



Distribution, Source Apportionment, and Risk Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in the Surficial Sediments from the Coastal Areas of Bangladesh

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Abstract

The surficial sediments were collected in winter and summer (2015) from the coastal areas of Bangladesh and analyzed for 16 USEPA priority polycyclic aromatic hydrocarbons (PAHs). The total concentration of PAHs (Σ PAHs) were 349.8–11,058.8 and 199.9–17,089.1 ng/g dry weight (dw) in winter and summer, respectively. Sediments from the areas with recent urbanization and industrialization (Chittagong, Cox's Bazar, and Sundarbans) were more contaminated with PAHs than the unindustrialized area (Meghna Estuary). The concentrations of Σ PAHs were slightly higher in summer than those in winter, but the seasonal variations were not statistically significant ($p > 0.05$). Molecular ratios suggested mixed sources of PAHs in the Bangladeshi coastal areas with a slight imposition of pyrolytic inputs closely related to shipping and fishing activities as well as industrial and municipal sewage discharge. According to ecological risk assessment, the measured levels of sedimentary PAHs exceeded some of the existing national and international environmental quality guidelines/standards, and thus might cause acute biological damage in the studied areas of the Bay of Bengal coast of Bangladesh.

Polycyclic aromatic hydrocarbons (PAHs) are common organic pollutants in soil, water, and sediments worldwide (Suess 1976). PAHs are now ubiquitous in the environment due to their persistence characteristics, long range transport, bioaccumulation, and known to be toxic, carcinogenic, and mutagenic (Barrie et al. 1992; Jones and de Voogt 1999). PAHs are generally formed by natural processes and anthropogenic activities and introduced into the environments

through various ways (Suess 1976; Lee et al. 1981; Barrie et al. 1992). PAHs are mainly enter the environment from two sources: petrogenic source, which is composed by PAHs associated with spills of crude and refined oil, and pyrogenic source, which includes compounds formed by fossil fuel combustion, waste incineration, biomass burning, and asphalt production (Suess 1976; Lee et al. 1981; Barrie et al. 1992). PAHs are widely distributed in marine aquatic environments, specially estuaries, coastal areas, and sometime even in deep sea due to anthropogenic processes and their comparatively long half-life (Neff 1979; Lee et al. 1981; Barrie et al. 1992; Zhang et al. 2016). PAHs finally find their ways into the marine environment particularly in Bangladesh through several courses, including discharge of industrial and municipal sewage, marine transport, shipping and fishing activities, ship building and ship breaking activities, terrestrial surface and riverine runoff, atmospheric deposition, and petroleum spills, etc. Due to their hydrophobic properties, PAHs in the water body tend to be absorbed rapidly by suspended particulate matter and to be subsequently deposited on sediments, which can be generally regarded as a sink for PAHs (Lee et al. 1981). In addition, the organic carbon plays an important role in sorption of PAHs in sediment particles (Rockne et al. 2002). However, source profiles and

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spatial distributions of PAHs in the environment may vary seasonally (Deshmukh et al. 2013).

Based on the evidences of high toxicological risk of PAHs, 16 selected PAHs have been listed as priority pollutants by the United States Environmental Protection Agency (USEPA), of which seven are potentially carcinogen according to the International Agency for Research on Cancer (IARC). The toxicity of PAHs is related directly to their composition and concentrations in the sediments (Johnston et al. 2015). In addition, PAHs are also readily taken up and bioaccumulated by marine organisms and affect negatively not only the growth and reproduction of native wildlife species but also human health via consumption of contaminated seafood (Gu et al. 2013; Sun et al. 2016). Therefore, PAHs have always been of a great concern and need to monitor their contamination levels, as well as their potential toxicity in the environment.

