

Dissemination and risk assessment of polycyclic aromatic hydrocarbons (PAHs) in water and sediment of Buriganga and Dhaleswari rivers of Dhaka, Bangladesh

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ABSTRACT

Concentration, source, ecological and health risks of sixteen polycyclic aromatic hydrocarbons (PAHs) were estimated for water and sediment samples of two urban rivers namely Buriganga River (BR) and Dhaleswari River (DR). The mean concentration of \sum PAHs in BR water and sediment were 9619.2 ngL⁻¹ and 351.6 ngg⁻¹, respectively. Furthermore, the average PAH concentrations detected in DR water and sediment were 1979.1 ngL⁻¹ and 792.9 ngg⁻¹, respectively. The composition profile showed that 3-ring PAHs were dominant in the water matrix; however, 5-ring PAHs were prevalent in the sediment samples of both rivers. Sources apportion study of PAHs indicated that mixed combustion and petroleum sources are responsible for PAHs contamination in the rivers. Ecological risk study of water suggested that the aquatic lives of both rivers are threatened by Fla, BbF, BkF, DahA, and IcdP, as presented above the threshold level. Comparison with sediment quality guidelines (SQGs) indicated that adverse effects might cause occasionally in the sediment ecosystem in DR at certain sampling sites for Nap, Acy, Fl, Phe, Ant, Pyr, Chr, BaP, and DahA. On the other hand, the presence of Nap, Acy and DahA might occasionally cause adverse biological effects in the BR sediment ecosystem. Estimated hazard quotient (HI > 1) and carcinogenic risk (CR_{total} > 10⁻⁴) values indicated that local inhabitants living in the vicinity of the rivers are prone to high health risks.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are planar aromatic molecules with variable carcinogenic potencies determined by their unique structures [1]. They have grabbed the fame of being a persistent organic pollutant from the Stockholm Convention 2001. These

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compounds comprise more than one complex aromatic ring and produce during the incomplete combustion of petroleum products and fossil fuels [2–4]. Source distribution study classified two types of PAHs, petrogenic and pyrogenic. Petrogenic PAHs are generated from fossil fuels, whereas pyrogenic PAHs are produced from the incomplete combustion of different waste materials during cooking and tobacco burning [5,6]. A further classification is based on their molecular weight related to their mischievous effect [7]. To a lesser extent, natural processes such as biomass burning and volcanic eruptions emit PAHs into the environment. Still, they are primarily introduced into the environment by anthropogenic activities in the urban and industrial areas [8].

PAHs are persistent and ubiquitous contaminants, so they are easily bio-accumulated into biota, especially aquatic organisms. In addition, they have a strong affinity for sediment, which disrupts the regular cycle of the water ecosystem due to long-term accumulation into the sediment [9–11]. These may cause chronic or acute toxicity as well as sub-lethal effects. Moreover, PAHs are toxic and harmful to humans since they are considered teratogenic, carcinogenic, and mutagenic to people [12]. Furthermore, among a large number of PAH congeners, the United States Environmental Protection Agency (US EPA) has listed sixteen PAH congeners as priority pollutants based on their acute toxicity to humans and aquatic life and long-term sustainability in nature [12–15]. Regular exposure to these persistent contaminants above the threshold level may adversely affect humans and aquatic organisms. Since PAHs are an issue of major concern, routine monitoring of their contamination levels in the atmosphere is very important. Therefore, to develop effective contaminants control strategies and environmental management systems, gathering information regarding these persistent contaminants, e.g., nature, source identification, and cause-effect relationships, is crucial.

The Buriganga River (BR) and the Dhaleswari River (DR), which are located in the vicinity of Dhaka, fulfill the fundamental water demand for drinking, industrial, and domestic purposes [16], and their water quality is noticeably poor [17]. Both rivers have received garbage and industrial discharge from the city and adjacent areas due to rapid industrial exploration over the last few decades [18,19]. Specifically, the BR has consumed approximately 5,000 cubic meters and 9,000 tons of liquid and solid effluent respectively discharged daily into river banks from 627 dyeing and 104 fertilizer industries, respectively [20]. In addition, this river receives around 63% of total municipal garbage daily [16]. Due to the lack of recycling facilities by Dhaka Water Supply and Sewerage Authority (DWSA), they typically dump the remaining trash at the riverside. Remarkably, the 18-km-long BR is frequently used as a key shipping route for transporting people and goods and consuming a huge amount of leakage fuel from water vehicles.

Furthermore, DR flowing adjacent to the Savar district meets local people's water demand and plays a crucial role in local livelihood. In 2017, the government of Bangladesh relocated the tannery industries from Dhaka city (Hazaribag) to the nearby Savar Export Processing Zone (EPZ), where the poorest inhabitants work to earn their livelihood. These industries discharge approximately 21,600 cubic meters of liquid waste and 88 tons of solid daily in DR [17]. Moreover, they apply open firing to burn leather waste as a waste management technique, contributing to a huge number of pollutants in the river. Additionally, discharge from heavy fuel oil-fired power stations also contributes to pollution. Hence, the targeted rivers are currently experiencing severe contamination stress.

In recent decades, many studies have been conducted on both rivers to investigate the occurrence and distribution of heavy metals and their ecotoxicities in the aquatic environment [21–23]. For example, Ref. [16] worked on heavy metals contamination in the BR. Ref. [17,21,22] investigated physical parameters and heavy metals contamination in the DR. The previous researchers did not concentrate on the contamination of organic pollutants in the targeted study areas, although both rivers consume a significant portion of organic contaminants. Additionally, no research article has been found to assess the contamination level and associated risks of PAHs in these two important rivers. The objectives of the study are (i) to quantify the concentration of PAHs in water and sediment of the Buriganga and Dhaleswari rivers, (ii) to identify possible sources of PAHs emissions, (iii) to evaluate the associated potential ecological and human health risks caused by the targeted PAHs in water and sediment and (iv) to provide valuable information regarding the source and potential threat of PAHs to the local ecosystem.

2. Materials and methods

2.1. Chemicals and reagents

PAH congeners were selected based on the priority pollutant addressed by the US EPA. A total of 16 PAHs compounds was taken for the analysis of collected samples; those compounds are Naphthalene (Nap), Acenaphthene (Ace), Acenaphthylene (Acy), Anthracene (Ant), Fluorene (Fl), Phenanthrene (Phe), Fluoranthene (Fla), Pyrene (Pyr), Benzo [a] anthracene (BaA), Chrysene (Chr), Benzo [b] fluoranthene (BbF), Benzo [a]pyrene (BaP), Benzo [k]fluoranthene (BkF), Dibenzo [a,h]anthracene (DahA), Indeno [1,2,3-cd]pyrene (IcdP), Benzo [ghi]perylene (BghiP). The standard mixture of 16 PAHs was bought from Supelco (EPA 610 Polynuclear Aromatic Hydrocarbons mix 2000 µg/mL) (Kuri and Co., Dhaka, Bangladesh). The naphthalene D8 was used as an internal standard purchased from Sigma Aldrich (Kuri and Co., Dhaka, Bangladesh). Anhydrous sodium sulfate was bought from Sigma Aldrich (Kuri and Co., Dhaka, Bangladesh). All solvents and reagents were obtained from Applichem (Germany) with high-performance liquid chromatography (HPLC) grade.

2.2. Study area

BR is a major water source in Dhaka city, with a population of nearly 10 million people, and flows past the southwest outskirts of the city. Its average depth is 7.6 m (25 ft), and its maximum depth is 18 m (58 ft). Several industrial zones have been established on the bank of this river due to the easy transportation of goods through water vehicles. Moreover, this channel is a major navigation route connecting the capital to the periphery districts. This river receives more than 60,000 cubic meters of toxic waste daily through municipal sewage drainage channels. The main road is just 1 km from the river bank, and air deposition and surface runoff may

contribute a significant portion of pollutants in this river. Besides this, port activities are another source of aquatic pollution. Twenty-six sampling locations (15 water and 11 surface sediment) were selected based on harbor, industrial and municipal activities for sampling. Samples were collected in March 2022. The sampling sites are selected by maintaining approximately 2 km distance from each other and starting from $23^{\circ}74'90.59''\text{N}$, $90^{\circ}33'90.07''\text{E}$ and ending at $23^{\circ}62'80.59''\text{N}$, $90^{\circ}45'10.07''\text{E}$ for BR. Global positioning system (GPS) was used to determine sampling positions (detail is given in the supplementary table, ST-1), and ArcGIS 10.5 software was applied to draw the maps (Fig. 1). For water samples, four sub-samples (0.5 L each) were collected separately from each sampling site. These sub-samples were mixed in an aluminum silo, and a 1 L sample was collected from the composite sample in a Durn glass bottle as a representative sample. Four sub-samples were collected from the river bank for the sediment sample, covering 1 m^2 surface sediment area with 0–20 cm depth for each location using stainless steel dustpan shovel. These sub-samples were mixed in an aluminum silo, and around 500 g of composite samples were stored in a wide-neck glass jar with a steel lid as a representative sample. After collection, all samples were stored in a cooler box and transported to the laboratory. Samples were stored at -20°C until further analysis, and all samples were analyzed within two weeks from the sampling date.

Another targeted river Dhaleshwari River (DR), which is a distributary of 160 km long, emerged from the Jamuna River near the northwestern tip of Tangail District and flowed through the Savar region, remarkable for the export processing zone (EPZ) and heavy fuel-fired power plant. River bank tannery industries and power stations are major contributors to contamination in the studied zone. Therefore, the sampling sites are chosen carefully to maintain an equivalent distance among them. Eighteen samples (9 water and 9 sediment samples) were collected from DR, as shown in Fig. 1. The same procedure was applied to collect the samples.

2.3. Sample extraction and analysis

2.3.1. Water

APHA 6440B method was followed to extract water samples for PAHs analysis. Briefly, the representative sample was first sonicated to make a uniform distribution. After sonication, 500 mL of the sample was taken for liquid-liquid extraction with $3 \times 30\text{ mL}$ Dichloromethane (DCM), and the extracts were combined later. An orbital shaker (brand: Stuart, model: SSL1) and a separating funnel were used to ensure the efficacy of separating the organic layer from the water matrix. The combined extract used deuterium labeled PAH(Naphthalene-D8) as an internal standard. Then, the combined extract was passed through the alumina-silica gel (1:2) with sodium sulfate to remove the aliphatic hydrocarbon. The final extract was intensified to 2 mL using rotatory evaporator (brand: BUCHI, Japan), then vialled for further analysis. Three subsamples were prepared for analysis from each sampling site.

