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Spatiotemporal Variation and Health Risk Assessment of Selected Polycyclic Aromatic Hydrocarbons and Pesticides in Ogun River, Lagos, Nigeria

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ABSTRACT: Studying the spatiotemporal distribution and health risks associated with polycyclic aromatic hydrocarbons and pesticides in a river is crucial for understanding the impacts of these contaminants on human health. Hence, the objective of this paper was to investigate the spatiotemporal variation and health risk assessment of selected polycyclic aromatic hydrocarbons and pesticides in Ogun River, Lagos, Nigeria. The water samples were analyzed using gas chromatography coupled with MSD model 5977A. The results show a diverse distribution of PAHs across the sites, with Naphthalene and Acenaphthylene being the most frequently detected, having significantly higher concentrations in the dry season than in the rainy season. There was a significant difference in the mean amounts of pesticides at site 1 compared to other sites. While some pesticides recorded higher mean values in the rainy season, others had higher mean values in the dry season. Similar to PAHs, most pesticides had higher mean values in year 1 than in year 2. Most of the pesticides and PAHs posed significant non-cancer risks (HQ > 1) through oral exposure, except Glyphosate and Imidacloprid at all the sites; and Acenaphthene and Anthracene at site 4. There was also a significant health risk through dermal exposure to Heptachlor and Dichlorvos at S4 and Aldrin at S1. The cancer risk through oral exposure to all the detected carcinogenic pollutants is significant (CR $>1\times10^{-4}$), except Lindane at S4, which was within the acceptable range. Benzo[a]anthracene and Benzo[a]pyrene posed significant cancer risk through dermal exposure at S3 and heptachlor at S1. The study indicated that the observed spatial and temporal variations are different for specific pollutants. There is a high probability of adverse carcinogenic and non-carcinogenic risks for people who rely on the river for various purposes.

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Water is a natural resource that plays a critical role in the sustenance of life for all living organisms. However, due to population growth, industrialization, agriculture, and other human activities, this vital resource is increasingly polluted and has become a global concern requiring urgent attention. Top among the pollutants of concern are organic pollutants, including pesticides and polycyclic aromatic hydrocarbons (PAHs). Some features of these organic pollutants are their persistence, bioaccumulation in the food chain, and the ability to cause various environmental and health problems (Oyugi *et al.*, 2021). PAHs are a group of organic compounds containing two or more fused aromatic rings formed during incomplete burning of organic substances like coal, oil, wood, animal parts, etc. (ATSDR, 1995).

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They generally have low solubility in water, which further decreases with an increase in molecular weight, and are highly lipophilic, which partly accounts for their toxicity (Essumang, 2020). PAHs have been ranked 9th on the ATSDR priority list of hazardous substances (ATSDR, 1995). Based on their toxicity, potential for human exposure, frequency of exposure, and extent of information available, the USEPA has listed 16 PAHs as priority pollutants (Keith and Telliard, 1979). Eight of them (mostly the higher molecular weight ones) are carcinogenic, while others have varying degrees of acute and chronic toxicities (ATSDR, 2009). PAHs enter water bodies through different routes, like atmospheric deposition, runoff, domestic and industrial waste, etc. (Poh and Kostecki, 2020). The application of agrochemicals like pesticides has resulted in a large increase in food production and a significant reduction in some diseases globally (Pelegrini and Fernandez, 2018). However, this has come with *consequences*. An estimated 0.3% of the applied pesticides go to the target pest, while 99.7% end up in the environment, with a wide range of environmental and health implications (Pimentel, 2003). Many are known carcinogens, while some are endocrine disruptors (Bergman *et al*., 2013), cause liver damage, nervous and reproductive disorders, immune suppression, and allergies (Holmgaard and Nielsen, 2009; Terziev and Petkova-Georgieva, 2019). Chronic exposure to pesticides has also been linked to neuropsychiatric disorders that may lead to suicide attempts (Faria *et al*., 2014). They also lead to a loss of biodiversity (Bourguet and Guillemaud, 2016). Water pollution is a significant problem in Lagos, Nigeria, resulting from the discharge of untreated industrial and agricultural wastewater and animal faeces into water bodies. The popular Kara Abattoir, which generates abattoir-based pollutants such as animal blood, paunch, manure, animal faeces, animal horns, bones, and spent oil from machines like generators, is located in the study area. This study aims to assess the spatiotemporal variation and health risk of selected polycyclic aromatic hydrocarbons and pesticides in Ogun River, Lagos, Nigeria.

MATERIALS AND METHODS

Study Area: The study area was Berger Princict, one of the tributaries of Ogun River, a bustling commercial area on the outskirts of Lagos, Nigeria (Figure 1). The Lagos Kara market, which housed Kara Abattoir, was also located in the area. The Ogun River basin is in southwestern Nigeria, with latitudes 6°26’N and 9°10’N and longitudes 2°28’E and 4°8’E. The land area was about 23,000 km^2. Ogun River flows southwards over about 480 km before discharging into the Lagos Lagoon. The two seasons in the study area were the wet season between April and October and the dry season from November to March.