Bangladesh is an agricultural country that has an irregular 580-km-long deltaic marshy coastline, which is divided by many rivers and streams that enter into the Bay of Bengal. The environmental and ecological integrity of the coastal areas of Bangladesh suffers from a number of anthropogenic activities, such as the development of industrial hubs, rapid human settlement, tourism and transportation, dumping of e-waste, widespread ship breaking and port activities, excessive operation of mechanized boats, deforestation, and intensive agriculture and aquaculture activities, discharges of untreated and semitreated land-based sewage, and effluents from various large and small local industries. A substantial amount of contaminants, such as PAHs, could be produced from these anthropogenic activities, which can accumulate in the coastal or marine food chains. Therefore, monitoring and assessing the level of PAH pollution in the coastal areas of Bangladesh is an urgent need.

The environmental PAHs, their distribution, sources, and environmental risk to ecological systems, including humans, have been extensively studied in the coastal regions worldwide (Wang et al. 2001; Horii et al. 2009; Yim et al. 2014; Sun et al. 2016; Zheng et al. 2016; Li et al. 2016; Goswami et al. 2016). However, in Bangladesh, there are very few studies monitored the concentrations of PAHs in aquatic environments (Zuloaga et al. 2013; Nøst et al. 2015). To the best on our knowledge, no comprehensive studies have been conducted in the coastal sediments of Bangladesh. Therefore, the present study was initiated to investigate the contamination levels of PAHs in the coastal sediments of Bangladesh. In addition, spatiotemporal distribution, possible sources, and potential ecological impacts of PAHs also were evaluated in this study.

Materials and Methods

Study Area and Collection of Samples

To explore the influence of the potential pollution sources, we investigated four coastal sites with 14 sampling locations in the southeast and southwest part of the Bay of Bengal coast of Bangladesh. These sampling sites located in Cox's Bazar, Chittagong, Meghna Estuary, and Sundarbans are shown in Fig. 1. An elaborative description of the study area is given in the supplementary information (SI). Please refer to Table S1 in the SI for the coordinates and IDs of sampling location.

A total of 28 sediment samples were collected in winter (January–February) and summer (August–September) 2015. Sampling was performed during low tide. Approximately 200 g of surface sediment samples (top 0–5 cm) were collected from each location by a portable Ekman grab sampler. Samples were kept in polyethylene (PE) Ziplock® bags and transported in ice-filled airtight insulating box to the laboratory of Fisheries Department of Dhaka University. Freeze-dried sediment samples were grounded and stored in PP bottles – 20 °C. Upon the permission from the Yokohama Plant Protection Station, all the pre-processed samples were brought to the Yokohama National University, Japan, for further chemical analysis.

Chemicals and Reagents

The 16 priority PAH compounds of the United States Environmental Protection Agency (USEPA), namely, Naphthalene (Nap), Acenaphthylene (Acel), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)-fluoranthene (BkF), Benzo(a)pyrene (BaP), Dibenz(a,h)anthracene (DahA), Benzo(g,h,i)perylene (BghiP), and Indeno(1,2,3-cd)pyrene (IP), were analyzed. Native calibration standards of a complete set of all 16 EPA PAH isomers (Z-013N-SET, Polycyclic Aromatic Hydrocarbon Kit 10MGx16) and two isotopically labeled internal standards (Acenaphthene-D10 (Ace-D10) and Benzo[a]pyrene-D12 (BaP-D12)) were purchased from AccuStandard (New Haven, CT, USA). Supelclean™ ENVI-18 solid phase extraction (SPE) cartridges (12 mL, 2 g) were purchased from SUPELCO® (PA, USA). All of the Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) extraction kits were obtained from Agilent Technologies (Santa Clara, CA). All solvents (*n*-hexane, acetone, methanol, and dichloromethane) used for sample processing and analysis were analytical grade and