2.3.2. Sediment

Sediment samples were air-dried for 3 days before extraction. Then, visible components such as twigs, broken plastics, leaves, glass fragments, etc., were removed before grinding and sieving to maintain homogeneity. Sieve no. 18 with a 1 mm opening was used for sieving. In the extraction process, around 10.0 g air-dried sediment sample was taken into a scott screw bottle where 30 mL of

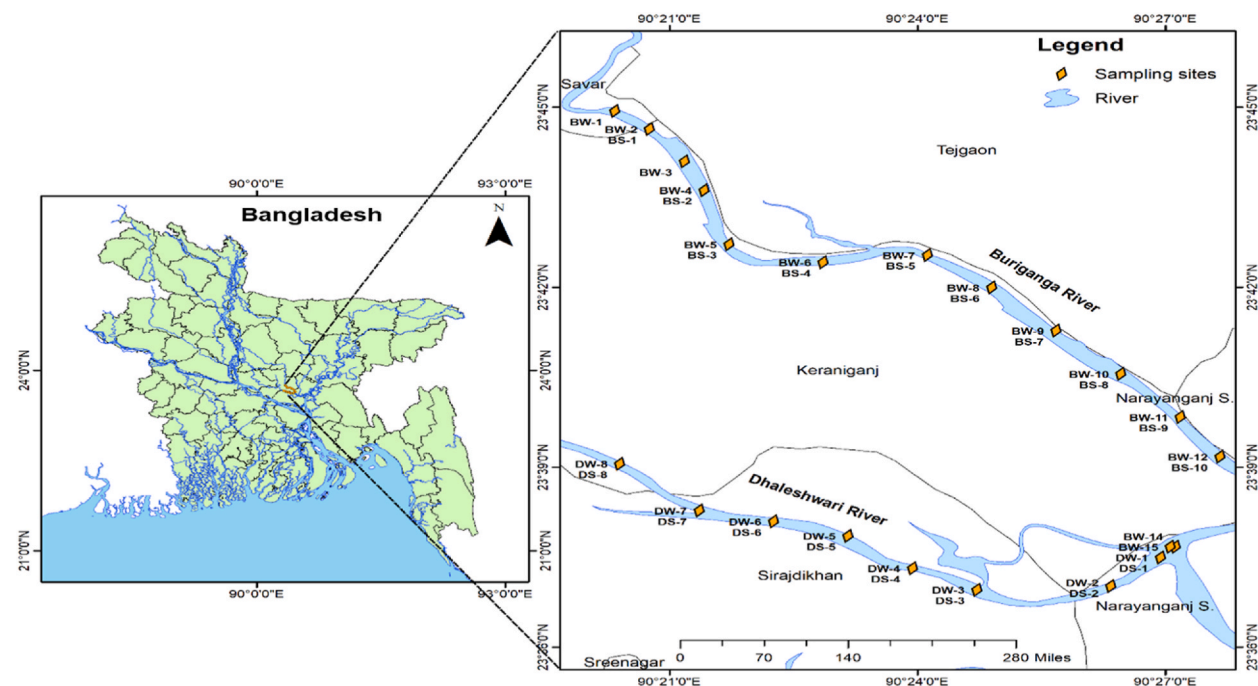


Fig. 1. Sampling sites of water and sediment of BR and DR.

dichloromethane (DCM) was added to the sample and sonicated in an ultrasonicator (brand: LAB Dex, UK) for 15 min and after that, shook in an orbital shaker (brand: Stuart, model: SSL1) with 200 rpm for 30 min. The sample was permitted to stay for 15 min to facilitate the effective separation of the organic layer and solid portion. The extraction process was repeated twice to ensure proper extraction of PAHs from sediment using 30 mL DCM each time. Later the total extract was combined, and naphthalene-D8 was added as an internal standard. Finally, the combined extract was moved through alumina: silica gel in a 1:2 ratio with sodium sulfate to remove the aliphatic hydrocarbon. The final combined extract was concentrated to 2 mL using a rotatory evaporator (brand:BUCHI, Japan) and then stored in 2 mL glass vials for further analysis. From each sampling site, three subsamples were prepared for analysis.

2.4. GC/MS analysis

The whole extract was analyzed by using Gas Chromatographic/Mass Spectroscopic (GC/MS) technique. For the GC/MS, the inert gas was usually used as the carrier gas at a 1 mL/min constant flow rate. In this case, helium gas was used as a carrier gas for the instrument. A specific column for the analysis was framed with fused silica SH-Rxi 5 sil capillary column with a thickness of $30.0 \text{ m} \times 0.25 \text{ mm} \times 0.25 \text{ }\mu\text{m}$. The model was Shimadzu 2010 plus, made in Japan. To evaporate the volatile compound for the final detection, the oven temperature was settled at an 80–300 °C temperature range with a 1 min hold time. The injection volume has been 1.0 μL for every analyzed sample with a split-less injection mode. Column flow was $1.00 \text{ }\mu\text{L min}^{-1}$. Additionally, the injector temperature was 250 °C, whereas the ion source temperature was kept at 200 °C and the interface temperature was 250 °C. In GC/MS, PAHs are quantified by creating a correlation between the amount or concentration of analyte flowing through the detector and the detector response, which utilizes peak area. The peak height in detector mass spectrometry might vary with the mobile phase flow rate.

2.5. Quality assurance and quality control

Firstly, the mixed standard solution with a concentration of 2000 mgL^{-1} was diluted to 5 mgL^{-1} concentration with DCM to prepare a stock standard solution. Secondly, the working standards with concentrations of 10, 20, 40, 50, 100, and $200 \text{ }\mu\text{gL}^{-1}$ were prepared from stock standard solution with the previously mentioned solvent. The instrument calibration was done using these freshly prepared standards. For each PAH, the R^2 value was calculated more than 0.95. After calibration, a solvent blank and a certain standard solution were run as the unknown samples to ensure the quality check. Furthermore, one solvent blank and a working standard were run for every ten samples to check instrument performance. The average spike recovery of all PAHs for certain concentration was detected between 85%–105% and 87%–94% for water and sediment samples, respectively. Furthermore, their relative standard deviation ranged from 1.439% to 14.278%. A similar procedure to that described by Ref. [13] was used to calculate the method detection limit (MDL). MDLs for targeted PAHs were calculated between 0.0027 and $0.0502 \text{ }\mu\text{gL}^{-1}$ for water and 0.0151 and 0.1598 ngg^{-1} for sediment samples, respectively (ST-2). Detected concentration less than MDL was considered as not detected (ND). Therefore, all processes were carried out according to APHA 6440B method with some modifications based on lab and instrument conditions.

2.6. Statistical analysis

Statistical analysis of all PAHs in the water and sediment samples was performed using IBM SPSS statistics software (version 26) for Windows. The normality test for each PAH was done by the Shapiro-Wilk normality test. Results show that all PAHs were normally distributed or log-normally distributed apart from Phe and Ant for the BR water matrix. Moreover, for the sediment of BR, all PAHs were normally distributed or log-normally distributed except DahA. Furthermore, all PAHs were found normally distributed or log-normally distributed unless DahA for DR water. Additionally, unlike Ace, Ant, Phe, and BaA, the remaining PAHs followed normal and log-normal distribution.

The ratio of LMW-PAHs/HMW-PAHs, Ant/(Ant + Phe), Fla/(Fla + Pyr), BaA/(BaA + Chr), and InP/(InP + BghiP) were used to identify the sources of PAHs in the water and sediment samples of both rivers. Moreover, principal components analysis (PCA) and multivariate linear regression (MLR) were applied to determine the PAH sources in the water and sediment samples of both rivers. PCA using the varimax rotation method with eigenvalues >1 was applied to find a reduced set of original variables of data.

2.7. Assessment of eco-toxicological risk

2.7.1. Risk quotient

The risk quotient (RQ) method effectively calculates individual PAH's impact on aquatic organisms [24,25]. The current study estimated RQ to determine the ecological risk to the water ecosystem. The equation and parameters for calculating RQ described in Ref. [26] were applied in the study.

$$RQ = C_i / PNEC$$

where C_i is the concentration of individual PAH. PNEC (predicted no effect concentration) value for individual PAH was taken from Refs. [27,28] and reported that an RQ less than 0.1 indicates low ecological risk, whereas 0.1 and 1 refer to medium risk, and above 1 implies high risk.

2.7.2. Sediment quality guidelines

Sediment Quality Guidelines (SQGs) [29] were recommended to evaluate the adverse biological toxicity of PAHs in the aquatic sediment. In the current study, the used SQG parameters consist of the effect range-low values/effect range–median value (ERL/ERM) and the threshold effect levels/probable effect levels (TEL/PEL). Effects of PAHs on organisms are categorized based on PAH concentrations and compared with two boundary levels, ERL (or TEL) and ERM (or PEL), where amounts less than ERL (or PEL) are considered to be dangerous frequently, and between ERL and ERM indicate occasional hazard risk [30,31]. Ecological risk due to the presence of multi-toxic elements in the sediment samples was calculated by using the mean ERM quotient (M-ERM-Q). According to Ref. [29], the equation is

$$M-ERM-Q = \sum \left(\frac{C_i}{ERM_i} \right) / n$$

where C_i is the concentration of the individual compound, ERM_i is the ERM value of each compound, and n is the total number of PAHs. According to Ref. [32], $M-ERM-Q \leq 0.1$ indicates no harmful biological effect, <0.1 denotes less adverse effect (11% probability of toxicity); $0.11-0.50$ for a potential adverse effect (30% probability of toxicity), $0.51-1.50$ for a moderate adverse effect (46% probability of toxicity), and >1.50 for a significant harmful impact (75% probability of toxicity).

2.7.3. Toxicity equivalents

The toxicity equivalents (TEQs) were applied to assess the potential toxicity of all carcinogenic PAHs in the sediment and were estimated by the given equation

$$TEQs = \sum C_i \times TEF_i$$

where C_i is the concentration of the individual compound and TEF_i represents the toxicity equivalency factor for each PAH.