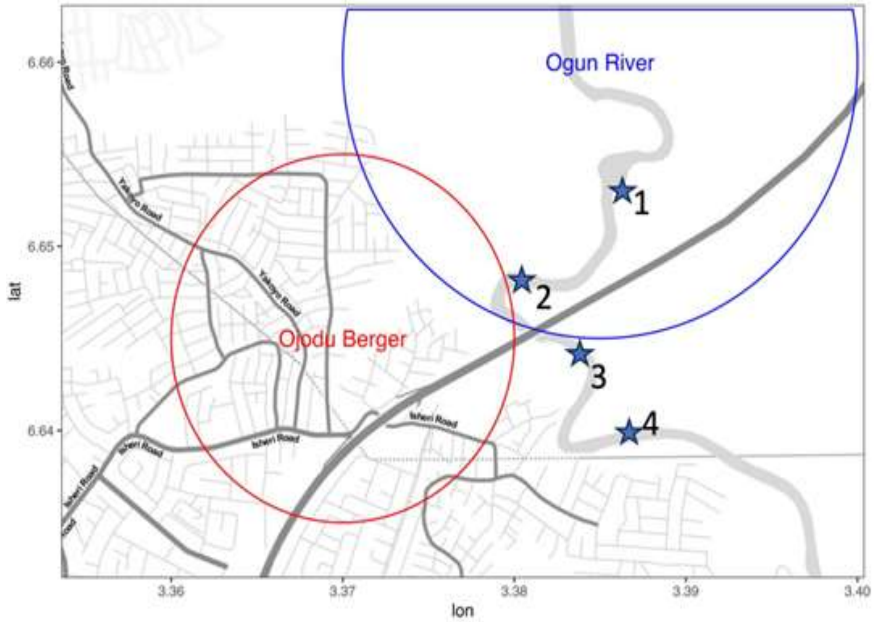

Fig 1: Map of the study area showing the sampling sites

The mean annual rainfall ranged from 900 mm in the North to 2000 mm towards the South. The vegetation zone identified here was of two major types: the high forest vegetation in the North and Central part and the swamp/mangrove forests on the Southern coast and floodplains next to the lagoon. The residents around the river's numerous flow points use the untreated water for domestic purposes (including drinking), irrigation, fishing, car wash, transportation, and recreation, among other purposes.

Sampling Procedure: Sampling plastic bottles used were washed and thoroughly rinsed with the river water. At each sampling time, nine grab water samples were taken from four locations or points across the river (S1, S2, S3, and S4) below the surface (25 cm) using the grab sampling technique. Samples were collected twice a year (February and August), corresponding to the peak of the dry season and the peak of the rainy (wet) season, over two years, resulting in 144 samples. The samples, put in a mobile cooler with ice packs maintained at ≤ 4°C, were immediately transported to the laboratory (Analytical Department, National Research Institute for Chemical Technology (NARICT) Zaria) for analysis within 48 hours.

Chromatographic Analysis: A 7890A (Agilent Technologies, USA) gas chromatography, coupled with MSD model 5977A, was used to analyze samples. Chromatographic separations were carried out using the HP-5 MS Ultra Inert column (30 m × 0.25 mm I.D.× 0.25-µm; Agilent Technologies, USA). Analyses were conducted in the SCAN mode. The following analysis parameters were used: samples injected in a splitless mode, injected volume—2 µL, carrier gas—helium (5.0 purity, flow 2.1 mL/min), and the MS ionization was carried out in the electron ionization mode at 70 eV. For pesticide residues, the temperature was 270 °C for the transfer line, 250 °C for the ion source, 150 °C for the quadrupoles, and 70–270 °C for the oven. For PAHs analysis, the temperature was 320 °C for the transfer line, 320 °C for the ion source, 150 °C for the quadrupoles, and 80–320 °C for the oven. Software Mass Hunter, version B.07.06, was used for data acquisition, control, and data processing of the analysis results. The total run time was 26 min for pesticide and PAHs.

The limit of detection (LOD) was used to determine the reliability of any low values obtained from the equipment. It is the smallest quantity of analyte that can be reliably detected. It is measured from a series of blank runs or standard curves.

$$LOD = \frac{3.3\sigma}{S} \quad (1)$$

Where σ = standard deviation at the y-intercept; *s* = standard error; 3.3 = a constant.
The limit of quantification (LOQ) is the smallest quantity that can be reliably used to quantify an analyte. The LOQ is generally taken as 3 x LOD. Values below the LOD and/or LOQ (as applicable) are regarded as unreliable and discarded from the discussion.

Exposure assessment: Exposure assessment is the extent of human exposure to the pollutants or the environmental agent (IPCS, 2010). To assess the exposure (to pollutants) in adults, the chronic daily intake (CDI) ((mg/kg/day)), which represents the lifetime average daily dose of exposure to contaminants (polycyclic aromatic hydrocarbons and pesticides), was used. The chronic daily intake via oral ingestion (CDI) and dermal route (DA_d) were calculated for the adult population using the equations 2 and 3 (USEPA, 1989; USEPA, 2004):

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

$$DA_d = \frac{C_p \times K_d \times SA \times EF \times ED \times ET \times CF}{BW \times AT} \quad (3)$$

Where: C_p stands for the concentration of the pollutants in the water sample (mg/L); IR is the injection rate per unit time (2 L/day); EF is the exposure frequency (350 days/year for both ingestion and dermal absorption); ED is the exposure duration (30 years); BW represents the average body weight (70 kg); AT means the average time (non-carcinogenic effects AT = ED x 365 d/yr; carcinogenic effects AT = 70 yr x 365 d/yr); SA stands for the exposed skin area (18,000 cm^2); K_p (cm/h) is the dermal permeability coefficient (contaminant-specific: Table 1); ET is the exposure time (0.58 h/day) and CF represents the unit conversion factor (L/1000 cm).

