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Risk Assessment of Polyaromatic Hydrocarbon (PAH) in Asiko River, in Ajaokuta, Kogi State, Nigeria

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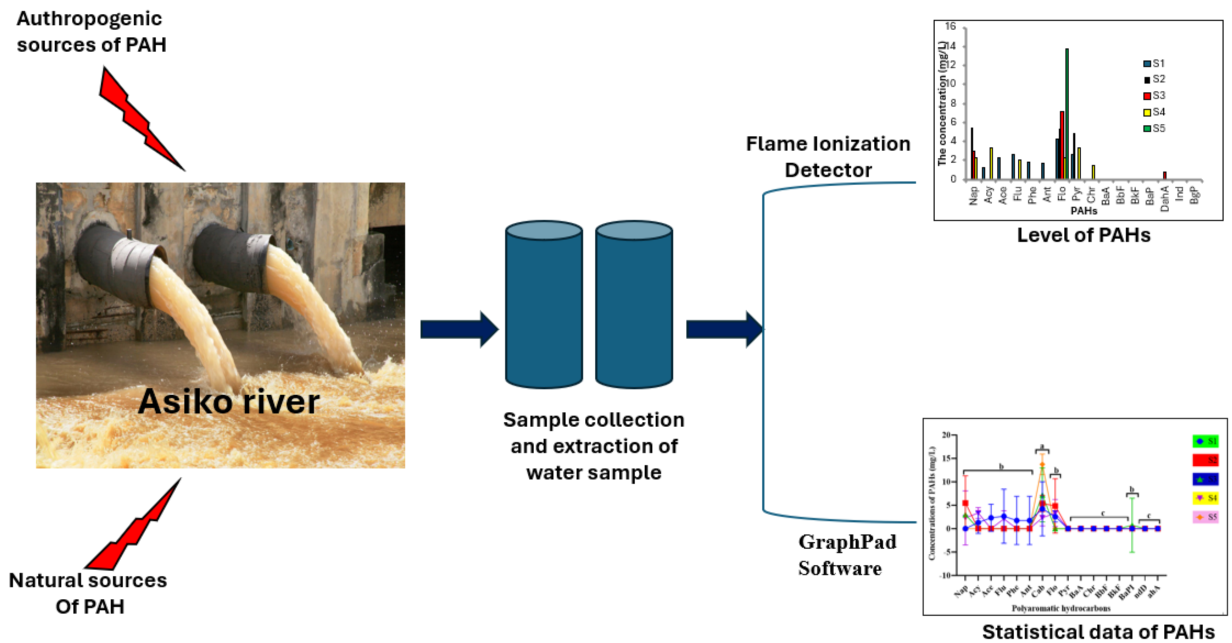
Abstract

Polyaromatic hydrocarbons (PAHs) are persistent environmental pollutants with significant health risks due to their toxicity, carcinogenic potential, and resistance to degradation. This study assessed PAH contamination in the Asiko River, situated in Ajaokuta, Kogi State, Nigeria, by analyzing water samples from five designated points (S_1-S_5) for hydrophobic PAH concentrations. The collected water samples were preserved in dark brown glass vials containing preservatives and transported to the laboratory. PAHs extracts were isolated through a liquid-liquid extraction method and then the extract was subjected. The results indicated varying PAH levels across sampling sites, with S_1 recording acenaphthylene ($1.28 \pm 2.3 \times 10^{-5}$ mg/L), acenaphthene ($2.30 \pm 2.9 \times 10^{-5}$ mg/L), and pyrene ($2.60 \pm 1.2 \times 10^{-5}$ mg/L), while S_2 and S_3 had notable detections of naphthalene ($5.445 \pm 5.7 \times 10^{-5}$ mg/L and $3.00 \pm 2.7 \times 10^{-5}$ mg/L, respectively). S_4 exhibited the highest diversity of PAHs, including naphthalene ($2.28 \pm 5.7 \times 10^{-6}$ mg/L) and pyrene ($3.32 \pm 2.9 \times 10^{-5}$ mg/L), whereas S_5, serving as the control, showed fluoranthene ($13.70 \pm 2.2 \times 10^{-5}$ mg/L). The concentrations exceeded the Agency for Toxic Substances and Disease Registry (ATSDR) permissible level of 0.2 μg/L and the US Environmental Protection Agency (USEPA) limit of 0.1 μg/L, indicating significant pollution. Risk assessment parameters, including average daily dose (ADD), hazard quotient (HQ), and lifetime average daily dose (LADD), were computed following USEPA methodologies. The ADD values for adults and children ranged from 0.39 to 0.56 and 1.20 to 2.29, respectively, while LADD values were below the threshold of 10^{-3}, suggesting no immediate health risks. The cancer risk ($<10^{-6}$) and HQ (<1) were within acceptable limits, indicating that despite contamination, local inhabitants are not currently at high risk of carcinogenic effects. These findings underscore the necessity of stringent regulatory measures to mitigate PAH pollution and protect aquatic ecosystems and human health. Continuous monitoring, pollution control strategies, and public awareness are recommended to reduce PAH exposure in the Asiko River region.

Keywords: Flame ionization detector, Polycyclic aromatic hydrocarbons; silica gel column; solvent extraction; Water pollution.

Graphical Abstract



Introduction

Pollution refers to harmful contaminants in the environment, including heat, light, sound, and other energy sources, which pose a threat to biological, ecological, and physical systems [1,2]. Environmental pollution, characterized by the increase in trace elements exceeding environmental tolerance limits, poses a threat to ecosystems, human health, and sparks conflict between industrial operations and the public [3, 4]. This phenomenon is commonly ascribed to human activities, such as industrial and agricultural practices [5]. Water pollution, a worldwide problem that endangers ecosystems and public health, is one of the most urgent environmental issues [6]. Polycyclic aromatic hydrocarbons (PAHs), dangerous organic chemicals mostly produced by human activities such as industrial discharges, vehicle emissions, and inappropriate waste disposal, are a significant class of contaminants of concern [1,7]. PAHs pose significant threats to aquatic ecosystems due to their toxicity, persistence, and bioaccumulation, leading to mutagenesis, endocrine disruption, and increased cancer risks in humans and animals [1,8].

The sources of PAHs can be broadly classified into three categories: biogenic, petrogenic, and pyrogenic [9]. Petrogenic PAHs are found in crude and refined petroleum products and are introduced into aquatic environments through routine tanker operations, urban runoff, and other pathways [10,11]. Pyrogenic PAHs, on the other hand, are produced when biomass, fossil fuels, and wood are burned, releasing these compounds as exhaust and solid residues. PAHs are organic pollutants with structures made up of multiple fused aromatic rings [1,12]. They have low volatility, low solubility in water, and significant chemical stability [13]. Anthropogenic sources usually outweigh natural ones in areas affected by human activity, except for certain compounds like perylene [14]. Once released into the environment, PAHs spread through various pathways and infiltrate living organisms. Human exposure occurs primarily through occupational contact, passive or active smoking, and the ingestion of contaminated food and water [1,15]. Additionally, natural sources contribute to PAH levels in the environment. For instance, perylene is thought to form during early diagenesis through the in-situ transformation of perylene quinone pigments or other organic materials [14,16]. After being discharged into the environment, PAHs spread via various routes and enter living things. Ingestion of tainted food and water, passive or active smoking, and occupational contact are the main ways humans are exposed [1,17]. This complex threat emphasises how urgently policies to reduce PAH pollution and protect the environment and public health are needed.