2.8. Health risk assessment

According to USEA standard model, carcinogenic and non-carcinogenic risks to the local inhabitants were estimated as they expose to surface sediments and waters through dermal contact and ingestion pathways. The exposure dosage via ingestion and the dermal route was calculated by the following equations

$$CDI = (C_w \times IR \times ED \times EF) / (BW \times AT)$$

$$DAD = (DA_{event} \times EV \times ED \times EF \times SA) / (BW \times AT)$$

$$DA_{event - watercontact} = 2 \times FA \times Kp \times C_w \times \sqrt{(6 \times \tau_{event} \times t_{event}) / \pi}$$

$$DA_{event - sediment} = C_s \times CF \times AF \times ABS_d$$

where CDI is chronic daily ingestion through water ($\text{mg.kg}^{-1}\text{day}^{-1}$), DAD represents dermal absorbed dose through sediment and water contact ($\text{mg.kg}^{-1}\text{-day}$), DA_{event} shows absorbed dose per event ($\text{mg.cm}^2\text{-event}$) [33]. Also, dermal and ingestion cancer risks and hazard quotients were defined as:

$$\text{Ingestionhazardquotient} = CDI / RfDo$$

$$\text{Dermalhazardquotient} = DAD / (RfDo \times ABS_{GI})$$

$$\text{Ingestioncancerrisk} = CDI \times SFo$$

$$\text{Dermalcancerrisk} = DAD \times SFo / ABS_{GI}$$

$$HI = \sum HQs$$

All parameters for computing hazard quotient (HQ) and cancer risk (CR) described by Ref. [34] were applied in this study. Moreover, HI is a hazard index of non-cancer carcinogenic effects through the two exposure pathways. If the HI (non-cancer risk) is < 1 , it indicates no significant non-carcinogenic risk; however, if the HI is > 1 , there is a chance that non-carcinogenic effects might occur. Cancer risks are classified as: very low when the estimated value is $\leq 10^{-6}$, low from 10^{-6} to $<10^{-4}$, moderate from 10^{-4} to $<10^{-3}$, high from 10^{-3} to $<10^{-1}$ and very high when the value is $\geq 10^{-1}$ [35].

Table 1
Concentration of PAHs in the water and sediment samples from BR and DR.

PAHs	BR							DR							MACwater (μgL^{-1}) ^a		
	Water (n = 15)				Sediment (n = 11)			Water (n = 9)				Sediment (n = 9)					
	Mean (ngL^{-1})	SD	Range (ngL^{-1}) Min Max		Mean (ngg^{-1})	SD	Range (ngg^{-1}) Min Max	Mean (ngL^{-1})	SD	Range (ngL^{-1}) Min Max		Mean (ngg^{-1})	SD	Range (ngg^{-1}) Min Max			
Nap	849.1	117.5	667.7	1080.1	27.0	14.2	10.4	54.2	338.5	167.1	200.3	747.4	40.9	27.7	11.3	88.3	
Acy	91.7	79.8	13.0	271.4	23.5	20.0	0.1	70.9	34.5	5.4	28.5	45.8	6.0	4.5	2.9	17.3	
Ace	255.5	260.1	52.2	989.1	1.9	1.4	0.5	5.6	171.2	45.4	120.3	254.1	11.0	7.3	6.1	24.1	
Fl	801.7	848.5	136.1	3247.8	4.5	2.2	1.4	8.3	93.0	8.6	78.6	106.4	21.1	10.5	9.3	38.9	
Phe	3262.3	1809.7	1113.3	5888.1	19.4	8.9	5.7	39.7	362.8	134.9	71.6	562.7	103.1	63.7	53.2	223.6	
Ant	590.9	625.3	81.4	2312.6	6.1	8.4	0.8	30.7	34.6	6.5	24.3	45.3	19.4	16.7	6.8	51.2	
Fla	1855.7	1623.4	365.1	5929.5	39.7	14.7	9.8	59.0	226.0	93.0	107.6	367.0	16.8	28.1	0.3	71.9	
Pyr	1072.6	1103.4	158.5	3737.1	22.4	14.0	4.5	46.9	207.1	70.5	103.9	289.7	93.6	71.1	10.7	213.9	
BaA	88.6	106.1	11.2	370.6	14.1	11.0	2.0	34.6	37.1	39.1	6.5	110.5	33.8	21.5	ND	49.6	1
Chr	149.8	181.7	20.6	683.4	15.4	10.5	2.2	35.3	53.3	44.4	6.3	144.8	81.3	44.7	30.5	161.2	2
BbF	458.8	218.6	172.0	757.4	131.0	107.9	ND	303.5	133.1	119.2	39.3	338.1	62.4	45.0	6.5	119.8	2
BkF	75.0	119.3	1.9	405.4	20.0	13.5	3.0	45.3	74.5	34.2	39.3	138.6	93.8	110.4	22.9	297.4	2
BaP	21.7	36.2	0.3	101.7	5.8	5.3	ND	16.4	60.7	79.1	10.2	207.7	119.0	88.1	32.6	273.9	2
DahA	12.1	18.8	0.4	67.8	7.7	6.7	0.5	20.2	60.5	75.3	14.0	195.1	31.7	32.3	ND	85.4	3
BghiP	3.4	3.8	0.2	12.2	1.7	2.1	ND	6.8	37.6	32.7	5.7	94.0	13.7	18.4	1.4	56.5	
IcdP	30.3	59.8	0.5	232.7	11.5	9.9	2.1	31.5	54.5	52.3	22.4	188.9	45.2	37.4	10.9	107.3	4
Σ PAH	9619.2	7212.0	2794.2	26086.8	351.6	198.8	90.7	734.2	1979.1	47.0	878.7	3836.0	792.9	407.9	205.4	1880.3	

ND= Not detected; SD= Standard deviation.

^a MAC-Maximum allowable concentration of PAHs for surface water according to United State Environmental Protection Agency (2013).

3. Results and discussion

3.1. Concentration of PAHs in the surface water and sediment of BR

3.1.1. Water

The sum of the average concentration of 16 PAHs was detected in the BR water ranging from 2794.2 ngL⁻¹ to 26086.8 ngL⁻¹ with a mean concentration of 9619.2 ngL⁻¹ as shown in Table 1. In detail, the concentration of 2-ring PAHs (Nap) was between 667.7 ngL⁻¹ and 1080.1 ngL⁻¹. The 3-ring PAHs (Acy, Ace, Fl, Phe, and Ant) were detected from 91.7 ngL⁻¹ to 3262.3 ngL⁻¹. The concentrations of 4-ring PAHs (Fla, Chr, BaA, and Pyr), 5-ring PAHs (BbF, BkF, BaP, and DahA), and 6-ring PAHs (BghiP and IcdP) ranged from 88.6 ngL⁻¹, 12.1 ngL⁻¹ and 3.4 ngL⁻¹ to 1072.6 ngL⁻¹, 458.8 ngL⁻¹ and 30.1 ngL⁻¹. The dominance of contaminants has followed the order of Phe > Fla > Pyr > Nap > Fl > Ant > BbF > Ace > Chr > Acy > BaA > BkF > IcdP > BaP > DahA > BghiP. The average \sum PAHs concentration of 15 water sampling sites varied from 3167.2 ngL⁻¹ (BW-11) to 20767.4 ngL⁻¹ (BW-1). According to the classification of pollution levels for river water provided by Ref. [36], all water sampling sites of the studied area were highly polluted as \sum PAHs concentration was detected to be more than (>1000 ngL⁻¹). Moreover, there was no significant difference in PAHs concentration among sites ($p < 0.005$) of both matrices, indicating the origin of the PAHs might be similar. However, the total PAH concentration was high at sampling locations from BW-1 to BW-8.

Moreover, the compositional profile of PAHs is shown in Fig. 2 (a). 3-ring PAHs were found as dominant contaminants in the water matrix contributing around 56.6% of total PAHs contamination in the studied area. High water solubility and relative vapor pressure might explain the abundance of low molecular weight PAHs (2-ring and 3-ring) in the water matrix [4,37,38]. This finding is consistent with the studies of Ref. [4,10,11,34]. Additionally, 2-ring, 4-ring, 5-ring, and 6-ring PAHs were responsible for 11.1% 26.5%, 5.6%, and 0.3% contributions, respectively.

3.1.2. Sediment

As mentioned in Table 1, a sum of the average concentration of 16 \sum PAHs detected in the BR sediment ranging from 90.7 ngg⁻¹ in dry weight (d.w.) to 734.2 ngg⁻¹ (d.w.) with a mean concentration of 351.6 ngg⁻¹ (d.w.). The 2-ring PAH (Nap) concentration was observed between 10.4 ngg⁻¹ and 54.2 ngg⁻¹. The 3-ring PAHs (Acy, Ace, Fl, Phe, and Ant) were detected between 1.9 ngg⁻¹ and 23.5

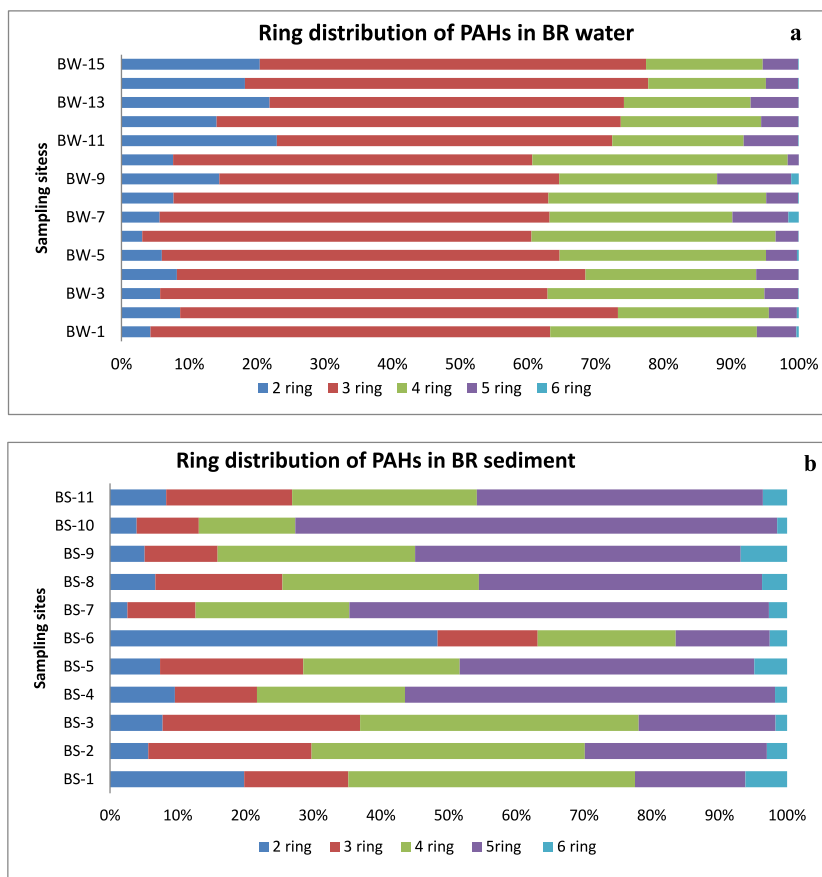


Fig. 2. Ring distribution of PAHs in BR water (a) in BR water, (b) in BR sediment, (c) in DR water, and (d) in DR sediment.