Non-carcinogenic Health Risk Assessment: Risk is the probability of adverse effects from exposure to an environmental agent or mixture of agents (IPCS, 2010). The non-carcinogenic risks through oral and dermal routes were estimated using the hazard quotient (HQ) and hazard index (HI) as shown in the Equations 4 and 5.

$$HQ_o = \frac{CDI}{RfD_o} \quad (4)$$

$$HQ_d = \frac{DA_d}{RfD_d} \quad (5)$$

The oral dose-response factor, adjusted for absorption, can be converted to an absorbed dose basis as follows (USEPA, 1992):

$$RfD_d = RfD_o \times ABS_{GI} \quad (6)$$

DA_d = Dermal Absorbed Dose (mg/kg-day); RfD_d = Absorbed reference dose (mg/kg-day)

RfD_o = Oral reference dose (mg/kg-day). (Reference dose is the daily exposure to a substance that will not result in any deleterious effect in a lifetime for a given human population) (FAO/WHO, 2013). ABS_{GI} = Fraction of contaminant absorbed in the gastrointestinal tract.

If HQ is less than 1, it implies no significant risk; if HQ is equal to or greater than 1, it implies a significant non-cancer risk.

$$HI = \sum HQ \quad (7)$$

The value of the hazard index is proportional to the magnitude of the toxicity of the water to the population.

Carcinogenic Risk Assessment: Carcinogenic risk assessment estimates the probability of an individual developing cancer over a lifetime due to exposure to the potential carcinogen. Oral Carcinogenic Risk (CR_o) due to oral ingestion and carcinogenic risk due to dermal absorption (CR_d), were calculated using the Equations 8, 9 and 10 (USEPA, 2004):

$$CR_o = CSF_o \times CDI \quad (8)$$

$$CR_d = DA_d \times CSF_d \quad (9)$$

$$CSF_d = \frac{\mathrm{CSF_o}}{\mathrm{ABS_{GI}}} \quad (10)$$

CSF_d = Absorbed cancer slope factor $(mg/kg/day)^{-1}$; CSF_o = Oral slope factor $(mg/kg\text{-}day)^{-1}$ ABS_{GI} = Fraction of contaminant absorbed in the gastrointestinal tract; DA_d = Dermal Absorbed Dose (mg/kg-day). The CSF for the studied pollutants is in Table 1.

According to USEPA (2011), CR between 1×10^{-6} to 1×10^{-4} represents a range of permissible predicted lifetime risk for carcinogens. Chemicals for which the risk factor falls below 1×10^{-6}, may be eliminated from further consideration as a chemical of concern

RESULTS AND DISCUSSION

Variations in PAHs and pesticide concentrations across the sites: The analysis of PAH concentrations across different sites revealed variations in the amounts of these compounds. Site 1 has detectable levels of only Naphthalene and Acenaphthylene (Figure 2). At site 2, four PAHs were detected: Naphthalene, Dibenzothiophene, Acenaphthene, and Fluoranthene. Among the PAHs found in this site, Naphthalene had the highest concentration at 58.218 ± 34.785 mg/L, followed by Fluoranthene at 2.286 ± 0.001 mg/L. Other PAHs also showed varying average amounts, indicating a diverse distribution of PAHs at this site. At site 3, Naphthalene had the highest level of 62.902 ± 40.516 mg/L. Other PAHs were at far lower concentrations than Naphthalene. Samples from site 4 showed only four PAHs. Naphthalene showed moderate variability, with a standard error of 29.92 mg/L. Acenaphthylene, Acenaphthene, and Anthracene were also detected, but at substantially lower concentrations than Naphthalene. Naphthalene and Acenaphthylene were the most frequently measured PAHs and displayed considerable variability among the different sampling sites. They belong to low molecular weight PAHs.

Table 1: Toxicological characteristics of the organic pollutants[1]

Pollutants	RfD_o (mg/g/day)	ABS_{GI}	Kp (cm/hr)	CSF_O $(mg/kg/day)^{-1}$
Heptachlor	5.0×10^{-4}	1	8.6×10^{-3}	4.50
Dichlorvos	5.0×10^{-4}	1	8.5×10^{-4}	0.29
Aldrin	3.0×10^{-5}	1	1.4×10^{-3}	17.00
Lindane	3.1×10^{-4}	1	1.1×10^{-2}	1.10
Chlorpyrifos	1.0×10^{-3}	1		
Paraquate Dichloride	4.5×10^{-3}	1		
Cypermethrin	1.0×10^{-2}	1		
Glyphosate	1.0×10^{-1}	1		
Imidacloprid	$5.7x10^{-2}$	1		
Cyhalothrin	1.0×10^{-3}	1		
λ-Cyhalothrin	5.0×10^{-3}	1		
Benzo[a]pyrene	3.0×10^{-4}	1	7.0×10^{-1}	1.00
Benzo[a]anthracene	3.0×10^{-2}	1	4.7×10^{-1}	0.10
Naphthalene	2.0×10^{-2}	1	4.7×10^{-2}	
Pyrene	3.0×10^{-2}	1	3.2×10^{-1}	
Fluoranthene	4.0×10^{-2}	1	2.2×10^{-1}	
Acenaphthylene	4.0×10-	1		
Acenaphthene	6.0×10^{-2}	1		
Anthracene	3.0×10^{-1}	1		
Dibenzothiophene	1.0×10^{-2}	1		

