgkg}^{-1}$ in crab, $0.5 \pm 0.00 \text{ mgkg}^{-1}$ in oyster, and $0.17 \pm 0.00 \text{ mgkg}^{-1}$ in prawn, while Pyrene was found to be of a concentration of $0.47 \pm 0.00 \text{ mg/kg}$ in fish, $0.064 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.13 \pm 0.00 \text{ mg/kg}$ in crab, $0.21 \pm 0.00 \text{ mg/kg}$ in oyster and $0.35 \pm 0.00 \text{ mg/kg}$ in prawn. on the other hand, Benz(a)anthracene was found to be of concentrations of $0.41 \pm 0.00 \text{ mg/kg}$ in fish, $0.017 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.1 \pm 0.00 \text{ mg/kg}$ in crab, $0.06 \pm 0.00 \text{ mgkg}^{-1}$ in oyster, and $0.16 \pm 0.00 \text{ mgkg}^{-1}$ in prawn.

In the same vein, varying concentrations of Chrysene were observed in the biotics. Concentrations of $0.3 \pm 0.00 \text{ mg/kg}$ was found in fish, $0.07 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, $0.08 \pm 0.00 \text{ mg/kg}$ in crab, $0.1 \pm 0.00 \text{ mgkg}^{-1}$ in oyster and $0.14 \pm 0.00 \text{ mgkg}^{-1}$ in prawn. For Benzo(b)Fluoranthene, conc. of $0.33 \pm 0.00 \text{ mg/kg}$ was found in fish, $0.017 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.05 \pm 0.00 \text{ mg/kg}$ in crab and $0.07 \pm 0.00 \text{ mg/kg}$ in oyster and prawn. Benzo(k)Fluoranthene was found to be of the same concentration of 0.04 mg/kg in crab, oyster, and prawn, and varying concentration of $0.02 \pm 0.00 \text{ mgkg}^{-1}$ in fish and $0.044 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, while Benzo(a) Pyrene was $0.59 \pm 0.00 \text{ mg/kg}$ in fish, $0.089 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.07 \pm 0.00 \text{ mg/kg}$ in crab, $0.15 \pm 0.00 \text{ mgkg}^{-1}$ in oyster and $0.98 \pm 0.00 \text{ mgkg}^{-1}$ in prawn. The concentration of Benzo(a,h)anthracene was observed to be 0.01 mg/kg in fish, $0.752 \pm 0.00 \text{ mg/kg}$ in periwinkle, $0.05 \pm 0.00 \text{ mg/kg}$ in crab, $0.6 \pm 0.00 \text{ mg/kg}$ in oyster and $0.47 \pm 0.00 \text{ mg/kg}$ in prawn. And for Indeno(1,2,3-cd) pyrene, mean value concentration of $0.04 \pm 0.00 \text{ mg/kg}$ was found in crab and oyster, $0.03 \pm 0.00 \text{ mgkg}^{-1}$ in fish, $0.054 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, and $0.08 \pm 0.00 \text{ mgkg}^{-1}$ in prawn; whereas conc. of

Benzo (ghi) Perylene was $5.63 \pm 0.00 \text{ mg/kg}$ in fish, $0.097 \pm 0.00 \text{ mgkg}^{-1}$ in periwinkle, $1.91 \pm 0.00 \text{ mg/kg}$ in crab, 0.03 mg/kg in oyster, and $3.36 \pm 0.00 \text{ mg/kg}$ in prawn.

Mean concentrations for total carcinogenic PAHs (sum of BaA, Chr, BkFL, Bap, BbFL, Ind, DBA, BP) accounted for **92%, 89%, 85%, 82%, and 50%**, respectively in crab, prawn, fish, periwinkle, and oyster of the total PAHs.

Total mean carcinogenic PAHs concentrations were higher in fish

(7.37 mg/kg) than other biotics and differences in concentrations between the species is quite significant. Total mean PAH concentrations were also higher in fish (8.64 mg/kg) than the other biotics.