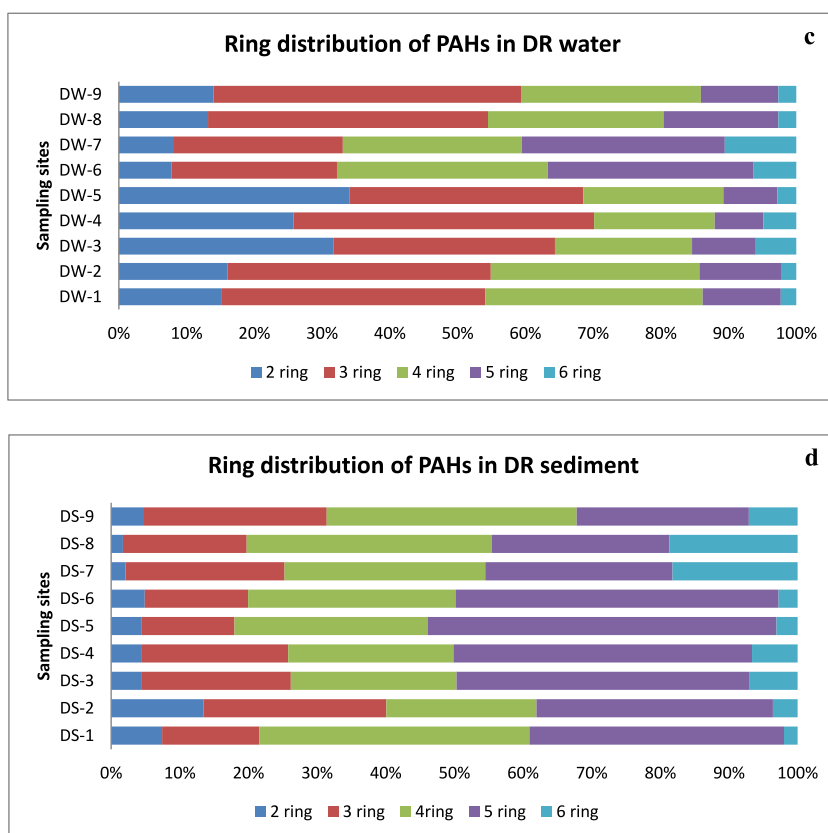


Fig. 2. (continued).

ngg^{-1} (d.w.). The concentration of 4-ring PAHs (Fla, Chr, BaA, and Pyr), 5-ring PAHs (BbF, BkF, BaP, and DahA) and 6-ring PAHs (BghiP, and IcdP) varied from 14.1 ngg^{-1} , 5.8 ngg^{-1} and 1.7 ngg^{-1} to 39.7 ngg^{-1} , 131 ngg^{-1} , and 11.457 ngg^{-1} , respectively. The dominance of PAHs has followed the descending order of BbF > Fla > Nap > Acy > Pyr > BkF > Phe > Chr > BaA > IcdP > DahA > Ant > BaP > Fl > Ace > BghiP. BbF was the most dominant with a mean value of 131 ngg^{-1} . According to the classification of sediment pollution level given by Ref. [39], each sampling site in this study area is moderately polluted except sampling site BS-6, which concentration was detected lower than 100 ngg^{-1} . Furthermore, as provided in Fig. 2(b), the ring distribution study shows that 5-ring PAHs strongly polluted the sediment of the studied area with 40.068% of the total PAHs. Significantly, these contaminants contributed more than 50% contamination at sampling sites BS-4, BS-7, BS-9, and BS-10. The strong hydrophobic nature and resistance to HMW PAHs' biodegradability could be the reason for their higher enrichment in the river sediment [37,40,41]. The abundance of 5-ring PAHs in the river sediment was also found by Refs. [11,36]. In addition, 2-ring, 3-rings, 4-ring, and 6-ring PAHs contributed 11.4%, 16.7%, 28.3%, and 3.5% pollution, respectively. Therefore, LMW PAHs were found dominant in the water, and HMW PAHs have been detected as dominants in the sediment of the studied area, and this observation is coherent with the findings of Ref. [4,10,11,34,42].

3.2. Concentration of PAHs in the surface water and sediment of DR

3.2.1. Water

The total mean concentration of 16 PAHs detected in the DR water ranged from 878.7 ngL^{-1} to 3836 ngL^{-1} with a mean concentration of 1979.1 ngL^{-1} as shown in Table 1. The concentration of 2-ring PAH (Nap) was between 200.3 ngL^{-1} and 747.4 ngL^{-1} . The 3-ring PAHs (Acy, Ace, Fl, Phe, Ant) were detected from 34.5 ngL^{-1} to 362.8 ngL^{-1} . Ranged from 53.3 ngL^{-1} , 60.5 ngL^{-1} , and 37.6 ngL^{-1} to 226 ngL^{-1} , 133.1 ngL^{-1} , and 54.5 ngL^{-1} were found for 4-ring PAHs (Fla, Chr, BaA, and Pyr), 5-ring PAHs (BbF, BkF, BaP, and DahA) and 6-ring PAHs (BghiP and IcdP) respectively. The availability of PAHs is followed by the descending order of Phe > Nap > Fla > Pyr > Ace > BbF > Fl > BkF > BaP > DahA > IcdP > Chr > BghiP > BaA > Ant > Acy. The most dominant contaminant, Phe contributed a total of 18.3%, closely followed by Nap, contributing 17.1% of the total pollution. The mean concentration of \sum PAHs based on sampling location varied from 1107.5 ngL^{-1} (DW-3) to 2580.8 ngL^{-1} (DW-7). Depending on the classification of water pollution levels introduced by Ref. [37], each sampling site of the studied area is highly polluted as \sum PAHs levels were detected higher than 1000 ngL^{-1} . Likewise, BR, all PAHs were found to be significantly distributed across the sampling site ($p < 0.05$) in both matrices. Furthermore, Fig. 2(c) displays the compositional analysis of PAHs as a mean of ring distribution. The study shows 2-ring, 3-ring,

4-ring, 5-ring, and 6-ring contributed 18.4%, 36.1% 25.7%, 15.2%, and 4.5% of the total contamination, respectively. Additionally, like the BR water system, 3-ring Σ PAHs were detected as dominant contaminants in the DR water system. 3-ring PAHs occupied more than 50% of the total PAHs at five sampling locations, namely DW-1, DW-2, DW-4, DW-8, and DW-9. High water solubility and benthic recycling could be the reasons for the availability of LMW PAHs in the aquatic system [11,43].

3.2.2. Sediment

As mentioned in Table 1, the concentration of 16 Σ PAHs detected in the DR sediment ranged from 205.4 ngg⁻¹ in dry weight (d.w.) to 205.4 ngg⁻¹ (d.w.) with a mean concentration of 792.9 ngg⁻¹ (d.w.), which is two-fold higher than the found concentration of BR. The 2-rings PAH (Nap) concentration was observed between 11.3 ngg⁻¹ and 88.3 ngg⁻¹. The 3-ring PAHs (Acy, Ace, Fl, Phe, and Ant) were detected between 6.1 ngg⁻¹ and 103.1 ngg⁻¹. Ranged from 16.8 ngg⁻¹, 31.7 ngg⁻¹, and 13.7 ngg⁻¹ to 93.6 ngg⁻¹, 119 ngg⁻¹, and 45.2 ngg⁻¹ were investigated for 4-ring PAHs (Fla, Chr, BaA, and Pyr), 5 ring PAHs (BbF, BkF, BaP, and DahA) and 6-ring PAHs (BghiP and IcdP) respectively. The dominance of PAHs has followed the descending order of BaP > Phe > BkF > Pyr > Chr > BbF > IcdP > Nap > BaA > DahA > Fl > Ant > Fla > BghiP > Ace > Acy. According to Ref. [39] classification of pollution levels in sediments, each sampling site is moderately polluted, except for DS-3 and DS-4, where concentrations exceeded 1000 ngg⁻¹. Among HMW PAHs, Pyr, Chr, BkF, and BaP were found to be dominant pollutants in these sampling sites. Furthermore, as given in Fig. 2(d), the ring distribution study shows that the sediment samples of the studied area were strongly polluted through 5-ring PAHs with a contribution of 37.1%. These contaminants contributed more than 50% contamination at sampling sites DS-5 and DS-6. More resistance to microbial degradation and the high adsorptive nature of HMW PAHs might be the possible reasons for dominance in the sediment [44]. Additionally, 2-ring, 3-ring, 4-ring, and 6-ring PAHs contributed 5.3% 20%, 30%, and 7.7%, respectively. Hence, the water matrix was strongly loaded with LMW PAHs, and the sediment was highly loaded with HMW PAHs. These findings are similar to the study of Ref. [4,10,11,34,42].

The sum of the mean PAHs in BR water was detected fourth times higher than in DR water. Rapid port and navigation activities, discharge from fuel refineries, municipal waste discharge, and burning and anthropogenic activities might be the probable reasons for detecting high concentrations in the BR water matrix. In contrast, the studied DR area is not utilized as the major navigation route, which might be the possible reason for detecting low PAH levels. Additionally, compared with other polluted rivers in the world (Table 2), the total PAHs level in two rivers was found to be higher than Daliao river (748.8 ngL⁻¹; [45]), Tiber river (90.5 ngL⁻¹; [11]), Songhua river (342.2 ngL⁻¹; [46]), Shadegan wetland (78 ngL⁻¹; [34]), Hoor Al-Azim wetland (76.94 ngL⁻¹; [12]), Yangtze River (19.8 ngL⁻¹; [47]), and Yellow river delta (496 ngL⁻¹; [48]). In contrast, the detected concentration is considerably lower than in Algoabay (74.6 μ gL⁻¹; [36]).

The sum of the mean PAH concentration in the DR sediment was detected as two-fold higher than BR. When compared to other rivers around the world, the concentration of PAHs in the DR was found to be higher than the average PAHs concentration in the Yangtze (690 ngg⁻¹; [47]), Mahakam River (611.1 ngg⁻¹; [49]), Pearl River (346.8 ngg⁻¹; [50]), Subarnarekha River estuary (223.5 ngg⁻¹; [51]), Han River (679.6 ngg⁻¹; [52]), Beibu Gulf (146 ngg⁻¹; [53]), and Hoor Al-Azim wetland (51.7 ngg⁻¹; [12]). Furthermore, the detected concentration in BR was observed to be higher than Pearl River (346.8 ngg⁻¹; [50]), Subarnarekha River estuary (223.5 ngg⁻¹; [51]), Beibu Gulf (146 ngg⁻¹; [53]), and Hoor Al-Azim wetland (51.7 ngg⁻¹; [12]).