The ability of polycyclic aromatic hydrocarbons (PAHs) to elicit genotoxic and carcinogenic effects via drinking water is a considerable concern, especially in rivers tainted with these substances [18]. Research in the Nigerian Niger Delta has demonstrated a link between PAH contamination and widespread health problems. Owing to their hydrophobic characteristics and persistence, PAHs can bioaccumulate in adipose tissues and biomagnify through the food chain [19].

This bio-accumulative capacity, along with their mutagenicity, toxicity, and durability, has led to extensive investigations into their distribution in coastal ecosystems, where they are rigorously monitored [20]. Such contamination seriously impacts food security and human health, underscoring the urgent need for continuous PAH monitoring to ensure quality control and detect concentration variations before they exceed hazardous limits. PAHs are introduced into the environment via multiple pathways, including the decomposition of organic matter, petrogenic sources, and thermal degradation under hypoxic conditions. Pyrolysis, a process involving the exposure of organic materials to elevated temperatures with or without oxygen, significantly contributes to the formation of pyrogenic PAHs [21]. Pyrolytic processes entail the fragmentation of long-chain hydrocarbons into shorter chains, facilitated by heat or catalysts. Notable examples include the destructive conversion of coal into coke and coal tar, as well as the partial combustion of fuels in automobile engines, wood combustion, and fuel oil combustion in heating systems. These processes transpire at temperatures ranging from 350°C to 1200°C and are particularly prevalent in urban areas characterised by high anthropogenic activity.

PAH levels in the Asiko River are rising because of human activities such as agricultural runoff and the disposal of industrial waste. By determining their sources and comprehending the mechanisms underlying their persistence, this study aims to alleviate PAH pollution. The sustainable use of water resources, biodiversity preservation, and public health protection all depend on efficient management and control.

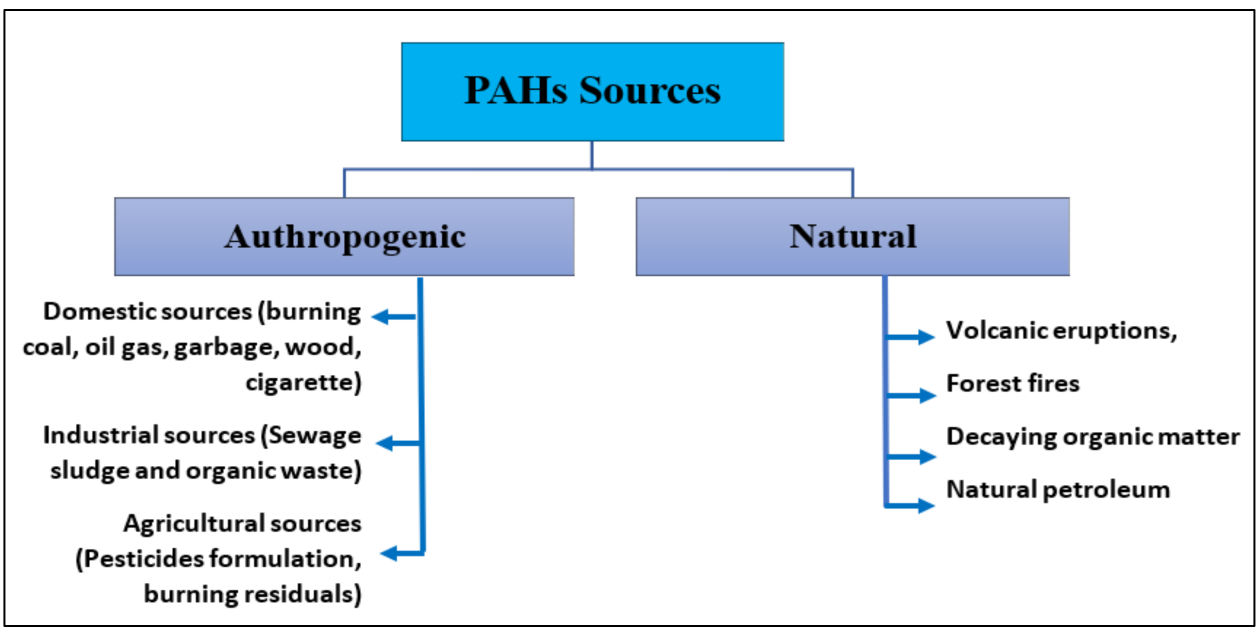


Figure 1: Mode of PAHs formation

In sediments, water sources, wastewater, and aquatic creatures like crustaceans, PAHs are often found in mixes and often co-occur with other contaminants [22]. Although incomplete fuel combustion is the main way that these compounds enter the environment, cooking, industrial waste, and agricultural fires can also release them into the atmosphere. The melting and boiling temperatures of PAHs grow with increasing molecular weight, but their water solubility falls [23]. Higher molecular weight PAHs, such as benzo[a]pyrene and chrysene, are essentially insoluble in water.

Numerous studies have documented the presence of PAHs in aquatic environments [1,3,22]. While individual PAHs vary in health effects, they are collectively classified as priority pollutants due to their significant health risks to humans and aquatic biota. Accordingly, these compounds demand serious attention. The following PAHs, identified by the Agency for Toxic Substances and Disease Registry, are of particular concern: acenaphthene, acenaphthylene, anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(e)pyrene, benzo(b)fluoranthene, benzo(ghi)perylene, benzo(j)fluoranthene, benzo(k)fluoranthene, chrysene, dibenzo(ah)anthracene, fluoranthene, fluorene, indeno(1,2,3-cd)pyrene, phenanthrene, and pyrene [24,25].

Materials and Methods

Materials

The materials used in this study include n-hexane, dichloromethane, anhydrous sodium sulphate (Na_2SO_4), silica gel (as an adsorbent), distilled water, and acetone. A standard mixture of 16 priority polyaromatic hydrocarbons (PAHs) at a concentration of 1000 μg/mL was prepared and serially diluted for calibration of the flame ionization detector (FID) coupled with gas chromatography (GC). o-Terphenyl was used as a surrogate standard. All chemicals were of analytical grade. Additional equipment included a gas chromatography flame ionization detector (GC-FID) and a rotary evaporator.

Description of Sampling Points

The Asiko River, situated in Ajaokuta, Kogi State, North Central Nigeria, served as the study area (Figure 2 and Table 1). Samples were collected from designated points along the river between 8:00 AM and 10:00 AM.