(USEPA, 2004; USEPA, 2011; USEPA, 2017; USEA, 2023)

Although low molecular weight PAHs have high volatilization, high oxidation, and easily degraded in water bodies (Qiu *et al.*, 2009; Patricia *et al.*, 2019), their high level in water bodies have been widely reported (Mujić *et al.*, 2017; Awe *et al.*, 2020) as a result of higher solubility in water, continuous release into the aquatic environment, high petrogenic dry deposition from the atmosphere that contains adsorbed low molecular weight PAHs (Ekanem *et al.*, 2019; Patricia *et al.*, 2019; Yao *et al.*, 2023). The results indicating relatively high standard errors for Naphthalene suggest that its concentrations fluctuate considerably across the samples. This variation may indicate different sources (Zhang *et al.,* 2015; Shitandayi *et al.*, 2019). These pollutants can be emitted from natural and anthropogenic sources, including vehicle exhaust, industrial emissions, and combustion of fossil fuels (Jimenez *et al.*, 2009; Tang *et al.,* 2020; Yi *et al.,* 2023). The results for pesticides (Figure 3) indicate that there were also variations across the sites; each site had a different pesticide with the highest concentration. Specifically, Cypermethrin was highest in site 1, Paraquat dichloride in sites 2 and 4, and Chlorpyrifos in site 3. There was a significant difference in the mean amounts of pesticide components at site 1 compared to other sites. There was, however, a lack of statistically significant differences in the mean amounts of pesticide components at site 2, site 3, and site 4. This finding suggests that site 1 experienced unique pesticide usage patterns or different environmental conditions. The major source of pesticide to surface water is runoff from agricultural land. Increased application leads to higher levels (Maloschik *et al*., 2007; Nyantakyi *et al*., 2022). A study by Pimentel (2003) showed that 99.9% of pesticides applied to agricultural targets end up in the environment. Soil characteristics, topography, and chemical properties of the pesticides (such as solubility, sorption capacity, persistence, volatilization, etc.) also play a significant role in the distribution of pesticides across different sites (Stamatis *et al*., 2013; Obidike *et al*., 2020).

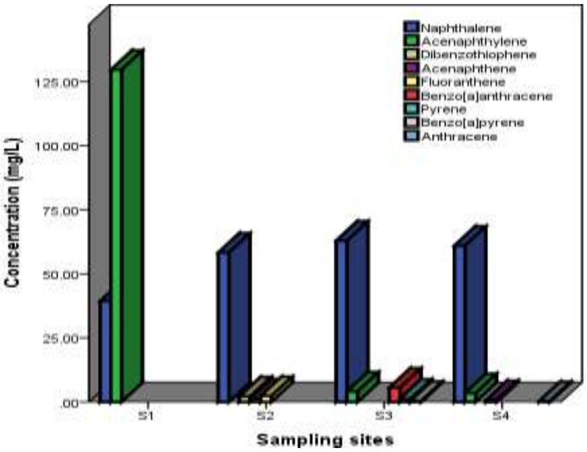

Fig 2: Mean concentrations of PAHs at different sites at the River

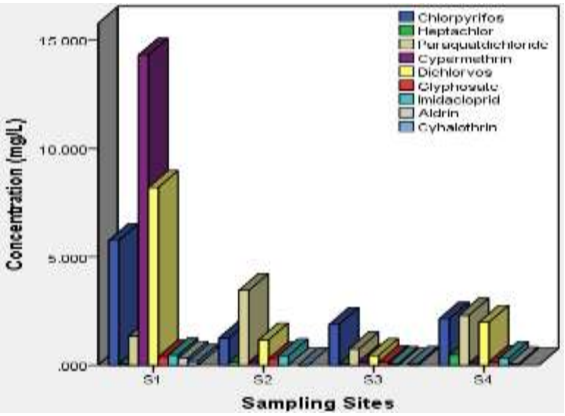

Fig 3: Mean Pesticide concentration in samples collected from different sites

Variations in PAHs and pesticide concentrations across different Seasons and years: During the dry season, the mean values of Polycyclic Aromatic Hydrocarbons (PAHs) (Figure 4) were as follows: Naphthalene (77.84 ± 27.48 mg/L), Acenaphthylene (159.66 ± 157.89 mg/L), Dibenzothiophene (2.09 ± 0.00 mg/L), Fluoranthene (2.29 ± 0.00mg/L), Pyrene (1.22 ± 0.00 mg/L), and Benzo[a]pyrene (0.37 ± 0.00 mg/L).