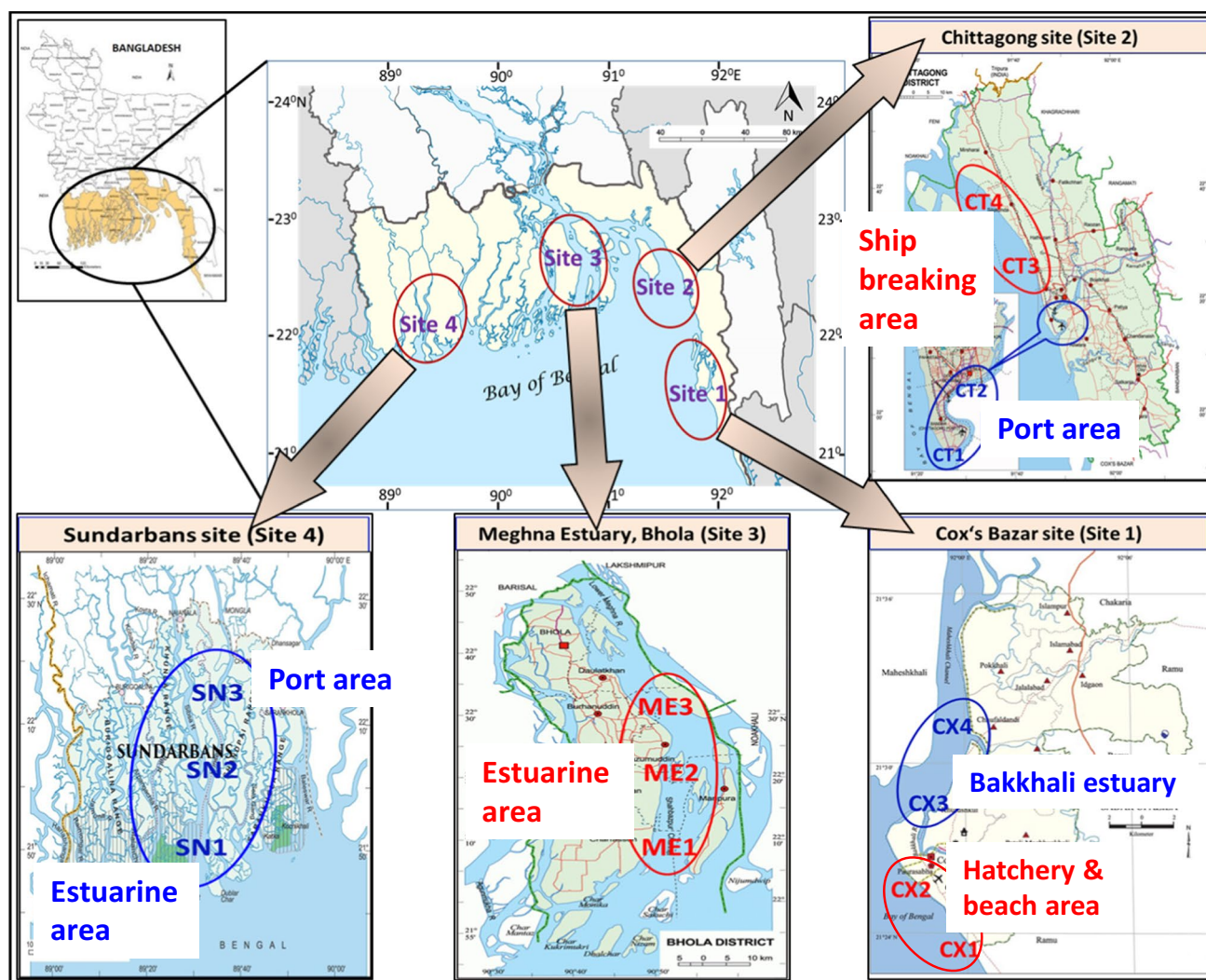


Fig. 1 Maps showing 4 sampling sites with 14 sampling locations in the coastal areas of Bangladesh

purchased from Wako Chemical (Osaka, Japan). Milli-Q ($> 18.2 \text{ M}\Omega$) water generated from an ultrapure water purification system (Millipore, Billerica, MA) was used throughout the experiment. Filter membranes ($0.45 \mu\text{m}$, 47 mm i.d.) were purchased from ADVANTEC® (Tokyo, Japan).