3.3. Sources identification

3.3.1. BR

Diagnostic ratios and PCA are widely used to evaluate PAH sources in the water and sediment matrices. LMWs/HMWs, Ant/(Ant + Phe), Fla/(Fla + Pyr), BaA/(BaA + Chr), and InP/(InP + BghiP) are considered prominent molecular ratios to identify the sources of

Table 2

Concentration ranges of Σ PAHs in the water and sediment samples from the different rivers in the world.

Location	Number of PAHs	Water ngL ⁻¹	Sediment ngg ⁻¹	Reference
		Range	Range	
Daliao river, China	16	71.12–4255.43 ngL ⁻¹	374.84–11588.85 ngg ⁻¹	[43]
Tiber river, Italy	16	10.3–951.6 ngL ⁻¹	36.2–545.6 ng g ⁻¹	[11]
Songhua river, China	16	32.5–108 ng L ⁻¹	–	[44]
Shadegan wetland, Iran	16	42–136 ng L ⁻¹	10000–317000 ng g ⁻¹	[32]
Hoor Al-Azim wetland, Iran	16	15.3–160.15 ngL ⁻¹	15780–410200 ngg ⁻¹	[12]
Yangtze River, China	16	6.95–43.2 ngL ⁻¹	32.8–3790 ngg ⁻¹	[45]
Yellow river delta, China	16	113 to 1533 ngL ⁻¹	–	[46]
Algoabay, South Africa	16	ND–24660 ngL ⁻¹	ND–5230 ngg ⁻¹	[40]
Mahakam River, Indonesia	16	–	54.7–2256.15 ngg ⁻¹	[47]
Pearl River, China	16	–	73.68–933.25 ngg ⁻¹	[48]
Subarnarekha River estuary, India	16	–	36.8–670.8 ngg ⁻¹	[49]
Han River, China	16	18.3–146.8 ngL ⁻¹	137.1–1478.4 ngg ⁻¹	[50]
Beibu Gulf, China	16	57.9–90.8 ngL ⁻¹	–	[51]
Buriganga River, Bangladesh	16	2794.2–26086.8 ngL ⁻¹	90.7–734.2 ngg ⁻¹	This study
Dhaleswari rivers, Bangladesh	16	878.7–3836.0 ngL ⁻¹	205.4–1880.3 ngg ⁻¹	This study

ND = Not detected.

PAHs [2,54]. LMWs/HMWs >1 indicates PAHs are generated from petrogenic sources, and LMWs/HMWs <1 indicates the pyrogenic origin of PAHs [2,55]. LMWs/HMWs >1 was detected in the water (ST-3), suggesting fuel or light petroleum products could be the possible sources of PAHs. Generally, LMW PAHs such as Nap, Acy, Ace, Fl, Fla, Phe, and Ant are found abundantly in fossil fuels as alkylated derivatives [56,57]. Bi and tricyclic aromatic compounds were responsible for approximately 67.7% pollution of BR water. Therefore, spillage/accidental release of fossil fuel from water vehicles like launch, speed boats, cargo, motor boats, etc., might be the primary culprit of pollution as the studied area is one of the major navigation routes. Additionally, municipal sewage discharge, surface runoff, and effluent from oil refinery industries near the port area could be other possible sources of LMW PAHs in the studied area. In contrast, LMWs/HMWs <1 were found for 89% of sediment sampling sites indicating pollutants discharged due to the combustion of biomass and fuel (ST-4). Combustion and incomplete combustion products usually emit HMW PAHs [18]. In our studied area, HMW PAHs contributed to about 71.9% of contamination in the sediment sampling sites, and we suspect that combustion products might be the possible sources of pollution. Open burning of municipal waste, black smoke from automobiles and road construction sites emit vast amounts of HMW PAHs, which might mix with the river through air deposition and municipal drainage system, and surface runoff results in pollution in the aquatic environment. As shown in Fig. 3(a), $\text{Ant}/(\text{Ant} + \text{Phe}) > 0.1$ was detected for 46.7% of water sampling sites (BW-1 to BW-7) and all sediment sampling sites, which attributes the pyrolytic origin of PAHs. Combustion of municipal solid wastes, incomplete combustion of fossil fuel in automobiles, wood, and biomass combustion in road construction activities, etc., are prominent sources of pyrogenic Ant and Phe [3,58,59]. It is assumed that municipal waste burning, black smoke from heavy-duty automobile vehicles on the main road, and road construction activities might be responsible for Ant and Phe in those sites, where the influx of pollutants might runoff from non-point sources to the studied area through the municipal drainage system and air transportation. In contrast, $\text{Ant}/(\text{Ant} + \text{Phe}) < 0.1$ was found for water sampling location BW-8 to BW-15, which attributes petrogenic regeneration of PAHs (Fig. 3(a)). These mentioned sites are adjacent to or at the jetty, from where more than a thousand water vehicles navigate at different destinations of the country, and more than 200 launches are docked in the harbor. Fuel spillage from water vehicles might significantly contribute to Ant and Phe contamination in the respective water sampling sites. Moreover, discharged effluent from oil refineries in the harbor area might be another probable source. Besides, $\text{Fla}/(\text{Fla} + \text{Pyr}) < 0.4$ indicates petroleum origin, $\text{Fla}/(\text{Fla} + \text{Pyr}) = 0.4\text{--}0.5$ represents liquid fossil fuel combustion, and $\text{Fla}/(\text{Fla} + \text{Pyr}) > 0.5$ attributes grass, wood, or coal combustion [2,60]. $\text{Fla}/(\text{Fla} + \text{Pyr}) > 0.5$ ratio was found in water and sediment, indicating the pyrogenic origin of Fla and Pyr (Fig. 3(a)). Like pyrogenic Ant and Phe, municipal waste and biomass burning in road construction might be the possible sources of Fla and

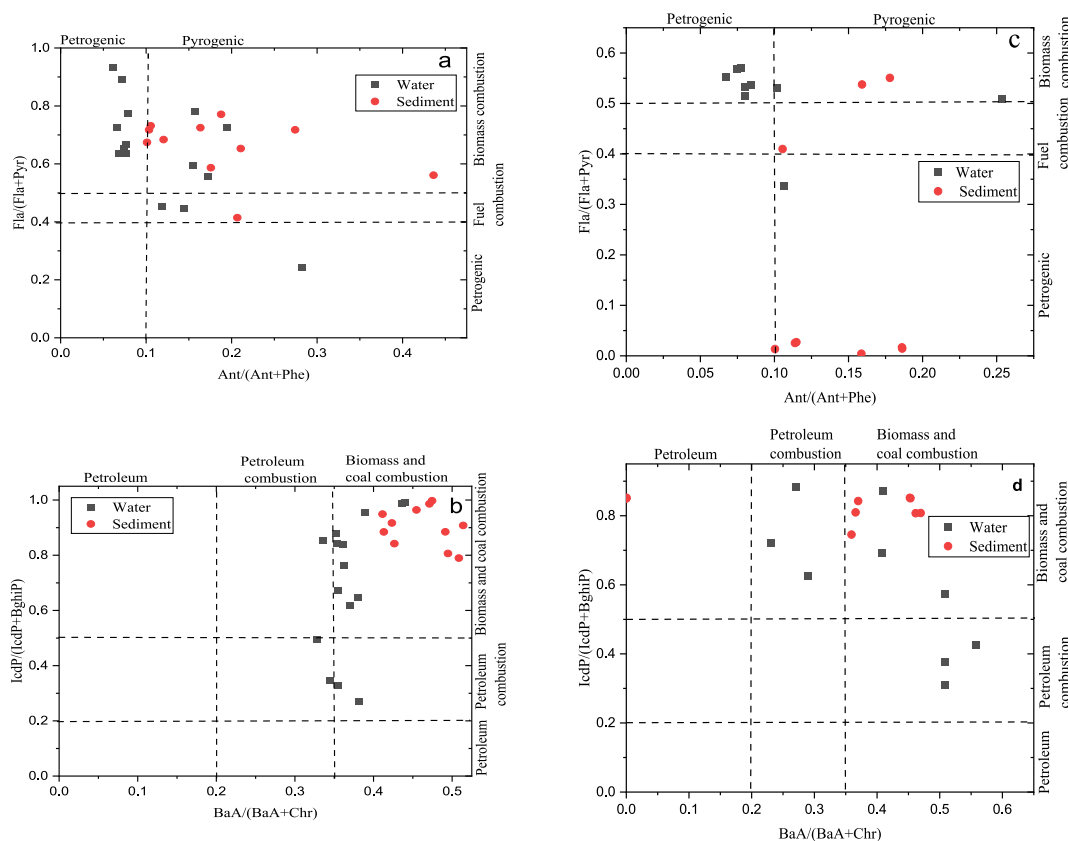


Fig. 3. (a) Isomeric ratios $\text{Ant}/(\text{Ant} + \text{Phe})$ vs. $\text{Fla}/(\text{Fla} + \text{Pyr})$ for BR water and sediment, (b) isomeric ratios $\text{BaA}/(\text{BaA} + \text{Chr})$ vs. $\text{IcdP}/(\text{IcdP} + \text{BghiP})$ for BR water and sediment, (c) Isomeric ratios $\text{Ant}/(\text{Ant} + \text{Phe})$ vs. $\text{Fla}/(\text{Fla} + \text{Pyr})$ for DR water and sediment, and (d) isomeric ratios $\text{BaA}/(\text{BaA} + \text{Chr})$ vs. $\text{IcdP}/(\text{IcdP} + \text{BghiP})$ for DR water and sediment.

Pyr in the studied area. However, for water sampling sites BW-1, BW-3, and BW-4, Fla/(Fla + Pyr) was found to be < 0.4 and $0.4\text{--}0.5$, respectively, indicating differential impacts of fossil fuel discharge and traffic emission [11]. BaA/(BaA + Chr) > 0.35 was assessed for water ($0.3\text{--}0.4$) and sediment ($0.4\text{--}0.5$) sampling sites, representing vehicular emissions that might be the possible sources of pollution (Fig. 3(b)). Moreover, IcdP/(IcdP + BghiP) > 0.5 was observed for the studied area, which suggests biomass burning could be a possible source of pollution in both matrices (Fig. 3(b)).