For individual concentration of PAHs, Benzo(ghi)Perylene was the most dominant congener in crab, fish, and prawn samples, and the concentrations are significantly higher than the other congeners, with mean concentrations of $1.91 \pm 0.00 \text{ mg/kg}$, $5.63 \pm 0.00 \text{ mg/kg}$, and $3.36 \pm 0.00 \text{ mg/kg}$, accounting for 75%, 65%, and 56% of the total PAHs in crab, fish, and prawn respectively. Benzo(a,h)anthracene was the most dominant congener in periwinkle and oyster with mean concentrations of $0.752 \pm 0.0 \text{ mg/kg}$ and $0.60 \pm 0.0 \text{ mg/kg}$ respectively, accounting for 54% and 28% of the cumulative PAHs in both species.

The PAH composition pattern by ring type from marine base creek also showed a considerable predominance of the three-ring, four-ring, and five-ring type PAHs. The mean percentage concentration of the higher molecular weight PAHs (HMWPAHs) (four to six rings) was higher than the lower molecular weight PAHs (LMWPAHs) (two to three rings) in all the aquatic biotics with crab, prawn, fish/periwinkle, and oyster accounting for 100%, 98%, 95%, and 83% respectively. Differences in concentrations between the HMWPAHs and LMWPAHs among the species were also statistically significant.

3.2 Human Health Risk Index from consumption PAHs via the biotics

Table 3.2.1. Human HRI from consumption of PAHs in Fish, Isaka Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.00607143		
Acenaphthylene	0.01	0.000121	NR			
Acenaphthene	0.01	0.000121	NR			
Fluorene	0.03	0.000364	0.04	0.00910714		
Anthracene	0.1	0.001214	0.04	0.03035714		
Phenanthrene	0.173	0.002101	0.04	0.05251786		
Fluoranthene	0.3	0.003643	0.04	0.09107143		
Pyrene	0.04	0.000486	NR			

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Benz(a)anthracene	0.06	0.000729	NR		0.1	7.28571E-05
Chrysene	0.12	0.001457	NR		0.01	1.45714E-05
Benzo(b)Fluoranthene	0.01	0.000121	NR		0.1	1.21429E-05
Benzo(k)Fluoranthene	0.023	0.000279	NR		0.1	2.79286E-05
Benzo(a) Pyrene	0.06	0.000729	NR		1	0.000728571
Benzo(a,h)anthracene	0.03	0.000364	NR		1	0.000364286
Indeno(1,2,3-cd) pyrene	0.02	0.000243	NR		0.1	2.42857E-05
Benzo (ghi) Perylene	1.794	0.021784	0.04	0.54460714	0.01	0.000217843

$\Sigma HQ = 0.733732143$; $\Sigma CR = 0.001462486$

Table 3.2.2. Human HRI from consumption of PAHs in Periwinkle, Isaka Creek

PAHs Conc. (mg/kg)	Mean	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.006071		
Acenaphthylene	0.01	0.000121	NR			
Acenaphthene	0.01	0.000121	NR			
Fluorene	0.01	0.000121	0.04	0.003036		
Anthracene	0.07	0.00085	0.04	0.02125		
Phenanthrene	0.08	0.000971	0.04	0.024286		
Fluoranthene	0.26	0.003157	0.04	0.078929		
Pyrene	0.11	0.001336	NR			
Benz(a)anthracene	0.04	0.000486	NR		0.1	4.86E-05
Chrysene	0.13	0.001579	NR		0.01	1.58E-05
Benzo(b)Fluoranthene	0.01	0.000121	NR		0.1	1.21E-05
Benzo(k)Fluoranthene	0.04	0.000486	NR		0.1	4.86E-05
Benzo(a) Pyrene	0.09	0.001093	NR		1	0.001093
Benzo(a,h)anthracene	0.03	0.000364	NR		1	0.000364
Indeno(1,2,3-cd) pyrene	0.07	0.00085	NR		0.1	0.000085
Benzo (ghi) Perylene	0.61	0.007407	0.04	0.185179	0.01	7.41E-05