Sample Pretreatment

After homogenizing with a silica mortar and pestle, the sediment samples were sieved (2-mm mesh) to remove debris and coarse fraction of greater than 2 mm in size class (IAEA 2003). After that, they were extracted by a Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) method validated by Salem et al. (2016) following some modifications. Briefly, 5 g of sediment was weighed into a 50-mL PP tube. After adding 5 mL of ultrapure water, the tube was shaken manually. Then, the sample was spiked with $100 \mu\text{L}$

of 500 ng/mL of Ace-D₁₀ and BaP-D₁₂ as internal standards (IS) for quantification. Fifteen milliliters of extraction solvent (hexane:acetone:dichloromethane $\approx 1:1:1$, v/v/v) and two ceramic bars (Agilent p/n 5982–9313) were added to facilitate the extraction and the tube was hand shaken vigorously for 5 min . Afterwards, the QuEChERS salts (4 g of magnesium sulfate (MgSO_4), 1 g of sodium chloride (NaCl), 1 g of trisodium citrate dihydrate ($\text{Na}_3\text{C}_6\text{H}_5\text{O}_7$), and 0.5 g of disodium hydrogen citrate sesquihydrate ($\text{C}_6\text{H}_8\text{Na}_2\text{O}_8$); Agilent p/n 5982–6650) were added, and the tube was immediately shaken for 1 min to resist the agglomeration of salts followed by ultrasonic agitation for 20 min . Finally, the tube was centrifuged for 3 min at 3500 rpm , and 10 mL of the supernatant were transferred to a QuEChERS dispersive solid-phase extraction (d-SPE) clean-up tube (15 mL) containing 0.9 g of anhydrous magnesium sulfate, 0.15 g of primary secondary amine (PSA), and 0.15 g of C18EC (Agilent p/n 5982–5156). One ceramic bar (Agilent p/n 5982–9312)

was added, and the tube was shaken vigorously by hand for 1 min followed by centrifugation for 3 min at 3500 rpm. The extracts were then treated with activated copper for desulfurization and transferred into a glass test tube and then were evaporated to near dryness under a gentle stream of high-purity nitrogen. The residue was finally redissolved in 1 mL of *n*-hexane and stored at $-20\text{ }^{\circ}\text{C}$ until GC–MS/MS analysis.

Instrumental Analysis

Gas chromatograph–tandem mass spectrometry (GC–MS/MS) analysis was performed using an Agilent 7890A GC, coupled with an Agilent 7000C triple-quadrupole MS. A computer with MassHunter software (version B.05.00412) was used for data acquisition and processing (Agilent Technologies, Palo Alto, CA). Chromatographic separation was achieved on an DB-5MS capillary column ($30\text{ m}\times 0.25\text{ mm}$ ID, $0.25\text{-}\mu\text{m}$ film thickness; Agilent p/n 122-5532) using Helium as a carrier gas at a flow rate of 1.2 mL/min . The GC oven temperature was initiated at $70\text{ }^{\circ}\text{C}$ for 1 min, increased to $300\text{ }^{\circ}\text{C}$ for 4 min at $10\text{ }^{\circ}\text{C min}^{-1}$, and finally held at $310\text{ }^{\circ}\text{C}$ for 2 min (total run time 31 min). The injection volume was set to $1\text{ }\mu\text{L}$ in splitless mode. Mass spectrometry was operated in multiple reactions monitoring (MRM) mode with a gain factor of 10. Electron impact (EI) ionization voltage was 70 eV . Nitrogen and helium were used as collision gas and quench gas in the collision cell at constant flows of 1.5 and 2.25 mL/min , respectively. Temperatures of transfer line, ionization source and triple quadrupole mass analyzer were $320\text{ }^{\circ}\text{C}$, $300\text{ }^{\circ}\text{C}$, and $150\text{ }^{\circ}\text{C}$, respectively. A solvent delay was set at 3 min. Both the first (Q1) and the third quadrupole (Q3) were operated at width resolution mode. Before analysis, MS/MS was auto-tuned with perfluorotributylamine. GC–MS/MS conditions and/or parameters for the analysis of PAHs are shown in Table S2. The analytes were identified by comparison of the retention times of the peaks detected in samples with the peaks obtained from a GC–MS/MS run using a standard solution containing a mixture of all 16 PAHs. For quantification, Ace-D₁₀ was used for Nap, Ace1, Ace, Flu, Phe, and Ant, and BaP-D₁₂ was used for Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP. The quantification of the PAHs was based on the area obtained for each analyte in the samples, the mass/area ratio obtained for the internal standard, the response factor obtained from the calibration curve, and the original sample weight. Concentrations of PAHs are given in nano gram per gram dry weight (ng/g dw).