Naphthalene and Acenaphthylene had the highest average concentration among the measured PAHs, while Dibenzothiophene, Fluoranthene, Pyrene, and Benzo[a]pyrene had comparatively lower average levels. During the rainy season, changes in PAH concentrations in decreasing order were as follows: Naphthalene (49.02 ± 26.29 mg/L), Benzo[a]anthracene (5.56 ± 0.00 mg/L), Acenaphthylene (4.84 ± 1.47 mg/L), Acenaphthene (1.20 ± 0.35 mg/L), and Anthracene (0.28 ± 0.00 mg/L). From the results, only two PAHs (Naphthalene and Acenaphthylene) were detected in dry and rainy seasons.

Dibenzothiophene, Fluoranthene, Pyrene, and Benzo[a]pyrene, which had comparatively lower average levels in the dry season, were not detected in the rainy season. Benzo[a]anthracene, Acenaphthene, and Anthracene, not found in the dry season, were found in the rainy season. Naphthalene and Acenaphthylene have significantly higher concentrations in the dry season than in the rainy season.

The higher PAH levels found in this season were due to low precipitation (low rainfall), leading to low water levels or less dilution (and higher PAH concentration) (Hassan *et al*., 2016; Awe *et al*., 2020), which also reduces the settling of PAH onto the sediment (Akinpelumi *et al*., 2023). Long-range airborne transportation, especially the LMW PAH, which is more significant in summer, also increases the level in the dry season (Wu *et al*., 2024).

The PAHs in the atmosphere usually found their way to water bodies following atmospheric deposition (EA, 2019; Kubo *et al*., 2020). The results also showed that these factors are not the only determinant of the distribution and level of PAHs in a river as seen in Benzo[a]anthracene, Acenaphthene, and Anthracene, which were detected only in the rainy season. The level and distribution of PAHs in water is a complex interplay of factors, including the pollutant source, environmental conditions, and. the physical and chemical properties of the PAH (Oyo-Ita *et al*., 2022; Akinpelumi *et al*., 2023)

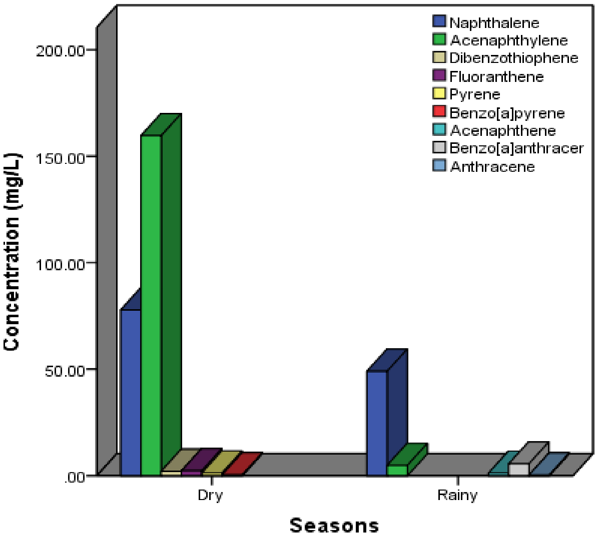

Fig 4: PAHs mean concentrations across seasons in the River

As shown in Figure 5, the following pesticides were detected in dry season: Aldrin (0.004 ± 0.002 mg/L), Chlorpyrifos (2.81 ± 2.02 mg/L), Cyhalothrin (0.10 ± 0.04 mg/L), Heptachlor (0.29 ± 0.17 mg/L), Paraquat dichloride (1.58 ± 0.84 mg/L), Cypermethrin (2.93 ± 2.85 mg/L), Dichlorvos (4.95 ± 3.70 mg/L), Glyphosate (0.19 ± 0.10 mg/L), Imidacloprid (0.17 ± 0.08 mg/L), and lambda-Cyhalothrin (0.08 ± 0.00 mg/L). In the rainy season, Chlorpyrifos (2.31 ± 1.05 mg/L) had the highest level followed by Paraquat dichloride (1.99 ± 0.94 mg/L) and Dichlorvos (0.81 ± 0.21 mg/L). Other detected pesticides are Aldrin (0.34 ± 0.27 mg/L), Heptachlor (0.04 ± 0.02 mg/L), Cypermethrin (0.19 ± 0.15 mg/L), Glyphosate (0.29 ± 0.12 mg/L), Imidacloprid (0.48 ± 0.24 mg/L), lambda-Cyhalothrin (0.22 ± 0.05 mg/L), and Lindane (0.06 ± 0.00 mg/L). The seasonal variation observed showed different patterns for different pesticides. While some recorded higher mean values in the rainy season (Aldrin, Paraquat dichloride, Glyphosate, Imidachlopride, and λ-Cyhalothrin), others had higher mean values in the dry season. Because of the complex nature of factors that determine the seasonal pattern of pesticides in aquatic environments, different studies have obtained different seasonal trends, while some have reported higher mean values of pesticides during the rainy season (Harnpicharnchai *et al*., 2013; Stamatis *et al*., 2013; Jayasiri *et al*., 2022; Nyantakyi *et al*., 2022;), others have reported higher levels in dry season (Konstantinou *et al*., 2006; Cunha *et al*., 2022; Toth *et al*., 2022). The major factor is the seasons of pesticide application, which depends on the locality. The highest levels of pesticides in aquatic environments occur during the period of pesticide application (Liu *et al*., 2018; Kruć-Fijałkowska *et al*., 2022; Wongsa *et al.*, 2023). During the rainy season, higher input from agricultural runoff, low temperature, and low solar radiation intensity (which decrease hydrolysis and photolysis of pesticides) increase the level of pesticides in water systems, while the dilution effect and biodegradation reduce their levels. The reverse is the case for the dry season (Chernyak *et al*., 1996; Harnpicharnchai *et al.*, 2013; Stamatis *et al*., 2013). The different seasonal patterns obtained for different pesticides in this study may also be due to different pesticide use at different seasons and locations in the area. The chemical and physical properties of different pesticides also determine processes like photochemical reaction, biodegradation, sorption, leaching, etc. (Kathrine, 2013; Obidike *et al*., 2020).