Molecular ratios of PAHs indicate that pyrogenic sources like combustion of biomass and fuel are the primary contributor to pollutants in the studied area; moreover, petrogenic sources such as fuel spillage from water vehicles are another possible source. Dhaka, the major industrial city of Bangladesh, situates on the bank of the BR having textile, chemical, pharmaceutical, garments, and petrochemical industries. The emission of particles due to the burning of solid effluent from these industries might transport through the municipal sewage system and air and deposited into the river. In addition, emissions from heavy vehicles load on the road and municipal solid waste burning sites result in severe air pollution, which might cause possible pyrogenic PAHs contamination in the studied area. Furthermore, heavy water traffic through the studied river channel might cause a significant level of petrogenic PAHs pollution. It is suspected that wood burning as fuel for cooking in the harbor area's restaurants could be a potential source as there are more than 50 restaurants located in the jetty area. Therefore, both petrogenic and pyrogenic activities might be the possible sources of PAHs in the mentioned area of BR.

Besides molecular ratio, PCA was applied to investigate point sources of PAHs emission. PCA was conducted for sixteen PAHs in the 15 water and 11 sediment sampling sites with the loading of 86.187% and 88.411% total variance, respectively (ST-7). For water sampling sites, the first principal component (PC-1) contained 65.4% of total variance with positive loading of LMW PAHs (Acy, Ace, Fl, Phe, and Ant) and HMW PAHs (Pyr and Chr), indicating both pyrogenic and petrogenic activities were responsible for the pollution. Also, the 3-ring compounds might be discharged from regularly commuted water vehicles, refineries industries, and combustion of fuel and biomass [61,62]. Pyr and Chr came from liquid fossil fuel combustion from water vehicles and automobiles [63,64]. The second principal component (PC-2) contains 14% of the total variance with a strong association of BkF, BaP, DahA, and IcdP, indicating possible sources are related to vehicular exhaust emission and biomass combustion [34,65]. Remarkably, BkF, BaA, and BaP may consider a marker for PAH emission from vehicular exhaust emission and curd oil refineries [66]. PC-3 has a total of 6.7% variance with the loading of Nap and Fla, indicating emerging pollutants might occur due to petrogenic activities. The presence of Fla indicates heavy-duty diesel combustion [67] in the studied area. Moreover, river bank oil refinery centers near the port area could be another possible source of Nap.

In sediment, PC-1 (67.6% of total variance) was positively loaded with Pyr, BaA, Chr, BkF, BaP, DahA, BghiP, and IcdP (ST-7). HMW PAHs are the possible marker of incomplete combustion of fuel, open burning, and pyrolysis [11,68]; thus, fuel discharge from water vehicles and curd oil refineries is responsible for significant contamination [12]. The studied area sediment was highly contaminated with 5-ring PAHs with a value of about 44%, which could be resulted from the open burning of municipal waste like food, e-waste, garments, etc., in the nearest landfill or vehicular exhausts from the road and water traffic. Additionally, the rural pocket is situated on the other side of the river, where low-income people use wood as domestic fuel, which might be another possible source of pollution. PC-2 explained 12.6% of the total variance with a positive contribution of Nap, Ace, Fl, Phe, and Ant. Heavy loading of 3-ring PAHs indicates pollution by petroleum products and combustion of petroleum products [45]. Fuel discharged from docked water vehicles as fuel contains a higher percentage of LMW PAHs as alkylated derivate [69], and incomplete combusted fuel from commuted cars may result in sediment contamination in the studied area. PC-3 has a 14% of total variance with high loading of Acy, BbF, and Fla. High loading of BbF indicates intensive water vehicles transportation and port activities might be the possible reason for BbF contamination [10]. Specifically, BbF was detected higher at the sites close to the harbor area. Hence, petrogenic and pyrogenic activities caused the pollution of PAHs in BR.

3.3.2. DR

LMWs/HMWs > 1 was found for DR water except for sampling locations DW-6 and DW-7; however, LMWs/HMWs < 1 was detected for sediment representing pyrolytic activities responsible for the contamination (ST-5). LMW PAHs significantly polluted the water matrix of the studied area. Heavy fuel oil (HFO) discharge with effluent from river bank power stations, and diesel and oil spillage from water vehicles might be the possible sources of LMW PAHs. As given in Fig. (c), Ant/(Ant + Phe) < 0.1 was found for water except for sampling sites DW-6 and DW-7, indicating petrogenic sources were a possible reason for pollution. For DW-6 and DW-7, sampling sites were polluted through pyrogenic sources according to LMWs/HMWs < 1 (ST-6) and Ant/(Ant + Phe) > 0.1 . HFO firing for power production, and burning bio-waste for waste management, which is produced by tannery industries, might be the possible sources of pyrogenic Ant and Phe. Fla/(Fla + Pyr) > 0.3 was found for water representing varying impacts of fuel and biomass combustion. Fla/(Fla + Pyr) < 0.3 was detected for sediment except in locations DS-8 and DS-9, petrogenic activities like fuel spillage from water vehicles might initiate pollution (Fig. 3(c)). DS-8 and DS-9 were greatly polluted through pyrogenic activities like air deposition of PAHs from leather waste dumping grounds and power stations. As presented in Fig. 2(d), BaA/(BaA + Chr) > 0.35 was assessed for water sampling sites (DW-2 to DW-7) and sediment sampling sites with an exception at DW-1, DW-8, DW-9, DS-3, and DS-4, indicating the emergence of pollutants from fuel-burning. IcdP/(IcdP + BghiP) > 0.5 was found in most of the water sampling sites and sediment, which suggests variable impacts of biomass and fuel burning in the studied area (Fig. 3(d)).

In water, PC-1 has positive loading of 4–6 ring PAHs (BaA, Chr, BbF, BaP, DahA, BghiP, and IcdP) with 50.4% of the total variance (ST-8). Heavy fuel oil and leather waste combustion may be the possible sources of these HMW PAHs [67]. Additionally, Fl, Phe, Ant, and Fla, were strongly associated with PC-2, having 27.5% of the total variance, indicating the generation of these LMW PAHs from petroleum products. The availability of Fl and Fla suggests rapid port activities in the studied area [45]. Moreover, effluent from the power plant might be another possible source of Fl and Fla. PC-3 contains Nap and Acy with 10.7% of the total variance, which

indicates this was emitted by petrogenic activities [34]. In sediment, Ace, Fl, Phe, Ant, Pyr, Chr, BkF, BaP, and IcdP were positively loaded in PC-1 with 66.4% of the total variance. Loading of 3-ring, 4-ring, 5-ring, and 6-ring compounds in the sediment samples of DR suggests that the studied area was polluted by combustion products (ST-8). Pyr, BaA, Chr, and Fl are markers for coal combustion [70], and BaP for biomass burning [71]. PC-2, with 15.6% of the total variable, contains Nap and DahA, which suggests this was emitted by

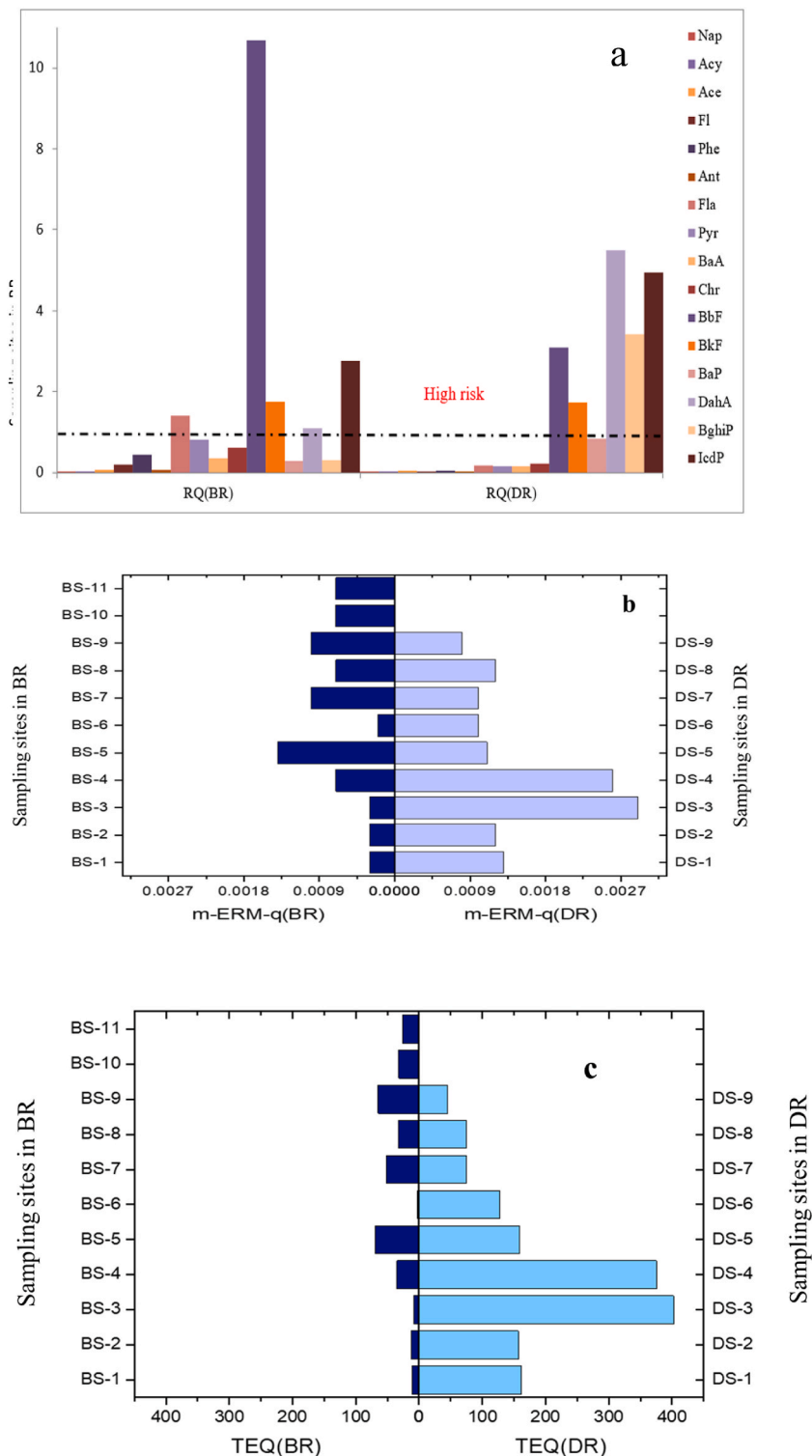


Fig. 4. (a) Risk quotient (RQ) of water sample of BR and DR, (b) *m*-ERM-*q*, and (c) TEQ of sediment sample of BR and DR.

petrogenic activities [34]. PC-3 (6.999%) is positively packed with Acy and BghiP, meaning heavy fuel spillage from water vehicles and biomass burning might be the probable sources.