Quality Assurance and Quality Control

Strict quality control procedure was maintained during the experiments. The containers and equipment used during the

whole procedure were precleaned with methanol followed by acetone. A (signal to noise) S/N ratio ≥ 3 was used to determine the limit of detection (LOD) for each analyte and an S/N ratio of 10:1 was defined as the limit of quantitation (LOQ). The LOQs were in the range of $0.06\text{--}0.82\text{ ng/g dw}$. The instrumental blanks (solvent without internal standard) and procedural blanks (Milli-Q water spiked with internal standards) analyzed with every batch of samples gave S/N values < 10 ($< \text{LOQs}$). To validate the accuracy of the methods, matrix spike recovery ($n=3$) was determined by spiking the target compounds into the samples at 10 ng/g dw , followed by similar extraction and analysis procedure as described in earlier sections. The mean recoveries of PAHs spiked into the water samples were $70\text{--}112\%$. The detailed QA/QC data are given in Table S3.

Data Analysis

The IBM SPSS (Version 23.0, IBM Corp., NY, USA) and XLSTAT (Version 2016.02.28451, Addinsoft, NY, USA) software were used for statistical analyses. Before analysis, the significance level was set at $p=0.05$. The normality of the data set was tested by a statistical distribution test called *P–P* plots. The differences among the concentrations of PAHs in the Bangladeshi coastal areas and seasonal variations were tested by one-way ANOVA. The spatial variations of PAHs in surface sediments were shown by using MapViewer™ software (Version 8, Golden Software Inc., CO, USA).

Results and Discussion

Concentration of PAHs in Surface Sediment

The target 16 PAHs were detected in all the samples between the two seasons. The concentrations of PAHs are summarized in Table 1 and illustrated in Fig. 2, whereas the detailed data are presented in Tables S4 and S5. The total concentrations of the ΣPAHs (sum of 16 PAHs) in the sediment samples ranged from 349.8 to $11,058.8\text{ ng/g dw}$ (mean: 4571.0 ng/g dw ; median: 4515.0 ng/g dw) in winter, and from 199.9 to $17,089.1\text{ ng/g dw}$ (mean: 5729.0 ng/g dw ; median: 4108.7 ng/g dw) in summer (Table 1).

In this study, we identified the dominant PAH compounds in the sediment both by occurrence and abundance by combining the detection rate and mean relative concentration of each compound across samples. Regardless of season, top four PAH compounds (Flt, Pyr, Chr, and Phe) were comprising up to $30\text{--}90\%$ (mean: 65%) of ΣPAHs by sites and highly correlated with ΣPAHs (Spearman rank correlation; $r=0.98$; $p<0.05$). In particular, Flt was the most abundant PAH compound in winter with a contribution of $12\text{--}43\%$

Table 1 Concentrations of PAHs in the surface sediments from the coastal areas of Bangladesh and standard pollution criteria of PAH components for sediment matrix (ng/g dw)