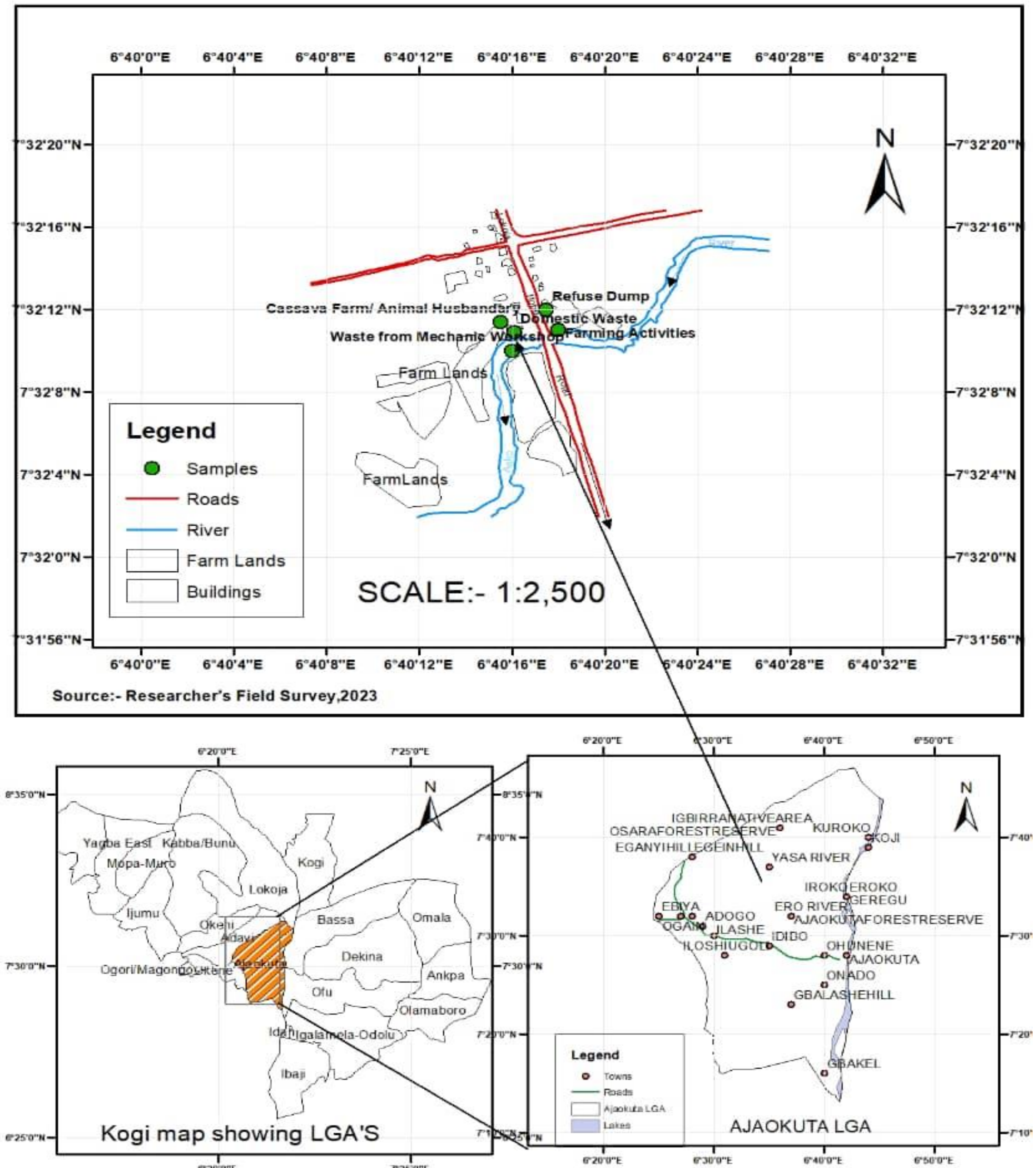


Figure 2: The map showing the area studied

Table 1: Co-ordinates of the studied site

Studied area	Longitude	Latitude	Description of sites
S_1	7° 32′ 11″ N	6° 40′ 18″ E	The wastes discharged from Banana, cassava and palm tree plantations.
S_2	7° 32′ 09″ N	6° 40′ 16″ E	Leached substances and wastes from dumpsite and food vendors.
S_3	7° 32′ 07″ N	6° 40′ 14″ E	Oil and grease, carbon particles and other wastes released from Mechanic workshop
S_4	7° 32′ 05″ N	6° 40′ 12″ E	Wastes discharged from domestic activities.
S_5	7° 32′ 04″ N	6° 40′ 10″ E	Little or no human activities (Pristine)

Collection, Preservation, and Extraction of Water Samples

Water samples were collected using 1-litre dark brown bottles. For preservation, 5 mL of diluted HCl (1:1) acid was added to each sample. The bottles were transported to the laboratory in ice-packed containers to maintain sample integrity. Approximately 500 mL of each sample was transferred into a separating funnel and extracted with 40 mL of dichloromethane. The extracts were concentrated to 5 mL using a rotary evaporator [25].

Clean-Up for PAHs

The extracts were cleaned using a glass column filled with 5 g of florisil and 2 g of anhydrous sodium sulfate (Na_2SO_4). The column was pre-eluted with 10 mL of dichloromethane before the extracts were eluted with 40 mL of the same solvent. Finally, the eluted extracts were concentrated to 2 mL using a rotary evaporator set at 37°C.

Quality Assurance

All glassware was thoroughly washed with detergent, rinsed with distilled water, followed by acetone, and oven-dried at 100°C [26]. For method validation, laboratory blank analyses were conducted. Samples spiked with the PAH standard mixture and surrogate were processed in the same manner as double-distilled water. The percentage recovery for each analyte was calculated to ensure the reliability and accuracy of the results.

$$\% \, Recovery = \frac{\text{Experimental value} \times 100}{\text{Experimental value}} \quad \text{equation (1)}$$

Instrumentation

The analysis was performed using a GC 6890 model equipped with an HP-5 column (30 m × 0.25 mm × 0.25 μm) and a flame ionization detector (FID) operated in splitless mode. Hydrogen gas at a flow rate of 35.0 mL/min was used as the carrier gas. A 1 μL aliquot of the sample extract was injected into the system at an injector temperature of 250°C.

The oven temperature was initially set at 50°C, ramped at 25°C/min, and increased to a final temperature of 310°C. The operational runtime was 20.40 minutes. To calibrate the instrument, a standard mixture of 16 PAHs was prepared at varying concentrations (10–600 μg/L). Additionally, the retention times of each analyte, as shown in Figure 3, were matched with those of the standards to ensure accurate identification and quantification.

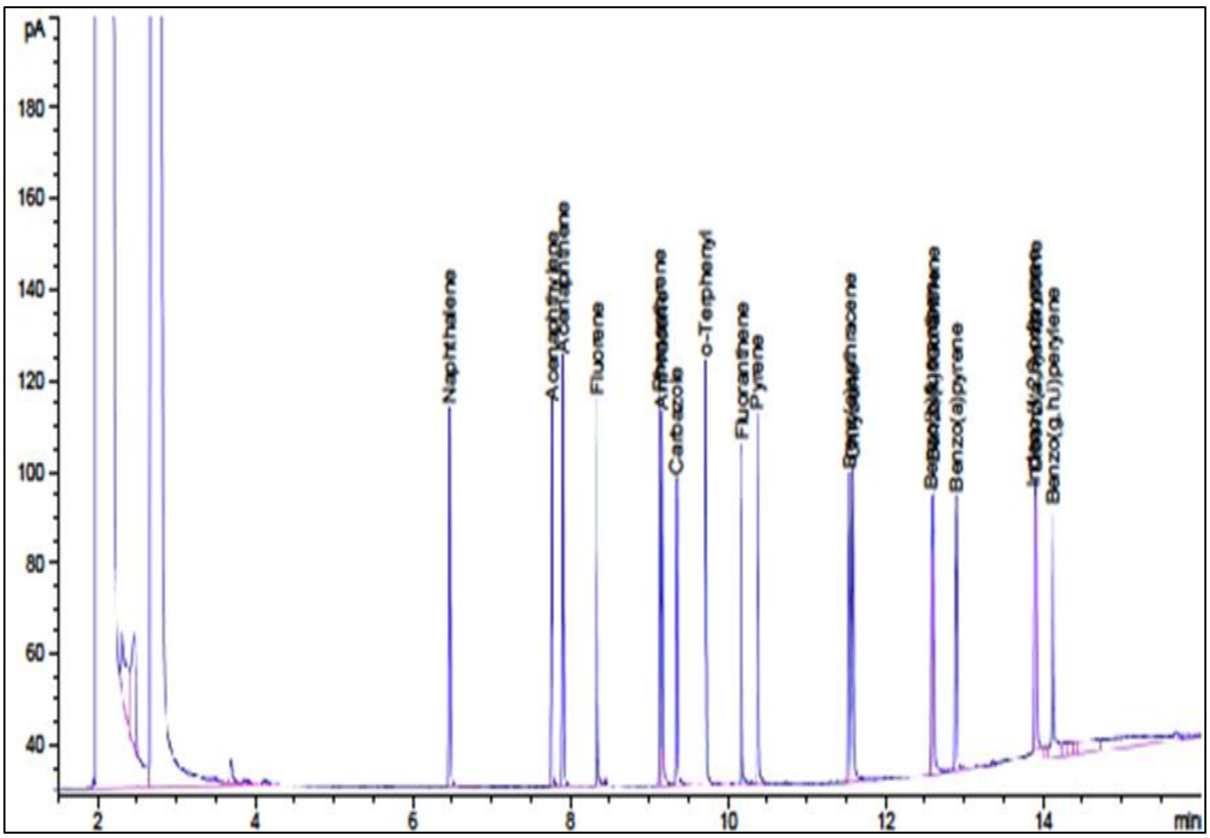


Figure 3: The Standard mixture of PAHs chromatogram

Statistical Analysis:

The GraphPad software was used for data analysis of variance, mean and standard deviation (SD) were obtained and correlation at a significant level of $p < 0.05$.