3.4. Ecological risk assessment

3.4.1. BR

The risk quotient (RQ) to assess the toxicity impact of individual PAHs on aquatic living organisms is given in (Fig. 4(a)). RQ results showed that Fla, BbF, BkF, DahA, and IcdP had posed the highest toxicity to aquatic organisms. Surprisingly, the RQ value of BbF was measured greater than 10, indicating aquatic organisms are in threat of toxicity through BbF exposure. Moreover, Fl, Phe, Pyr, BaA, Chr, BaP, and BghiP are responsible for moderate toxicity to the aquatic organism as RQ values measured between 0.1 and 1. In contrast, the remaining PAHs are not dangerous for aquatic organisms.

Compared with SQGs, the mean value for each PAH in each sampling site was found below TEL and ERL and much lower than PEL and ERM (Table 3). But, the mean concentration of individual PAHs, for 28% Nap, for 82% Acy, for 55% DahA of sample sites exceeded their respective TEL values, suggesting adverse biological effects might occur occasionally. Furthermore, the ERL values for Acy exceed 55% of sampling sites, indicating ecological impairment sometimes. Hence, the sediment ecosystem of BR is less threat to ecological imbalance. In addition, the mean ERM quotient (m -ERM- q) for each sediment site having 12 PAHs was detected between 0.0002 and 0.0014 with a mean value of 0.0007, indicating a less adverse effect on each sampling site in the studied area (Fig. 4(b)). Total toxic benzo [a]pyrene equivalent (TEQ_{carc}) was used to calculate the potential toxicity of sediment for seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA, and IcdP) as shown in Fig. 4(c). \sum TEQ_{carc} values were found between 3.0 and 69.1 ng TEQ/g d.w. with a mean value of 31.3 ng TEQ/g d.w. The contribution has followed the order of BbF > DahA > BaP > BkF > BaA > IcdP > Chr. According to the Canadian soil quality guidelines for protecting the ecosystem and human health, a safe TEQ_{carc} value should be less than 600 μ g/kg [34,71]. Compared to the Canadian Soil Quality Guideline for ecosystem protection and human health, the detected \sum TEQ_{carc} value in each site was found to be much lower than the safe value (600 ng TEQ/g), suggesting the studied area is less prone to biological risk for exposure to the PAHs contamination in sediments.

3.4.2. DR

According to Fig. 4(a), the RQ values of BbF, BkF, DahA, BghiP, and IcdP in water have posed a high risk to aquatic organisms. Furthermore, Fla, Pyr, BaA, Chr, and BaP cause moderate risk to aquatic organisms of DR. In sediment, similar to BR, the average concentration of individual pollutants in each sampling location was compared to TEL, PEL, ERL, and ERM (Table 4). The mean concentrations of Fl, BkF, and DahA were higher than ERL values in 45%, 11.11%, and 22.22% of total sampling sites, respectively, indicating seasonal ecological imbalance through mentioned pollutants. Additionally, mean concentrations of Nap, Acy, Fl, Phe, Ant, Pyr, Chr, BaP, and DahA were detected above TEL at 45%, 22.22%, 45%, 33.33%, 11.11%, 22.22%, 22.22%, 66.66%, and 66.66% of total sampling sites respectively, represents the probability of causing occasional impairment in sediment ecosystem. In contrast, the remaining pollutants pose toxicity rarely. Hence, organisms in sediment were targeted for occasional toxicity in specific sampling sites. Additionally, likewise to BR, m -ERM- q study for DR sediment showed that individual location is in the class of less probable toxic zone (around 11%) (Fig. 4(b)). Total toxic benzo [a]pyrene equivalent (TEQ_{carc}) was used to calculate the potential toxicity of sediment for seven carcinogenic PAHs like BaA, Chr, BbF, BkF, BaP, DahA, and IcdP. \sum TEQ_{carc} values were found between 45.289 and 402.468 ng TEQ/g d.w., with a mean value of 175.066 ng TEQ/g d.w. (Fig. 4(c)). Although BaP contributed the highest percentage (67.969%) of the total carcinogenic effect, \sum TEQ_{carc} values were found to be lower than the threshold value (600 ng TEQ/g).

Table 3

Comparison of individual PAH concentration in each sediment sample site of BR with Sediment quality guideline (SQGs) values for PAHs.

PAHs	BR					Sediment sites						
	ERL	ERM	TEL	PEL	Mean conc. (μ gKg ⁻¹)	Range	<ERL	ERL-ERM	>ERM	<TEL	TEL-PEL	>PEL
Nap	160	2100	34.6	391	27.0	10.4–54.2	All sites	–	–	8 sites	3 sites	–
Acy	16	640	6.71	88.9	23.5	0.1–70.9	5 sites	6 sites	–	2 sites	9 sites	–
Ace	44	500	NA	NA	1.9	0.5–5.6	All sites	–	–	–	–	–
Fl	19	540	21.2	144	4.5	1.4–8.3	All sites	–	–	All sites	–	–
Phe	240	1500	86.7	544	19.4	5.7–39.7	All sites	–	–	All sites	–	–
Ant	85.30	1100	46.9	245	6.1	0.8–30.7	All sites	–	–	All sites	–	–
Fla	600	5100	133	1494	39.7	9.9–59.0	All sites	–	–	All sites	–	–
Pyr	665	2600	153	1398	22.4	4.5–46.9	All sites	–	–	All sites	–	–
BaA	261	1600	74.8	693	14.1	2–34.6	All sites	–	–	All sites	–	–
Chr	384	2800	108	846	15.4	2.2–35.3	All sites	–	–	All sites	–	–
BbF	320	1880	NA	NA	131.0	ND-303.5	All sites	–	–	All sites	–	–
BkF	280	1620	NA	NA	20.0	3.0–45.3	All sites	–	–	–	–	–
BaP	430	1600	88.8	763	5.8	ND-16.4	All sites	–	–	All sites	–	–
DahA	63.40	260	6.22	135	7.7	0.5–20.2	All sites	–	–	5 sites	6 sites	–
BghiP	430	1600	NA	NA	1.7	ND-6.9	All sites	–	–	–	–	–
IcdP	NA	NA	NA	NA	11.5	2.1–31.5	–	–	–	–	–	–
\sum PAHs	4022	44792	1684	16770	351.6	90.7–734.2	All sites	–	–	All sites	–	–

NA= Not applicable; ND= Not detected.

Table 4

Comparison of individual PAH concentration in each sediment sample site of DR with Sediment quality guideline (SQGs) values for PAHs.

PAHs	DR				Sediment sites							
	ERL	ERM	TEL	PEL	Mean conc. (μgKg^{-1})	Range	<ERL	ERL-ERM	>ERM	<TEL	TEL-PEL	>PEL
Nap	160	2100	34.6	391	40.9	11.4–88.34	All sites	–	–	5 sites	4 sites	–
Acy	16	640	6.71	88.9	6.0	2.9–17.3	All sites	–	–	7 sites	2 sites	–
Ace	44	500	NA	NA	11.0	6.1–24.1	All sites	–	–	–	–	–
Fl	19	540	21.2	144	21.1	9.3–38.9	5 sites	4 sites	–	5 sites	4 sites	–
Phe	240	1500	86.7	544	103.1	53.2–223.62	All sites	–	–	6 sites	3 sites	–
Ant	85.30	1100	46.9	245	19.4	6.82–51.22	All sites	–	–	8 sites	1 sites	–
Fla	600	5100	133	1494	16.8	0.3–71.9	All sites	–	–	All sites	–	–
Pyr	665	2600	153	1398	93.6	10.7–213.9	All sites	–	–	7 sites	2 sites	–
BaA	261	1600	74.8	693	33.8	ND-49.6	All sites	–	–	All sites	–	–
Chr	384	2800	108	846	81.3	30.5–161.2	All sites	–	–	7 sites	2 sites	–
BbF	320	1880	NA	NA	62.4	6.5–119.8	All sites	–	–	–	–	–
BkF	280	1620	NA	NA	93.8	22.9–297.4	8 sites	1 site	–	–	–	–
BaP	430	1600	88.8	763	118.9	32.6–297.4	All sites	–	–	3 sites	6 sites	–
DahA	63.40	260	6.22	135	31.7	ND-85.4	7 sites	2 sites	–	3 sites	6 sites	–
BghiP	430	1600	NA	NA	13.7	1.4–56.5	All sites	–	–	–	–	–
IcdP	NA	NA	NA	NA	45.2	10.9–107.3	–	–	–	–	–	–
Σ PAHs	4022	44792	1684	16770	792.9	432.0–1567.1	All sites	–	–	All sites	–	–

NA= Not applicable; ND= Not detected.

3.5. Health risk assessment

3.5.1. BR

In water, the HQ_{dermal} was investigated between $5.1\text{E}-01$ and $9.2\text{E}+00$ (Table 5), where Phe (>1) value indicates the probability of adverse negative effects through the dermal route by Phe, while it is rare for other PAHs. $HQ_{\text{ingestion}}$ varied from $6.2\text{E}-07$ and $9.0\text{E}-04$, suggesting the likelihood of risk to local inhabitants is rare. It is assumed that local people and stakeholders involved in port activities usually use river water for bathing, washing, and other purposes but not for drinking. Furthermore, the assessed HI value indicated the probability of potential risk to local people through all possible exposure routes due to exposure to BR water.

In sediment, HQ_{dermal} and $HQ_{\text{ingestion}}$ were measured lower than 1, suggesting that the probability of adverse effects is rare through individual routes; however, $HI > 1$ indicates that combined exposure routes of PAHs may result in potential risk to inhabitants (Table 5). As represented in Table 5, CR_{dermal} for carcinogenic PAHs in the water samples was investigated between $6.3\text{E}-04$ and $1.9\text{E}-01$, indicating they might pose a moderate to high adverse risk to the people as they use river water in their everyday life. Only Chr poses a moderate risk. Conversely, the cancer risk to local people through the oral route ($CR_{\text{ingestion}}$) was less compared to the dermal pathway, with a value ranging from $9.1\text{E}-09$ to $2.8\text{E}-06$ (Table 5). Therefore, the CR_{total} value of the water sample indicates a potential cancer risk to local people who are involved in port activities. In sediment, obtained $CR_{\text{ingestion}}$ values varied from $6.6\text{E}-05$ to $5.6\text{E}-02$, representing people are at moderate to high risk of cancer (Table 5). Additionally, among all carcinogenic PAHs, BbF, BaP, and DahA showed a high level of carcinogenicity through the oral pathway. CR_{dermal} value was found to be lower than 10^{-6} . Although local people are safe from cancer risk through the dermal exposure route, CR_{total} ($1.3\text{E}-01$) indicates a high risk of carcinogenicity through the combined exposure effect.