PAHs	Winter				Summer				Guideline values	
	Range	Mean	Median	<i>df</i> ^a	Range	Mean	Median	<i>df</i>	ERL ^d	ERM ^d
Nap	11.5–202.2	69.3	54.1	100	18.5–1092.3	330.5	262.2	100	160	2100
Acel	4.2–246.8	50.0	23.2	100	7.8–420.53	127.9	78.1	100	44	640
Ace	0.5–106.7	26.8	23.05	100	2.1–816.7	170.2	76.2	100	16	500
Flu	1.2–126.1	52.3	45.0	100	0.6–545.6	97.7	39.1	100	19	540
Phe	17.3–1439.1	508.0	407.6	100	14.7–1538.3	482.6	502.7	100	240	1500
Ant	4.7–334.2	91.6	32.2	100	11.3–564.3	103.0	58.1	100	853	1100
Flt	59.9–3479.2	1118.2	950.6	100	30.6–3664.7	1135.2	816.9	100	600	5100
Pyr	46.4–2619.4	1195.8	1162.1	100	4.6–6190.3	1828.1	1125.9	100	665	2600
BaA	13.6–483.2	154.3	100.8	100	14.7–473.4	173.4	105.4	100	261	1600
Chr	28.9–1973.9	565.3	421.5	100	10.8–2907.9	565.9	193.5	100	384	2800
BbF	7.9–1108.6	308.9	262.1	100	4.6–657.2	147.4	54.0	100	NA	NA
BkF	1.8–347.3	110.6	109.0	100	4.7–584.9	159.4	75.2	100	NA	NA
BaP	4.4–373.5	132.1	110.3	100	17.8–352.7	102.9	65.2	100	430	1600
DahA	0.6–314.4	54.2	23.1	100	6.4–295	74.2	41.9	100	63.4	260
BghiP	3.4–271	77.4	33.4	100	2.6–207.8	60.0	34.8	100	NA	NA
IP	2.8–185.2	56.0	34.3	100	3.0–1475.5	170.5	20.2	100	NA	NA
Σ C-PAHs ^b	84.5–3638.2	1381.4	1430.7	100	76.9–4908.8	1393.7	643.6	100	NA	NA
Σ PAHs ^c	349.8–11,058.8	4571.0	4515.0	100	199.9–17,089.1	5729.0	4108.7	100	4000	44,792

NA not available

^aDetection frequency (%); *n* = 14 for each season

^bSum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA, and IP)

^cSum of 16 USEPA PAHs

^dThe effects range low (ERL) and the effects range median (ERM) (Long et al. 1995)

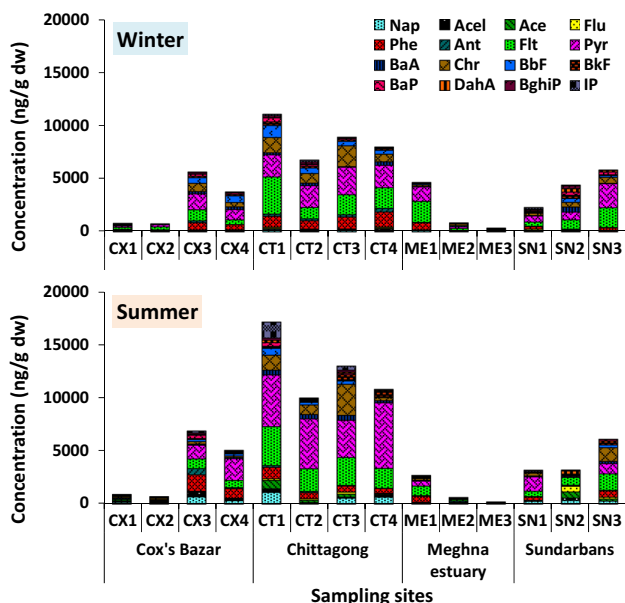


Fig. 2 Concentrations of PAHs in the surface sediments in the Bangladeshi coastal areas in winter and summer

(mean: 24%) of Σ PAHs, whereas Pyr was the most abundant PAH compound in summer contributing up to 2–57% (mean: 22%) of Σ PAHs. However, the dominance of these PAH compounds was also reported in the coastal and estuarine sediments in several studies (Poza et al. 2011; Gu et al. 2016; Goswami et al. 2016).