Risk Assessments

The health implications of PAHs in water were assessed for effects using average daily dose (ADD) in mg/kg/day, hazard quotient (HQ) and cancer risk (CR) in equations 2, 3, 4 as well as 5 respectively [27,28,29,30].

$$ADD = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (2)$$

$$HQ = \frac{ADD}{RfD} \quad (3)$$

$$LADD = \frac{C \times FI \times IR \times EF \times ED}{BW \times AT} \quad (4)$$

$$CR = ADD \times CSF \quad (5)$$

Exposure and Risk Assessment Parameters

- **C**: Level of PAHs in the sample (mg/kg)
- **IR**: Ingestion rate (24.7 g/day)
- **EF**: Frequency of exposure (350 days/year)
- **ED**: Time of exposure (30 years for adults)
- **BW**: Average body weight (60 kg for adults)
- **AT**: Average time (54.5 × 365 days)

Reference Doses (RfD):

- Ace: 60 µg/g
- Phe: 60 µg/g
- Flu: 40 µg/g
- Pyr: 30 µg/g [28,29,30].

Cancer Slope Factors (CSF):

- Bbf: 7.3×10^{-1}
- Bkf: 7.3×10^{-2}
- Bap: 7.3 [31]

Results and Discussion

The concentrations (mg/L) of PAHs, as shown in Table 2 and Figure 4, ranged from ND (not detected) to 1.80 ± 5.8 E-06 in sample S_1. In S_2 and S_3, three analytes were detected, while seven analytes were recorded in S_4. Only one analyte was detected in S_5, which served as a control. Several analytes, including naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, and pyrene, were recorded in S_1. A few, such as naphthalene, anthracene, and pyrene, were detected in S_2. This may be attributed to accidental oil discharges and waste releases from dye, pigment, and ceramic industries, with potentially poisonous waste percolating from a refuse site into S_1 and S_2 [32,33,34].

Many of the analytes were not detected in S_3, and those recorded were at low concentrations, which may be due to reduced anthropogenic activities in this environment. Additionally, some pollutants may have originated from non-point sources due to long-range atmospheric transport of persistent organic pollutants [19,35,36,37]. In S_4, naphthalene, fluorene, acenaphthylene, fluoranthene, pyrene, and chrysene were detected, while other analytes were below the detection limit. This could be linked to oil spills and industrial or residential discharges, highlighting the persistence of these compounds in the environment. At S_5, only one analyte was detected, with the others below the detection limit, likely due to the location being an upstream area with fewer anthropogenic activities and used as a control site. Most analytes were below the detection limit, with low concentrations in some places, while higher concentrations were recorded at certain sampling points. These findings align with the observations of [38], but contrast with the results of [39,40,41].

Table 2: The level of PAHs mg/L in Asoko river Values are means ± SD; (N = 3)

PAHs	**S_1**	**S_2**	**S_3**	**S_4**	**S_5**
Nap	**BDL**	5.45 ± 5.7 E-05	3.00 ± 2.7 E-05	2.28 ± 5.7E-06	**BDL**
Acy	1.28 ±2.3 E-05	**BDL**	**BDL**	3.33 ± 1.2 E-05	**BDL**
Ace	2.30 ±2.9 E-05	**BDL**	**BDL**	**BDL**	**BDL**
Flu	2.63 ±5.8 E-06	**BDL**	**BDL**	2.065 ± 1.7 E-05	**BDL**
Phe	1.80 ± 5.8 E-06	**BDL**	**BDL**	**BDL**	**BDL**
Ant	1.73 ± 5.1 E-4	**BDL**	**BDL**	**BDL**	**BDL**
Flo	4.20 ± 5.8 E-06	5.35 ± 1.73 E-05	7.19 ± 5.8 E-06	2.30 ± 1.7 E-05	13.70 ± 2.2 E-05
Pyr	2.60 ± 1.2 E-05	4.90 ± 5.8 E-06	**BDL**	3.32 ± 2.9 E-05	**BDL**
Chr	**BDL**	**BDL**	**BDL**	1.45 ± 1.7 E-05	**BDL**
BaA	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
BbF	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
BkF	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
BaP	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
DahA	**BDL**	**BDL**	0.72 ± 5.8 E-06	**BDL**	**BDL**
Ind	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
BgP	**BDL**	**BDL**	**BDL**	**BDL**	**BDL**
∑PAHs	**16.50 ± 5.9 E-04**	**15.60 ± 8.1 E-05**	**10.91 ± 4.04 E-05**	**14.74 ± 9.8 E-05**	**13.72 ± 2.2 E-05**

BDL: Below detection limit; S_1-S_5: Sampling points; S_5 (control)