$\Sigma HQ = 0.31875$; $\Sigma CR = 0.001741$

Table 3.2.3. Human HRI from consumption of PAHs in Crab, Isaka Creek

PAHs Conc. (mg/kg)	Mean	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	4.71429E-05	0.02	0.002357		
Acenaphthylene	0.01	4.71429E-05	NR			
Acenaphthene	0.01	4.71429E-05	NR			
Fluorene	0.034	0.00016029	0.04	0.004007		
Anthracene	0.12	0.00056571	0.04	0.014143		
Phenanthrene	0.18	0.00084857	0.04	0.021214		
Fluoranthene	0.5	0.00235714	0.04	0.058929		
Pyrene	0.21	0.00099	NR			
Benz(a)anthracene	0.06	0.00028286	NR		0.1	2.82857E-05
Chrysene	0.1	0.00047143	NR		0.01	4.71429E-06
Benzo(b)Fluoranthene	0.07	0.00033	NR		0.1	0.000033
Benzo(k)Fluoranthene	0.04	0.00018857	NR		0.1	1.88571E-05
Benzo(a) Pyrene	0.13	0.00061286	NR		1	0.000612857
Benzo(a,h)anthracene	0.5	0.00235714	NR		1	0.002357143
Indeno(1,2,3-cd) pyrene	0.034	0.00016029	NR		0.1	1.60286E-05
Benzo (ghi) Perylene	0.03	0.00014143	0.04	0.003536	0.01	1.41429E-06

$\Sigma HQ = 0.104186$; $\Sigma CR = 0.0030723$

Table 3.2.4. Human HRI from consumption of PAHs in Oyster, Isaka Creek

PAHs Conc. (mg/kg)	Mean	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.006071		
Acenaphthylene	0.01	0.000121	NR			
Acenaphthene	0.01	0.000121	NR			
Fluorene	0.037	0.000449	0.04	0.011232		
Anthracene	0.12	0.001457	0.04	0.036429		

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Phenanthrene	0.18	0.002186	0.04	0.054643		
Fluoranthene	0.5	0.006071	0.04	0.151786		
Pyrene	0.21	0.00255	NR			
Benz(a)anthracene	0.06	0.000729	NR		0.1	7.29E-05
Chrysene			NR		0.0	
	0.1	0.001214			1	1.21E-05
Benzo(b)Fluoranthene	0.07	0.00085	NR		0.1	0.000085
Benzo(k)Fluoranthene	0.04	0.000486	NR		0.1	4.86E-05
Benzo(a) Pyrene	0.13	0.001579	NR		1	0.001579
Benzo(a,h)anthracene	0.5	0.006071	NR		1	0.006071
Indeno(1,2,3-cd) pyrene	0.037	0.000449	NR		0.1	4.49E-05
Benzo (ghi) Perylene		0.000364	0.04	0.009107	0.0	3.64E-06
	0.03				1	

$\Sigma HQ = 0.269268$; $\Sigma CR = 0.007917$.

Table 3.2.5. Human HRI from consumption of PAHs in Prawn, Isaka Creek

PAHs Conc. (mg/kg)	Mean	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	4.71429E-05	0.02	0.002357		
Acenaphthylene	0.01	4.71429E-05	NR			
Acenaphthene	0.01	4.71429E-05	NR			
Fluorene	0.028	0.000132	0.04	0.0033		
Anthracene	0.096	0.000452571	0.04	0.011314		
Phenanthrene	0.102	0.000480857	0.04	0.012021		
Fluoranthene	0.027	0.000127286	0.04	0.003182		
Pyrene	0.031	0.000146143	NR			
Benz(a)anthracene	0.045	0.000212143	NR		0.1	2.12E-05
Chrysene	0.107	0.000504429	NR		0.01	5.04E-06
Benzo(b)Fluoranthene	0.003	1.41429E-05	NR		0.1	1.41E-06
Benzo(k)Fluoranthene	0.019	8.95714E-05	NR		0.1	8.96E-06
Benzo(a) Pyrene	0.043	0.000202714	NR		1	0.000203

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Benzo(a,h)anthracene	1.355	0.006387857	NR		1	0.006388
Indeno(1,2,3-cd) pyrene	0.018	8.48571E-05	NR		0.1	8.49E-06
Benzo (ghi) Perylene	0.028	0.000132	0.04	0.0033	0.01	1.32E-06

$\Sigma HQ = 0.035475$; $\Sigma CR = 0.006637$

The estimated **hazard quotients** (HQs) of PAHs from consumption of fish, periwinkle, crab, oyster, and prawn from the surface water from Isaka creek were 0.733732143, 0.31875, 0.104186, 0.269268, and 0.035475 respectively as shown in **tables 3.2.1 to 3.2.5**. The result indicates that HQs for all the aquatic biotics in the water is below the USEPA measurement limit of one. Thus, the possibility of human contacting any **non-carcinogenic** ailment is very unlikely. However, the **cancer risk(CR)** values were above the acceptable range; and as such there is the likelihood of cancer upon continuous exposure. The calculated CR values were 0.00146248, 0.001741, 0.0030723, 0.007917, and 0.006637 in fish, periwinkle, crab, oyster, and prawn respectively.