A comparison of total PAH concentrations in surface sediments of coastal and estuarine regions around the world are shown in Table 2. In general, the Σ PAHs concentrations in the present study were comparable or lower than those measured in the Kaohsiung Harbor (Taiwan), Boston Harbor (USA), Santander Bay (Northern Spain), Genoa-Voltri Harbor (Italy), Norwegian Harbor (Norway), and Estuarine and riverine area (Chennai, India). However, the levels were higher than those reported in the China coastal zones, Jinhae Bay (Korea), Tokyo Bay (Japan), Entire continental shelf (Sweden), Gemlik Bay (Turkey), Mediterranean coastal zone (Egypt), Lenga Estuary (Chile), Bahía Blanca estuary (Argentina), and Klang Strait (Malaysia) (Table 2). Furthermore, the levels of sediment contaminations evaluated by PAH concentrations could be classified into four categories: (a) low, 0–100 ng/g; (b) moderate, 100–1000 ng/g; (c) high,

Table 2 Concentrations of total PAHs in sediments (ng/g dw) from various estuary and coastal regions in the world

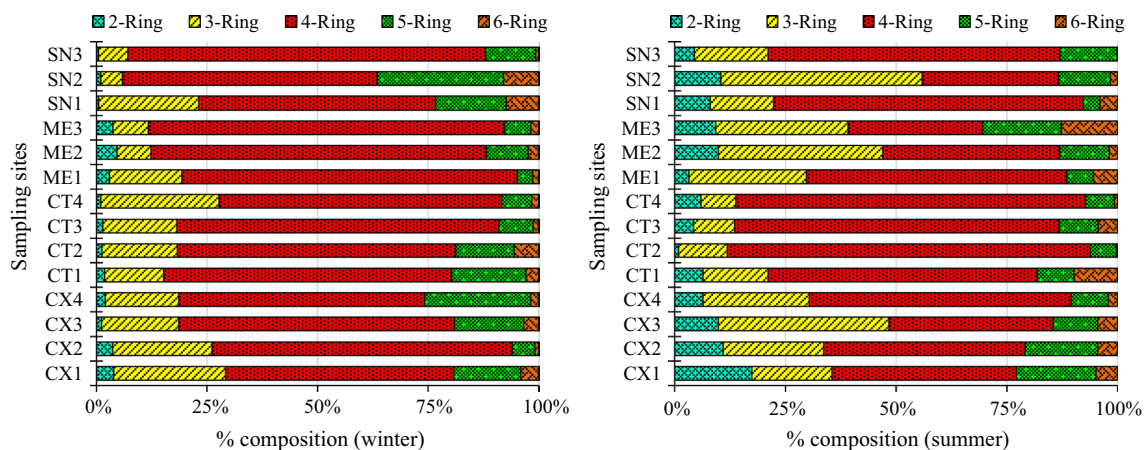
Locations	Sampling year	N ^a	∑PAHs	Pollution levels	References
China coastal zones	2014	16	195.9–4610.2 (W) ^b 98.2–2796.5 (S) ^b	Low to high	Li et al. (2016)
Kaohsiung Harbor, Taiwan	2006	17	34–16,700	Low to very high	Chen et al. (2013)
Jinhae Bay, Korea	2010	16	12.4–2430	Low to high	Yim et al. (2014)
Tokyo Bay, Japan	2004	16	109–1170	Moderate to high	Horii et al. (2009)
Boston Harbor, USA	1999	16	7300–358,000	Very high	Wang et al. (2001)
Entire continental shelf, Sweden	1995–2006	16	120–9600	Moderate to very high	Sánchez-García et al. (2010)
Santander Bay, Northern Spain	– ^c	16	20–25,800	Low to very high	Viguri et al. (2002)
Genoa-Voltri Harbor, Italy	2002	16	4500–20,800	Moderate to very high	Salvo et al. (2005)
Norwegian Harbor, Norway	–	16	2000–76,000	Moderate to very high	Oen et al. (2006)
Gemlik Bay, Turkey	–	14	50.8–13,482	Low to very high	Ünlü and Alpar (2006)
Mediterranean coastal zone, Egypt	–	16	3.51–14,100	Low to very high	Barakat et al. (2011)
Lenga Estuary, Chile	2002	16	290–6118	Moderate to very high	Pozo et al. (2011)
Bahía Blanca estuary, Argentina	2004–2005	18	15–10,260	Low to very high	Arias et al. (2010)
Klang Strait, Malaysia	2011–2012	16	100.3–3446.9	Moderate to high	Sany et al. (2014)
Estuarine and riverine area, Chennai, India	2014	16	13–31,425	Low to very high	Goswami et al. (2016)
Coastal area of Bangladesh	2015	16	349.8–11,058.8 (W) 199.9–17,089.1 (S)	Moderate to very high	This study