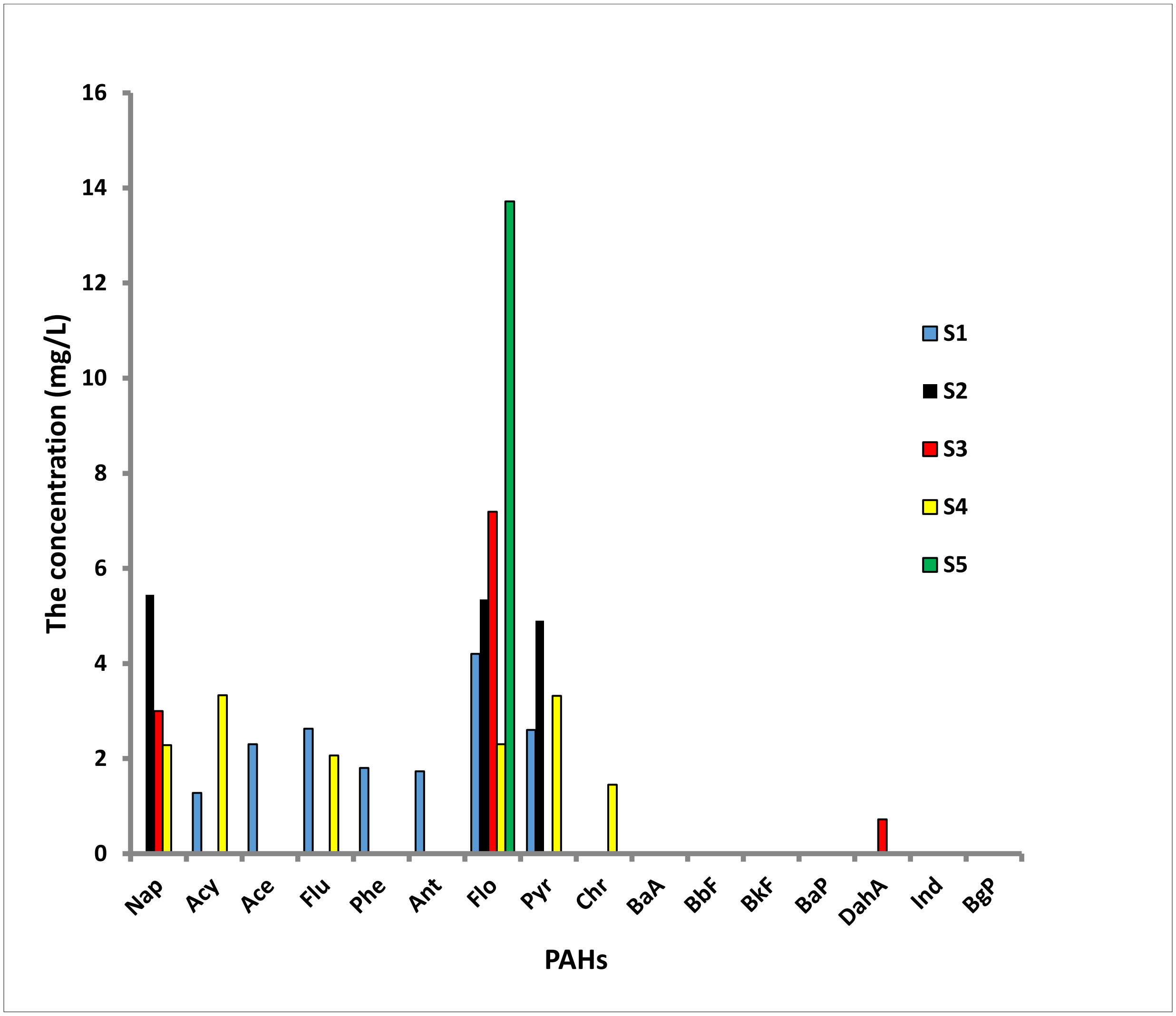


Figure. 4: The level of PAHs at five studied points across Asiko River

Figure 5 presents the data as mean ± standard deviation, derived from the meaning of three replicates (n = 3). A one-way analysis of variance (ANOVA) was conducted, followed by a multiple comparison test, with a significance level set at $p < 0.05$. The letters **a**, **b**, and **c** in the figure indicate statistically significant differences between the columns, where **a** represents the highest detected values, and **b** and **c** correspond to decreasing concentrations, observed at different sample points (S_1-S_5), respectively.

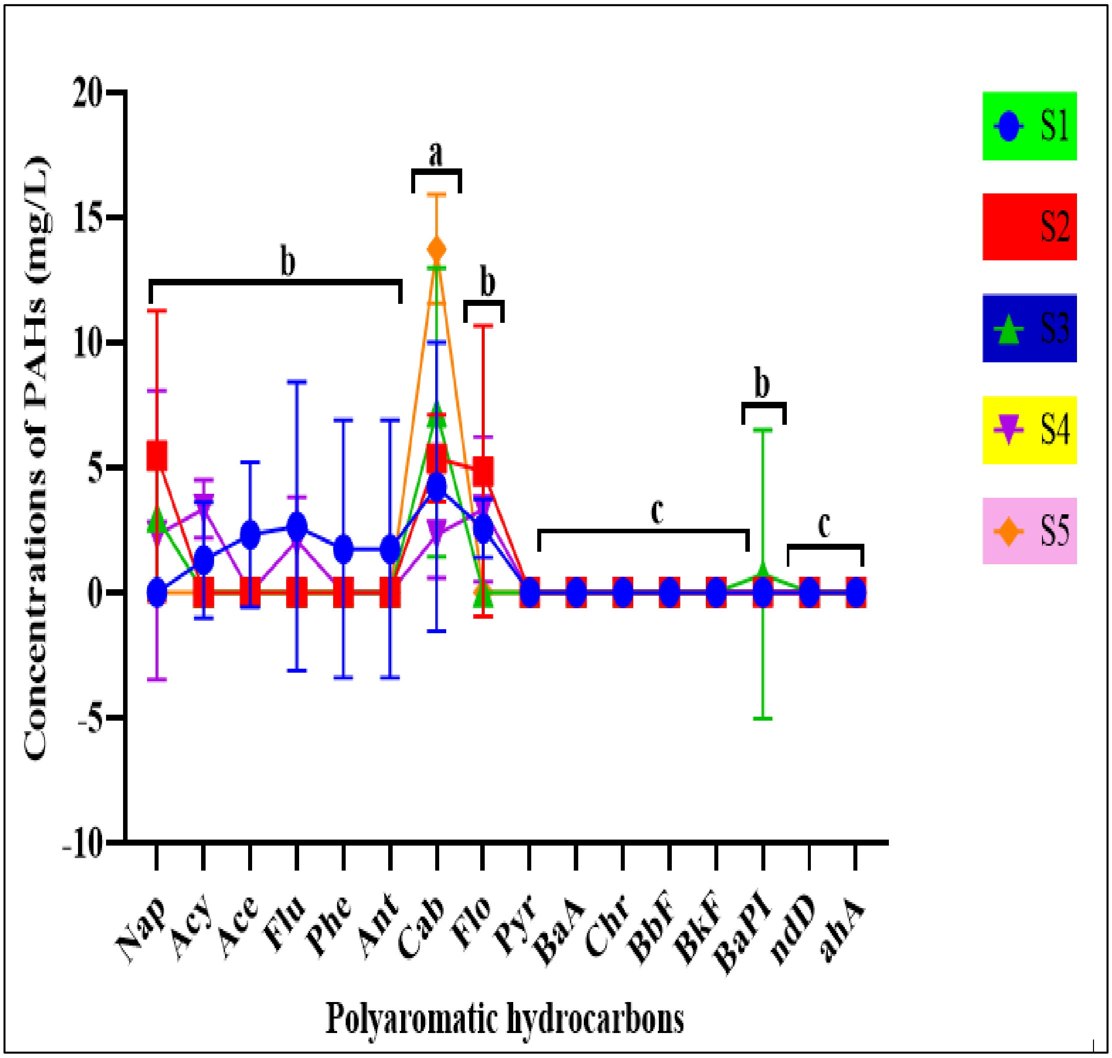


Figure 5: Statistical analysis results showing the level of PAHs recorded at different sample points.

The ADD (Figure 6 and 7) values for adults and children ranged from 0.39 to 0.56 and 1.20 to 2.29 in S_1, respectively. In S_2, the ADD ranged from below detection limit (BDL) to 1.06 for adults and 4.26 for children. In S_4, the ADD ranged from BDL to 0.72 for adults and 2.89 for children. However, in S_3, the ADD was below detection limit (BDL) for both adults and children.