Table 3.2.6. Human HRI from consumption of PAHs in Fish, Marine Base Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.006071		
Acenaphthylene	0.03	0.000364	NR			
Acenaphthene	0.06	0.000729	NR			
Fluorene	0.08	0.000971	0.04	0.024286		
Anthracene	0.13	0.001579	0.04	0.039464		
Phenanthrene	0.11	0.001336	0.04	0.033393		
Fluoranthene	0.38	0.004614	0.04	0.115357		
Pyrene	0.47	0.005707	NR			
Benz(a)anthracene	0.41	0.004979	NR		0.1	0.000498
Chrysene	0.35	0.00425	NR		0.01	4.25E-05
Benzo(b)Fluoranthene	0.33	0.004007	NR		0.1	0.000401
Benzo(k)Fluoranthene	0.02	0.000243	NR		0.1	2.43E-05
Benzo(a) Pyrene	0.59	0.007164	NR		1	0.007164
Benzo(a,h)anthracene	0.01	0.000121	NR		1	0.000121
Indeno(1,2,3-cd) pyrene	0.03	0.000364	NR		0.1	3.64E-05
Benzo (ghi) Perylene	5.63	0.068364	0.04	1.709107	0.01	0.000684

$\Sigma HQ = 1.927679$; $\Sigma CR = 0.008971$

Table 3.2.7. Human HRI from consumption of PAHs in Periwinkle, Marine Base Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.006071		
Acenaphthylene	0.01	0.000121	NR			
Acenaphthene	0.01	0.000121	NR			
Fluorene	0.01	0.000121	0.04	0.003036		
Anthracene	0.012	0.000146	0.04	0.003643		
Phenanthrene	0.013	0.000158	0.04	0.003946		
Fluoranthene	0.119	0.001445	0.04	0.036125		
Pyrene	0.064	0.000777	NR			
Benz(a)anthracene	0.017	0.000206	NR		0.1	2.06E-05
Chrysene	0.07	0.00085	NR		0.01	8.5E-06
Benzo(b)Fluoranthene	0.017	0.000206	NR		0.1	2.06E-05
Benzo(k)Fluoranthene	0.044	0.000534	NR		0.1	5.34E-05
Benzo(a) Pyrene	0.089	0.001081	NR		1	0.001081
Benzo(a,h)anthracene	0.752	0.009131	NR		1	0.009131
Indeno(1,2,3-cd) pyrene	0.054	0.000656	NR		0.1	6.56E-05
Benzo (ghi) Perylene	0.097	0.001178	0.04	0.029446	0.01	1.18E-05

$\Sigma HQ = 0.082268$; $\Sigma CR = 0.010393$

Table 3.2.8. Human HRI from consumption of PAHs in Crab., Marine Base Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	4.71E-05	0.02	0.002357		
Acenaphthylene	0.01	4.71E-05	NR			
Acenaphthene	0.01	4.71E-05	NR			
Fluorene	0.01	4.71E-05	0.04	0.001179		
Anthracene	0.01	4.71E-05	0.04	0.001179		

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Phenanthrene	0.02	9.43E-05	0.04	0.002357		
Fluoranthene	0.01	4.71E-05	0.04	0.001179		
Pyrene	0.13	0.000613	NR			
Benz(a)anthracene	0.1	0.000471	NR		0.1	4.71E-05
Chrysene	0.08	0.000377	NR		0.01	3.77E-06
Benzo(b)Fluoranthene	0.05	0.000236	NR		0.1	2.36E-05
Benzo(k)Fluoranthene	0.04	0.000189	NR		0.1	1.89E-05
Benzo(a) Pyrene	0.07	0.00033	NR		1	0.00033
Benzo(a,h)anthracene	0.05	0.000236	NR		1	0.000236
Indeno(1,2,3-cd) pyrene	0.04	0.000189	NR		0.1	1.89E-05
Benzo (ghi) Perylene	1.91	0.009004	0.04	0.02143	0.01	9E-05