3.5.2. DR

According to Table 6, HQ_{dermal} value ($2.01\text{E}-01$ to $2.69\text{E}+00$) for DR water indicates the possibility of non-carcinogenic risk to local people due to direct exposure to river water in terms of washing and bathing activities. Moreover, $HQ_{\text{ingestion}}$ was found to vary from $9.66\text{E}-07$ to $1.40\text{E}-04$, representing local people are at low to moderate non-carcinogenic risk as river water as they do not consume directly. Hence, local people are at risk due to exposure to non-carcinogenic PAHs through the combined routes, as the HI value was higher than 1. In sediment, Phe and BghiP were investigated greater than 1 for $HQ_{\text{ingestion}}$, indicating these pollutants might be responsible for the adverse negative effects on humans; in contrast, HQ_{dermal} values indicate inhabitants are at less risk through the dermal pathway. Therefore, HI values represent the probability of non-carcinogenic risk through combined approaches.

In water samples, local people are at moderate to high cancer risk through dermal exposure pathways by carcinogenic PAHs as CR_{dermal} was detected greater than 10^{-4} ($2.2\text{E}-04$ and $9.7\text{E}-01$) (Table 6). However, $CR_{\text{ingestion}}$ was found below 10^{-6} ($3.2\text{E}-09$ to $3.7\text{E}-06$), indicating carcinogenic PAHs have no risk to local inhabitants through the oral pathway as they consume river water directly. In the sediment samples, the $CR_{\text{ingestion}}$ was responsible for posing a moderate to high cancer risk to local people with a value between $3.5\text{E}-04$ and $5.1\text{E}-01$ (Table 6). CR_{dermal} was estimated between $1.6\text{E}-09$ and $2.4\text{E}-06$, indicating low carcinogenic risk. Hence, the CR_{total} value for water ($2.5\text{E}+00$) and sediment ($7.7\text{E}-01$) suggests that local inhabitants are at high carcinogenic risk and demand close monitoring.

4. Conclusion

This study assessed PAH levels for water and surface sediment in two major rivers (Buriganga and Dhaleshwari) of Dhaka, Bangladesh. For both rivers, investigated PAH levels were higher in the sediment samples compared to river water; moreover, LMW

Table 5
HI and CR assessment for BR water and sediment.

PAHs	Water			Sediment			Water			Sediment		
	HQ _{dermal}	HQ _{ingestion}	HI	HQ _{dermal}	HQ _{ingestion}	HI	CR _{dermal}	CR _{ingestion}	CR _{total}	CR _{dermal}	CR _{ingestion}	CR _{total}
Nap	5.1E-01	3.5E-04	5.1E-01	3.7E-06	7.9E-01	7.9E-01	–	–	–	–	–	–
Acy	–	3.8E-05	3.8E-05	3.2E-06	6.9E-01	6.9E-01	–	–	–	–	–	–
Acp	–	3.5E-05	3.5E-05	8.8E-08	1.9E-02	1.9E-02	–	–	–	–	–	–
Fl	–	1.7E-04	1.7E-04	3.1E-07	6.6E-02	6.6E-02	–	–	–	–	–	–
Phe	9.3E+00	9.0E-04	9.3E+00	1.8E-06	3.8E-01	3.8E-01	–	–	–	–	–	–
Ant	–	1.6E-05	1.6E-05	5.6E-08	1.2E-02	1.2E-02	–	–	–	–	–	–
Fla	–	3.8E-05	3.8E-05	2.7E-07	5.8E-02	5.8E-02	–	–	–	–	–	–
Pyr	–	2.9E-04	2.9E-04	2.1E-06	4.4E-01	4.4E-01	–	–	–	–	–	–
BaA	–	–	–	–	–	–	3.7E-02	5.4E-07	3.71E-02	2.8E-08	6.0E-03	6.0E-03
Chr	–	–	–	–	–	–	6.3E-04	9.1E-09	6.26E-04	3.1E-10	6.6E-05	6.6E-05
BbF	–	–	–	–	–	–	3.9E-01	2.8E-06	3.95E-01	2.6E-07	5.6E-02	5.6E-02
BkF	–	–	–	–	–	–	–	4.5E-08	4.53E-08	4.0E-09	8.6E-04	8.6E-04
BaP	–	–	–	–	–	–	1.8E-01	1.3E-06	1.84E-01	1.2E-07	2.5E-02	2.5E-02
DahA	–	–	–	–	–	–	1.9E-01	7.5E-07	1.90E-01	1.5E-07	3.3E-02	3.3E-02
BghiP	–	6.2E-07	6.2E-07	1.1E-07	2.45E-02	2.45E-02	6.2E-02	1.8E-07	6.20E-02	3.3E-08	7.1E-03	7.1E-03
IcdP	–	–	–	–	–	–	3.1E-02	1.8E-07	3.06E-02	2.3E-08	4.9E-03	4.9E-03

Table 6
HI and CR assessment for DR water and sediment.

PAHs	Water			Sediment			Water			Sediment		
	HQ _{dermal}	HQ _{ingestion}	HI	HQ _{dermal}	HQ _{ingestion}	HI	CR _{dermal}	CR _{ingestion}	CR _{total}	CR _{dermal}	CR _{ingestion}	CR _{total}
Nap	2.0E-01	1.4E-04	2.0E-01	5.6E-06	1.2E+00	1.2E+00	–	–	–	–	–	–
Acy		1.4E-05	1.4E-05	8.3E-07	1.8E-01	1.8E-01	–	–	–	–	–	–
Acp		2.4E-05	2.4E-05	5.1E-07	1.1E-01	1.1E-01	–	–	–	–	–	–
Fl		1.9E-05	1.9E-05	1.5E-06	3.1E-01	3.1E-01	–	–	–	–	–	–
Phe	1.0E+00	1.0E-04	1.0E+00	9.4E-06	2.1E+00	2.1E+00	–	–	–	–	–	–
Ant		9.7E-07	9.7E-07	1.8E-07	3.8E-02	3.8E-02	–	–	–	–	–	–
Fla		4.7E-06	4.7E-06	1.2E-07	2.56E-02	2.5E-02	–	–	–	–	–	–
Pyr		5.7E-05	5.7E-05	8.6E-06	1.8E+00	1.8E+00	–	–	–	–	–	–
BaA	–	–	–	–	–	–	1.5E-02	2.2E-07	1.5E-02	6.8E-08	1.5E-02	1.5E-02
Chr	–	–	–	–	–	–	2.2E-04	3.2E-09	2.2E-04	1.6E-09	3.5E-04	3.5E-04
BbF	–	–	–	–	–	–	1.2E-01	8.0E-07	1.2E-01	1.3E-07	2.7E-02	2.7E-02
BkF	–	–	–	–	–	–	–	4.5E-08	4.5E-08	1.9E-08	4.0E-03	4.0E-03
BaP	–	–	–	–	–	–	5.1E-01	3.7E-06	5.1E-01	2.4E-06	5.1E-01	5.1E-01
DahA	–	–	–	–	–	–	9.7E-01	3.7E-06	9.7E-01	6.4E-07	1.4E-01	1.4E-01
BghiP	2.7E+00	7.9E-06	2.7E+00	9.4E-07	2.1E-01	2.0E-01	7.9E-01	2.3E-06	7.9E-01	2.8E-07	5.9E-02	5.9E-02
IcdP	–	–	–	–	–	–	5.6E-02	3.3E-07	5.6E-02	9.1E-08	1.9E-02	1.9E-02

PAHs were found abundant in river water, while HMW PAHs were detected as dominant in river Sediment. PAHs concentration in the sediment of DR was assessed two-fold higher than BR sediment; whereas, PAHs concentration in the water of BR was detected five times higher than DR. Source identification study confirmed that pyrolytic activities such as burning of municipal waste and vehicle emissions due to incomplete combustion of fuel, burning of heavy oil in power station, and petrogenic activities for instance spillage of fuel from water vehicles and improper management of fuel in the power station are major sources for river pollution. Certain PAHs like BbF, BkF, DahA, and IcdP pose high toxicity to aquatic living organisms in both river ecosystems. The mean concentrations of certain PAHs (Nap, Acy, Fl, BkF, Phe, Ant, Pyr, Chr, BaP, and DahA), were detected as greater than ERL and/or TEL (but lower than ERM and/or PEL), they might pose adverse ecological effect on aquatic organisms occasionally. m -ERM-q and \sum TEQcarc values of each sampling site were found to be lower than the threshold value, suggesting probably of ecological risk and potential bio-toxicity risk were quite low at each sampling site. Detected HI and CR total indicates that local people are at high health risk. A more comprehensive study is required to determine the levels of PAH exposure to the local people, especially the dermal and oral pathways.

Ethics declarations

This article does not involve any ethical issues, so the ethics approval is not required for this paper.

Consent to participate and publish

All authors read and approved the final manuscript. All authors agree that the paper is not being submitted elsewhere and approves to publish.

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Data availability statement

All the relevant data are provided with the supplementary materials

Author's contribution

Aynun Nahar- Conceived and designed the experiments; performed the experiments; analyzed and interpreted the data; wrote the paper.

Md. Ahedul Akbor- Conceived and designed the experiments; analyzed and interpreted the data; fund management.

Shudeepta Sarker- Conceived and designed the experiments; performed the experiments; analyzed and interpreted the data; wrote the paper.

Md. Abu Bakar Siddique- Conceived and designed the experiments; contributed reagents, materials, analysis tools or data.

Md. Aftab Ali Shaikh- Conceived and designed the experiments; contributed reagents, materials, analysis tools or data.

Nushrat Jahan Chowdhury- Analyzed and interpreted the data; wrote the paper.

Shamim Ahmed- Contributed reagents, materials, analysis tools or data.

Mehedi Hasan - Analyzed and interpreted the data; wrote the paper.

Shahnaz Sultana- Analyzed and interpreted the data; wrote the paper.

N.B. Aynun Nahar and Shudeepta Sarker did equal contribution.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.heliyon.2023.e18465>.

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