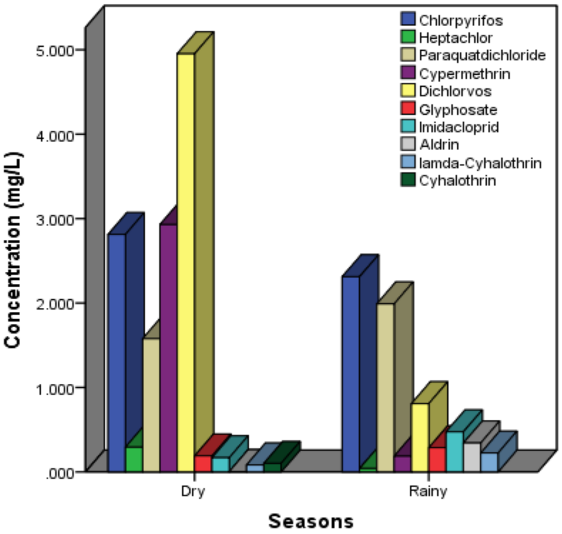

Fig 5: Pesticide concentration in samples collected in the dry and rainy seasons

In year 1, Naphthalene, Acenaphthylene, Dibenzothiophene, Acenaphthene, Benzo[a]anthracene, Fluoranthene, Pyrene, and Benzo[a]pyrene were found, with Acenaphthylene having the highest concentration (159.656 ± 157.889 mg/L) (Figure 6). Next to Acenaphthylene, Naphthalene had the second-highest PAH concentration (71.912 ± 26.302 mg/L⁾. The concentrations of the remaining PAHs detected in year 1 were generally less than 10 mg/L. The levels of PAHs in year 2 were somewhat different from those reported in year 1. First, fewer PAHs were detected in year 2, with only four PAHs compared to eight detections in year 1. Naphthalene also has the highest mean concentration of 21.482 ± 12.421 mg/L, closely followed by Acenaphthylene, with significantly lower levels than the values in year 1. The concentrations of

Acenaphthene and Anthracene were less than 1.00 mg/L in all the samples collected in year 2. Temporal variation in PAHs level is affected by changes in pollution sources coupled with variations in meteorological conditions such as annual rainfall (which changes the water level), temperature, and relative humidity (Launay *et al*., 2013**;** Hassan *et al*., 2019).

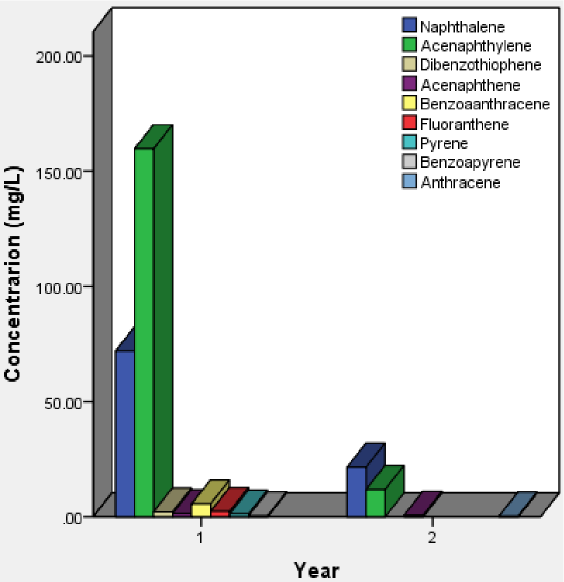

Fig 6: Mean Concentrations of PAHs at different years in the River

In year 1, the pesticides detected were Aldrin, Chlorpyrifos, Cyhalothrin, Heptachlor, Paraquat dichloride, Cypermethrin, Dichlorvos, Glyphosate, Imidacloprid, lambda_Cyhalothrin, and Lindane (Figure 7). The pesticides with the highest concentrations in year 1 were Dichlorvos (4.279 ± 3.113 mg/L), Chlorpyrifos (2.858 ± 1.472 mg/L) and Cypermethrin (2.188 ± 2.024 mg/L). The remaining pesticides had levels generally less than 2.00 mg/L, with Lindane having the lowest detected concentration at 0.062 ± 0.002 mg/L.