$\Sigma HQ = 0.233357$; $\Sigma CR = 0.000768$

Table 3. 2.9. Human HRI from consumption of PAHs in Oyster, Marine Base Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	0.000121	0.02	0.006071		
Acenaphthylene	0.01	0.000121	NR			
Acenaphthene	0.01	0.000121	NR			
Fluorene	0.04	0.000486	0.04	0.012143		
Anthracene	0.12	0.001457	0.04	0.036429		
Phenanthrene	0.18	0.002186	0.04	0.054643		
Fluoranthene	0.5	0.006071	0.04	0.151786		
Pyrene	0.21	0.00255	NR			
Benz(a)anthracene	0.06	0.000729	NR		0.1	7.29E-05
Chrysene	0.1	0.001214	NR		0.01	1.21E-05
Benzo(b)Fluoranthene	0.07	0.00085	NR		0.1	0.000085
Benzo(k)Fluoranthene	0.04	0.000486	NR		0.1	4.86E-05
Benzo(a) Pyrene	0.15	0.001821	NR		1	0.001821
Benzo(a,h)anthracene	0.5	0.007286	NR		1	0.007286
Indeno(1,2,3-cd) pyrene	0.037	0.000486	NR		0.1	4.86E-05

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Benzo (ghi) Perylene 0.03 0.000364 0.04 0.009107 0.01 3.64E-06
 $\Sigma HQ = 0.270179$; $\Sigma CR = 0.009378$

Table 3.2.10. Human HRI from consumption of PAHs in Prawn., Marine Base Creek

PAHs	Mean Conc. (mg/kg)	CDI	RfD	HQ	SF	CR
Naphthalene	0.01	4.71E-05	0.02	0.002357		
Acenaphthylene	0.01	4.71E-05	NR			
Acenaphthene	0.01	4.71E-05	NR			
Fluorene	0.01	4.71E-05	0.04	0.001179		
Anthracene	0.05	0.000236	0.04	0.005893		
Phenanthrene	0.06	0.000283	0.04	0.007071		
Fluoranthene	0.17	0.000801	0.04	0.020036		
Pyrene	0.35	0.00165	NR			
Benz(a)anthracene	0.16	0.000754	NR		0.1	7.54E-05
Chrysene	0.14	0.00066	NR		0.01	6.6E-06
Benzo(b)Fluoranthene	0.07	0.00033	NR		0.1	0.000033
Benzo(k)Fluoranthene	0.04	0.000189	NR		0.1	1.89E-05
Benzo(a) Pyrene	0.98	0.00462	NR		1	0.00462
Benzo(a,h)anthracene	0.47	0.002216	NR		1	0.002216
Indeno(1,2,3-cd) pyrene	0.08	0.000377	NR		0.1	3.77E-05
Benzo (ghi) Perylene	3.36	0.01584	0.04	0.396	0.01	0.000158

$\Sigma HQ = 0.432536$; $\Sigma CR = 0.007166$

From tables 3. 2.6, the total non-CR or HQ for consumption of PAHs in fish was estimated as above one. This implies that continuous consumption of fish from this creek directly or indirectly may cause non-carcinogenic health issues. The sum CR for PAHs in the fish of marine Base Creek was 0.008971 which is beyond the tolerable limit of 10^{-6} - 10^{-4} showing the possibility of cancer risk.

Meanwhile, as shown from table 3.2.7 to 3.2.10, the estimated hazard quotients of PAHs from Marine Base creek surface H₂O by consumption of periwinkle, crab, oyster, and prawn were **0.082268, 0.233357, 0.270179, and 0.432536 respectively**, which is lesser than one, implying that there is no possibility of non-carcinogenic health issues. However, the cancer risk values of periwinkle, oyster and prawn were above the permissible range depicting the tendency of cancer upon continuous consumption. The estimated values were **0.010393, 0.009378,**

and 0.007166 respectively. Cancer Risk for PAHs in crab is within the acceptable limit, there is thus no likelihood of cancer upon continuous consumption of prawn.

4.0 Conclusions

The concentrations PAHs in most of the aquatic biotics of both creeks were found to be of a risk upon consumption by man, which implies that the major sources of these pollutants within the creeks must be checked to forestall serious public health issues.

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