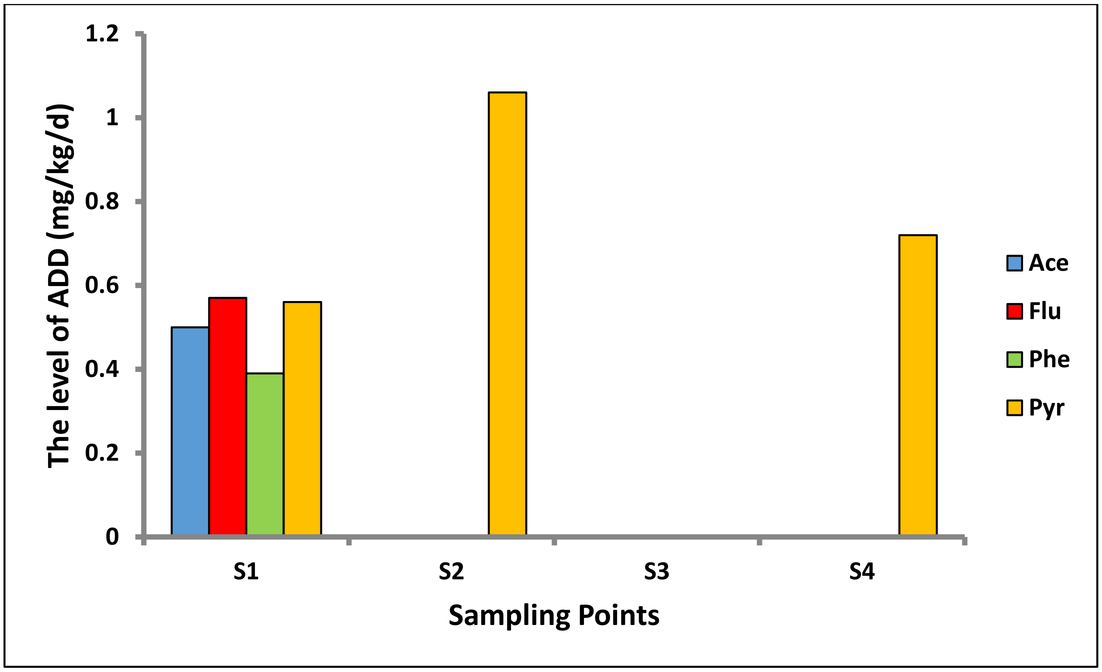


Figure 6: The ADD in adult at different sampling points

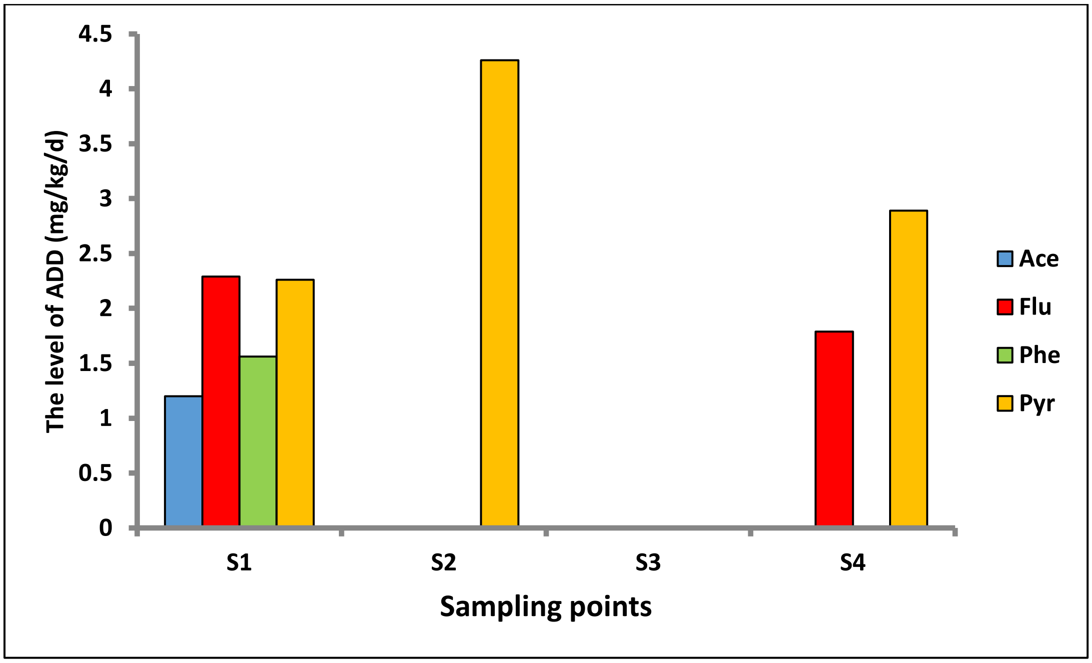


Figure 7: The ADD in children at different sampling points

Risk Assessment

Risk is the likelihood that organs (receptors) such as the kidney, lungs, and skin, among others, may develop cancer based on the exposure time to the toxicant and the degree of toxicity of the pollutants. Besides exposure to dangerous chemicals and hazardous substances, habits, age, and family history of inherited cancer could increase the individual's risk of having cancer [13]. The level of risk can be estimated by comparing the calculated values with USEPA cancer risk values [34,42]. However, there is a possibility that the dwellers of this river are probably exposed to the contaminants in water via inhalation or ingestion and dermal contact. ADD is the rate of consumption of pollutants per day. The ADD were not detectable in some sampling points while the ones recorded were still below the permissible limit of $10^{-4.}$ Also, if ADD> 10^{-4}. it implies possibility of life cancer risk [34,42].

The HQ (Figure 8 and 9) in adult and children were from 0.01- 0.02 and 0.02 – 0.07; ND – 0.03 and 0.14; BDL - 0.70 and 0.10 in S_1, S_2 and S_4 corresponding but below detection limit (BDL) in S_3. The hazard Quotient is the level at which living organisms are exposed to dangerous chemicals when there is no health effect (zero effect) on the living organism [42]. Furthermore, when the value > 1, it means harmful effect but < 1 implies low risk [23,42]. However, the HQ observed in all the samplings points less than 1, hence, there is no health risk.