In year 2, the following pesticides were detected: Chlorpyrifos, Heptachlor, Paraquat dichloride, Cypermethrin, Dichlorvos, Glyphosate, Imidacloprid, and lambda_Cyhalothrin. Similar to PAHs, most pesticides had higher mean values in year 1, than in year 2. The results showed that Chlorpyrifos had the highest mean concentration (1.758 ± 1.151 mg/L), and was closely followed by Paraquat dichloride (1.582 ± 0.837 mg/L) and Dichlorvos (1.059 ± 0.322 mg/L). The remaining pesticides had levels lower than 1.00 mg/L in year 2. Also, Cypermethrin had the lowest detected concentration (0.041 ± 0.031 mg/L) in year 2.

Non-carcinogenic risk assessment: Assessment of non-carcinogenic risk through oral exposure to the adult human population shows that there was a significant non-cancer risk (HQ > 1) due to the pesticides Chlorpyrifos, Heptachlor, and Paraquat dichloride at all the sites. Aldrin was detected only at sites 1 and 4, while λ-Cyhalothrin was detected only at sites 3 and 4, both with significant non-cancer risks at these sites.

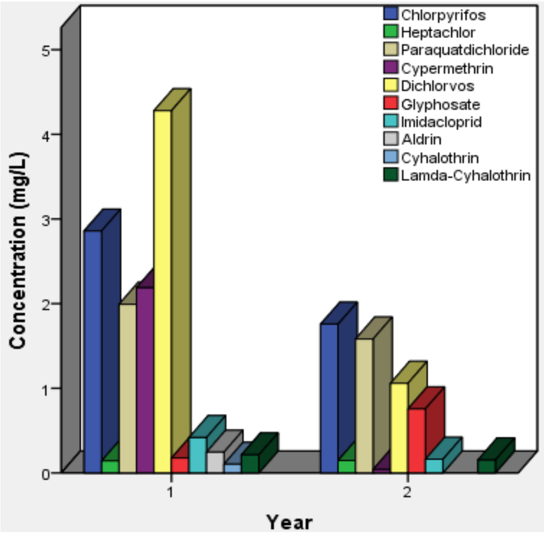

Fig 7: Pesticide concentrations in samples collected in Year 1 and Year 2

Lindane was detected only at site 4 with significant non-cancer risk. There was no non-cancer risk due to Cyhalothrin and Cypermethrin at sites 2, 3, and 4. The level of Glyphosate and Imidacloprid at all the studied sites did not pose a significant non-carcinogenic risk to the population. Exposure to pesticides has been linked to various health disorders such as endocrine disruption (Bergman *et al*., 2013), liver damage, nervous and reproductive disorders, immune suppression, and allergies (Holmgaard and Nielsen, 2009; Terziev and Petkova-Georgieva, 2019). Chronic exposure to pesticides has also been implicated in neuropsychiatric disorders that may lead to suicide attempts (Faria *et al*, 2014). On the other hand, the results of the non-carcinogenic risk from PAHs through oral exposure showed that these pollutants posed significant non-carcinogenic risk at all the sites where they were detected, except Acenaphthene and Anthracene at site 4. PAHs are known endocrine system disruptors (Wilson et al., 2001); many are teratogenic and mutagenic, and some have high incidences of coronary heart disease and diabetes (Baird, 1995). Non-carcinogenic risk, through dermal absorption, was calculated for the pollutants with available dermal permeability coefficients data, and the results are presented in Table 3. The results for pesticides show a significant health risk through dermal exposure for Heptachlor and Dichlorvos at S4 and Aldrin at S1. However, PAHs posed significant non-carcinogenic risks at all the sites detected. The value of the hazard index is proportional to the magnitude of the non-carcinogenic risk of the water to the population. From the calculated hazard index, there was variation in non-cancer risk across the studied sites for both oral and dermal routes. For oral route, HI decreases as follows: S1> S4 > S3 > S3; while for dermal route, it was as follows: R3 > R2 > R4 > R1. The residents around the river's numerous flow points use the river for domestic purposes, irrigation, and often for drinking p. The results show that the residents are at high risk from these pollutants.

Table 2: Non-Cancer Health Risk of Pesticides and PAHs through oral route

	S1		S2		S3		S4	
Pollutants	CDI	HQ_o	CDI	HQ_o	CDI	HQ_o	CDI	HQ_o
Chlorpyrifos	0.1807	785.71	0.0393	170.81	0.0588	255.53	0.0669	291.06
Heptachlor	0.0013	2.5143	0.0038	7.543	0.0013	2.514	0.0148	29.543
Paraquatdichloride	0.0418	9.289	0.1081	24.025	0.0233	5.168	0.0723	16.063
Cypermethrin	0.4500	45.006	0.0038	0.3771	0.0050	0.503	0.0028	0.283
Dichlorvos	0.2577	515.429	0.0364	72.914	0.0138	27.657	0.0622	124.46
Glyphosate	0.0116	0.116	0.0088	0.088	0.0044	0.044	0.0044	0.044
Imidacloprid	0.0145	0.254	0.0135	0.237	0.0019	0.524	0.0094	0.165
Aldrin	0.0101	335.24	-	-	-	-	0.0016	52.381
Cyhalothrin	0.0057	1.14	0.0013	0.26	0.0022	0.44	-	-
λ-Cyhalothrin	-	-	-	-	0.0057	5.70	0.0066	6.60
Lindane	-	-	-	-	-	-	0.00189	6.287
Naphthalene	1.2355	61.77	1.8298	91.49	1.9769	98.84	1.9140	95.70
Acenaphthylene	9.9801	166.34	-	-	0.1267	2.11	0.1059	1.77
Acenaphthene	-	-	0.0657	1.10	-	-	0.0233	0.39
Fluoranthene	-	-	0.0720	1.80	-	-	-	-
Dibenzothiophene	-	-	0.0657	21.90	-	-	-	-
Pyrene	-	-	-	-	0.0383	1.28	-	-
Anthracene	-	-	-	-	-	-	0.0088	0.0293
Benzo[a]anthracene	-	-	-	-	0.1747	5.82	-	-
Benzo[a]pyrene	-	-	-	-	0.0116	38.76	-	-
HI		1926.57		396.06		439.76		627.20