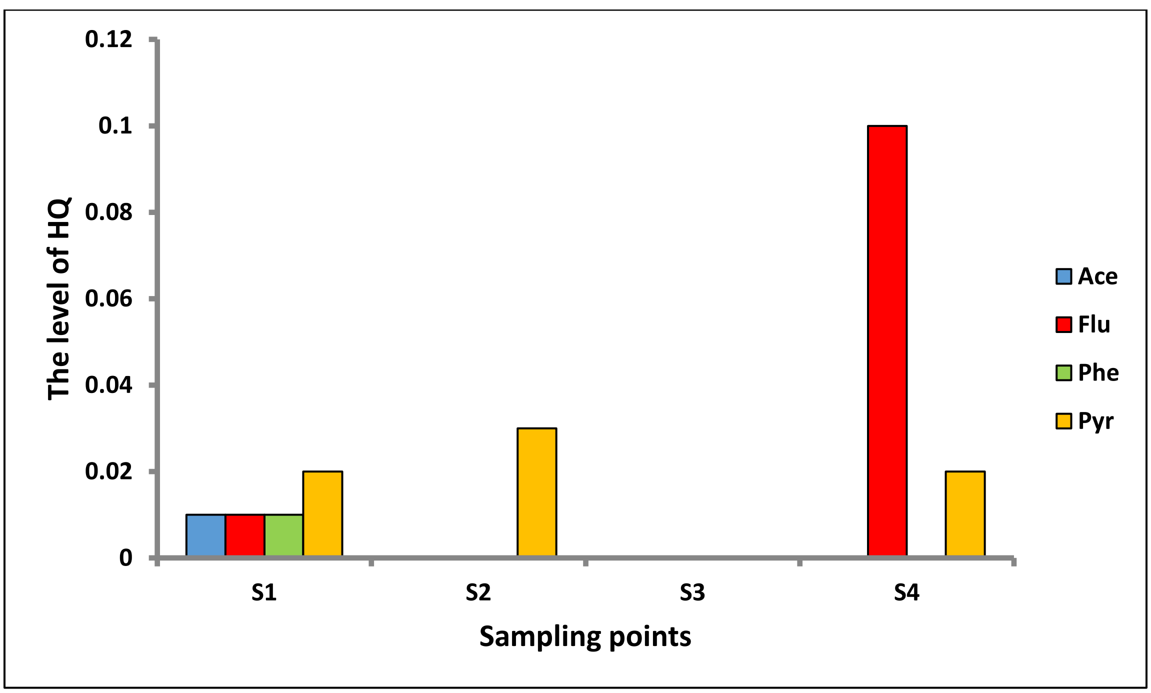


Figure 8: The HQ in adult at different sampling points

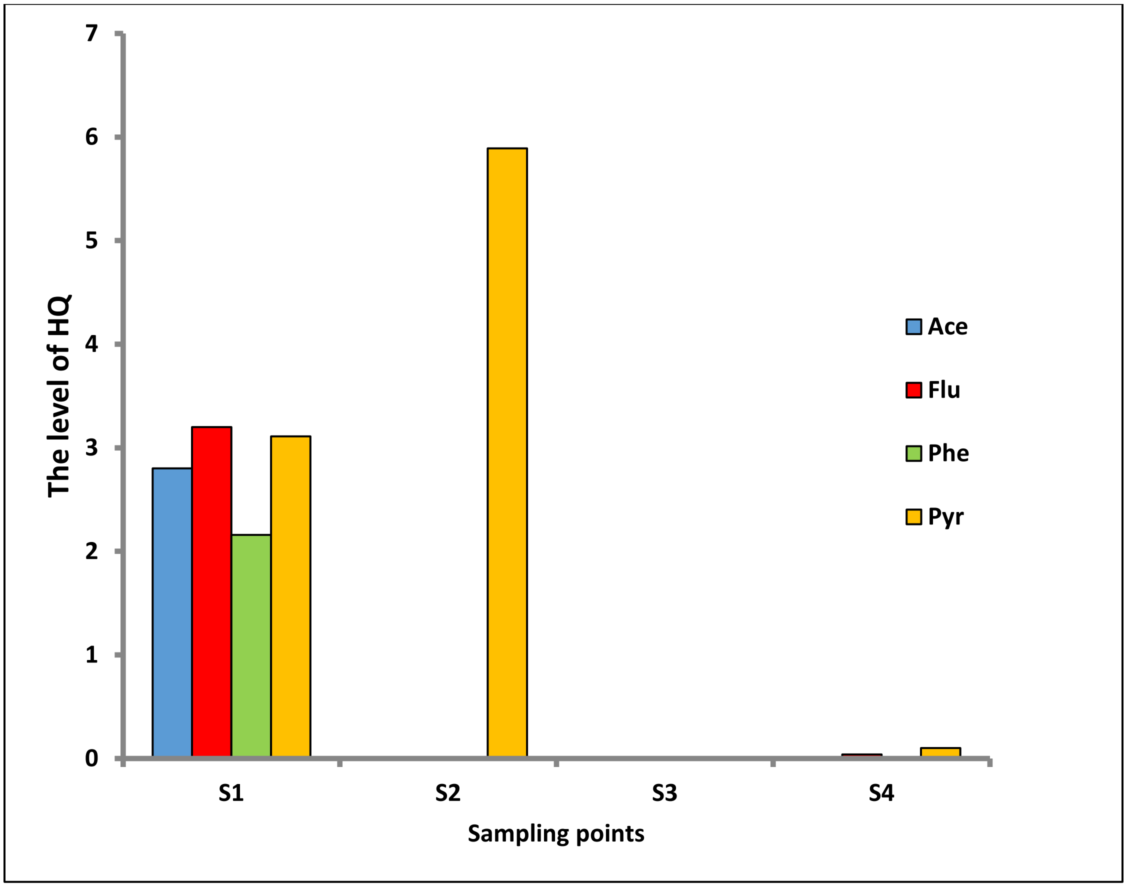


Figure 9: The HQ in children at different sampling points

The LADD (Figure 10 and 11) values for adults and children ranged from 0.29 to 0.44 and 2.16 to 3.11 in S_1, respectively. In S_2, the LADD ranged from below detection limit (BDL) to 0.80 for adults and 5.89 for children. In S_4, the LADD ranged from BDL to 0.70 for adults and 4.04 for children. However, LADD was not detected in S_3 for either adults or children. LADD represents the ingestion of organic pollutants in water over a lifetime. If LADD exceeds 10^{-3}, it indicates the need for protective measures [33,1,43]. Since the values are below 10^{-3}, no protective measures are required at any of the sampling points along the river.

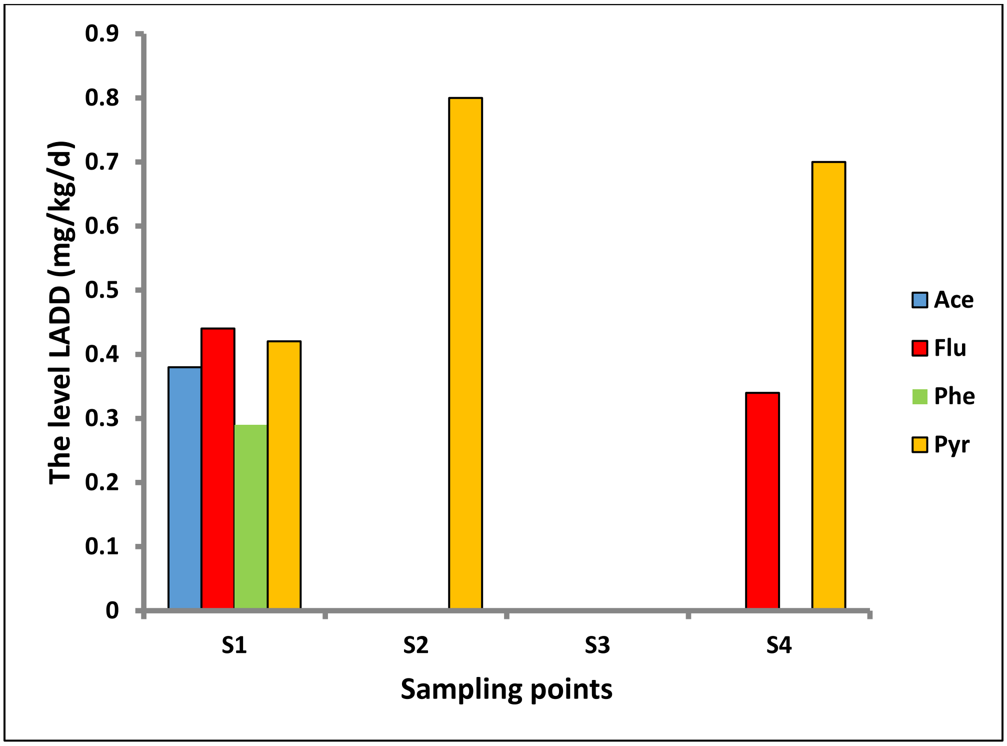


Figure 10: The LADD in adult at different sampling points

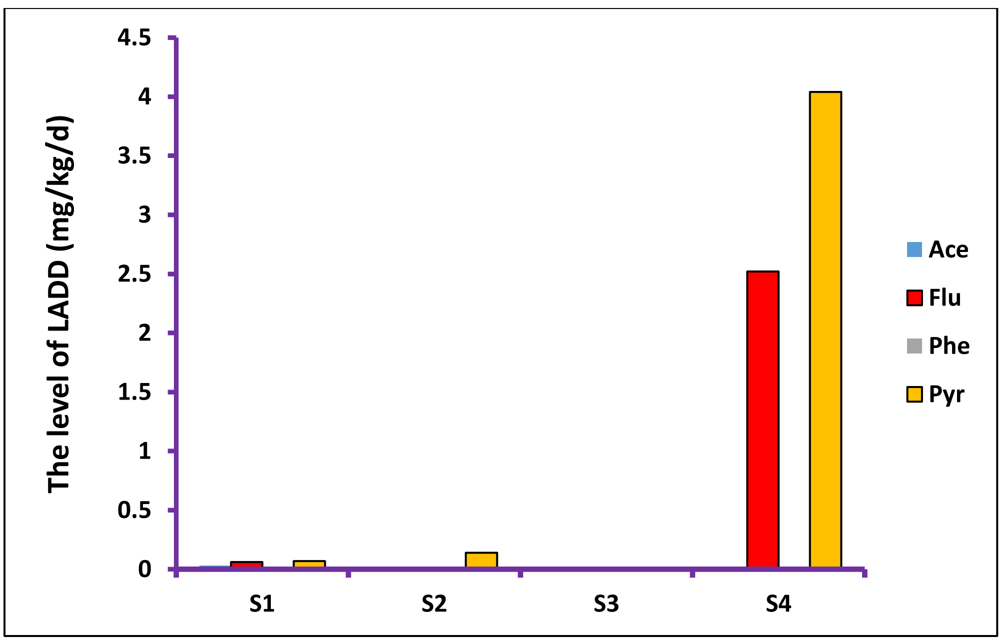


Figure 11: The LADD in children at different sampling points

The LADD (Figure 10 and 11) values for adults and children ranged from 0.29 to 0.44 and 2.16 to 3.11 in S_1, respectively. In S_2, the LADD ranged from below detection limit (BDL) to 0.80 for adults and 5.89 for children. In S_4, the LADD ranged from BDL to 0.70 for adults and 4.04 for children. However, LADD was not detected in S_3 for either adults or children. LADD represents the ingestion of organic pollutants in water over a lifetime. If LADD exceeds 10^{-3}, it indicates the need for protective measures [27,35,36]. Since the values are below 10^{-3}, no protective measures are required at any of the sampling points along the river.

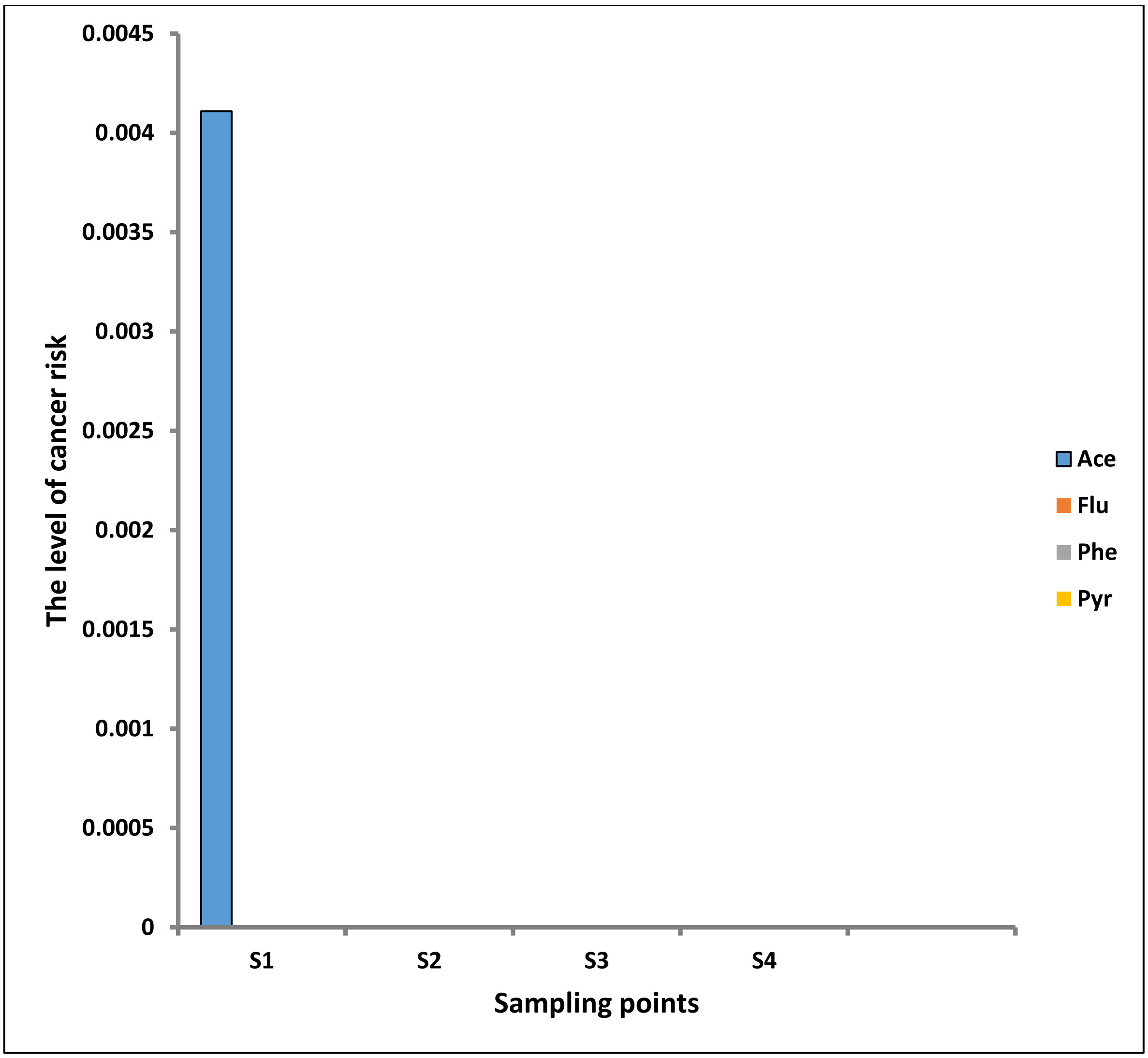


Figure 12: The cancer risk in adults at different sampling points

Conclusion

Samples were collected along the Asiko River and extracted using an organic solvent, with the USEPA 17 PAHs priority pollutants determined using GC-FID. A few of these analytes were detected, while others were below detection limits at certain sampling points. This variation could be attributed to anthropogenic activities, such as improper handling of hazardous chemicals and the release of domestic and industrial waste. Furthermore, the concentration of PAHs in the river water exceeded the USEPA's permissible limit of 0.1 µg/L, posing a potential threat to aquatic life and humans. However, the lower values of ADD, HQ, LADD, and cancer risk suggest that the contaminants may not pose an immediate health threat to humans or a significant biological risk to aquatic life. This study reveals elevated concentrations of PAHs in water samples from selected sites, surpassing the permissible limits set by both the USEPA and ATSDR. These findings confirm the presence of hazardous PAHs, posing risks to human health and aquatic ecosystems. Although health risk assessments, including LADD, HQ, ADD, and cancer risk, indicate that the cancer risk for residents remains within safe limits, long-term exposure to these pollutants still represents a significant threat. Therefore, proactive measures and continuous monitoring of industrial and human activities around these water sources are essential to reduce PAH contamination, safeguard public health, and maintain environmental integrity.

Author Contributions: Conceptualization, Methodology, A.E.E.A, Y.A, A.G, S.S.A

Writing-original draft preparation, A.E.E.A, Y.A, and H.A.M ; visualization,

A.E.E.A, Y.A, S.S.A., A.H.B., and A.N.A.; investigation, A.E.E.A, Y.A, and

O.K.D.; Sample pretreatment and characterizations, A.E.E.A, Y.A, S.S.A., O.K.D., O.V.F and

A.N.A.; Results interpretation, A.E.E.A, Y.A, B.O.M., and S.S.A.; writing reviewing and

editing, A.E.E.A, Y.A and A.G supervision and funding acquisition, Y.A.

All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: Authors declare no conflicts of interest.

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