Table 3: Non-Cancer Health Risk of Pesticides and PAHs through Dermal Route

	S1		S2		S3		S4	
Pollutants	DA_d	HQ_d	DA_d	HQ_d	DA_d	HQ_d	Dad	HQ_d
Heptachlor	4.71×10^{-5}	0.094	$1.51\ 10^{-4}$	0.30	5.20×10^{-5}	0.10	5.83×10^{-4}	1.17
Dichlorvos	1.0×10^{-3}	2.00	1.42×10^{-4}	0.28	5.37×10^{-5}	0.11	1.69×10^{-5}	0.03
Aldrin	$6.49\ 10^{-5}$	2.16	-	-	-	-	1.05×10^{-5}	0.35
Lindane	-	-	-	-	-	-	9.98×10^{-5}	O.002
Naphthalene	2.66×10^{-1}	6.65	3.94×10^{-1}	9.85	4.26×10^{-1}	10.65	4.12×10^{-1}	10.3
Pyrene	-	-	-	-	5.64×10-2	1.88	-	-
Fluoranthene	-	-	7.24×10^{-2}	1.81	-	-	-	-
BaP	-	-	-	-	3.76×10^{-1}	1.25	-	-
BaP	-	-	-	-	3.72×10^{-2}	1.24	-	-
HI		10.90		12.24		15.23		11.85

Table 4: Cancer Risk of PAHs and Pesticides through oral route (CR_o) and dermal route (CR_d)

Site	Route	Heptachlor	Dichlorvos	Aldrin	Lindane	Benzo[a] anthracene	Benzo[a] pyrene
S1	CR_o	2.09×10^{-3}	2.91×10^{-2}	6.70×10^{-2}	-	-	-
	CR_d	9.40×10^{-5}	1.29×10^{-4}	4.90×10^{-4}	-	-	-
S2	CR_o	7.05×10^{-3}	4.12×10^{-3}	-	-	-	-
	CR_d	3.04×10^{-4}	1.83×10^{-5}	-	-	-	-
S3	CR_o	2.31×10^{-3}	1.56×10^{-3}	-	-	6.80×10^{-3}	4.48×10^{-3}
	CR_d	1.04×10^{-4}	6.93×10^{-6}	-	-	1.67×10^{-2}	4.47×10^{-3}
S4	CR_o	2.60×10^{-2}	7.01×10^{-3}	1.08×10^{-2}	8.5×10^{-4}	-	-
	CR_d	1.17×10^{-3}	2.18×10^{-6}	7.91×10^{-5}	4.8×10^{-5}	-	-

Carcinogenic risk assessment: Carcinogenic risk (CR) was calculated only for the pollutants with available cancer slope factors. The CR is the probability of developing cancer over a lifetime as a result of exposure to a contaminant. According to USEPA, CR between 1×10^{-6} to 1×10^{-4} falls within the acceptable cancer risk; less than 1×10^{-6} means there is no risk, while CR above 1×10^{-4} indicates significant risk (USEPA, 2011; AEG, 2017). The results for both oral route (CR_o) and dermal route (CR_d) are presented in

Table 4. The results show that the cancer risks through the oral route for all the pollutants are more than 1×10^{-4}, except Lindane at S4 which was within the acceptable range. It implies that there is a possibility of contracting cancer over a lifetime through oral exposure for people who use the water. The results also showed that there is cancer risk through dermal exposure to the carcinogenic PAHs (Benzo[a]anthracene and Benzo[a]pyrene) at S3 and Heptachlor at S1. This means that there is a lifetime cancer risk for the adult population that uses the water for bathing, laundry, recreational activities, and other uses that lead to skin contact. Cancer risk due to dermal absorption for other pollutants falls within the acceptable limit, indicating a tolerable (but not recommendable range). Contaminants within this range are not removed as contaminants of concern but are recommended for continuous monitoring.

Conclusion: In this study, Naphthalene and Acenaphthylene were the most frequently detected PAHs and have significantly higher concentrations in the dry season than in the rainy season. The spatial and temporal variations in the level of the pollutants show that they are affected by complex factors such as the sources, meteorological conditions, and the physical and chemical characteristics of the pollutant. There is a high probability of adverse carcinogenic and non-carcinogenic risks for people who rely on the river for various purposes. From these results, it is recommended that the use of this river for domestic purposes should be highly discouraged, while restraint should be exercised in the use of the water for recreational purposes, laundry, car wash, etc.

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