^aNumber of PAHs^bW: Winter, S: Summer^cSampling year not mentioned

1000–5000 ng/g; (d) very high, > 5000 ng/g (Baumard et al. 1998). Therefore, based on the global comparison along with the proposed contamination classification, the surface sediment of the Bangladeshi coastal areas might be posed as moderately to heavily polluted by PAHs. More specifically, sediments of the study areas can be characterized as moderate for sites CX1, CX2, ME2, and ME3, high for ME1, SN1, and SN2, and very high for sites CX3, CX4, CT1, CT2, CT3, CT4, and SN3 in terms of PAHs contamination.

PAH Composition in Sediments and Source Characterization

The composition patterns and relative abundance of PAHs by the number of aromatic rings in the Bangladeshi coastal surface sediments in winter and summer are illustrated in Fig. 3. The compositions of PAHs in different sampling sites were almost similar and did not varied significantly between seasons ($p > 0.05$). In particular, 4-ring PAHs were the dominant compounds accounting 52–81% and 30–82%

**Fig. 3** Compositional profiles of PAH compounds in surface sediments taken in winter and summer

of Σ PAHs in winter and summer, respectively, followed by 5-ring PAHs (4–29% in winter and 4–18% in summer). In general, regardless of source and season, the patterns of PAHs contamination on the basis of ring number were in order of 4-ring > 5-ring > 3-ring > 2-ring > 6-ring. The results are consistent with the typical PAHs composition in the surface sediments reported in other studies (Ünlü and Alpar 2006; Sánchez-García et al. 2010; Pozo et al. 2011; Chen et al. 2013; Sany et al. 2014; Goswami et al. 2016; Li et al. 2016). About anthropogenic PAHs, low molecular weight (LMW, 2–3 rings) PAHs have both petrogenic and low-temperature combustion sources, whereas high molecular weight (HMW, 4–6 rings) PAHs have a predominantly high-temperature pyrolytic source (Mai et al. 2003). The HMW PAHs are resistant to degradation and less soluble in water (hydrophobic), and hence they associate with particulate matter that eventually deposit in sediments (Lee et al. 1981; Nagy et al. 2014). On the contrary, LMW PAHs having comparatively higher aqueous solubility tend to retain mainly in the dissolved phase (Lee et al. 1981; Aziz et al. 2014). As a result, the dominance of HMW PAHs have been commonly observed in sediments. In this study, the contribution of HMW PAHs (44–94%) was significantly higher than the LMW PAHs (6–56%). Zhou and Maskou (2003) reported that pyrolysis/combustion of fossil materials yields, such PAH assemblages, which are subsequently introduced into the marine environment by coastal and river runoff. Industrial and domestic wastes/sewerage were also suggested as another important local source.

The identification of PAHs source is important to reveal the transport and fate processes of PAHs in the environment. As a general practice, ratios of various individual PAH concentrations are used to distinguish the possible sources of PAHs (Yunker et al. 2002). Among these molecular ratios, Ant/(Ant + Phe), Flt/(Flt + Pyr), BaA/(BaA + Chr), and IP/(IP + BghiP) are more widely used

(Yunker et al. 2002; Tobiszewski and Namiesnik 2012). Although the use of PAH diagnostic ratios for source characterization has been criticized (Katsoyiannis et al. 2007; Galarneau 2008), it has been used widely and proven to effectively differentiate PAH sources (Jiang et al. 2009; Martins et al. 2010; Xue et al. 2013), and consequently, in the present study, it is utilized as an indicative tool of PAHs source and distribution.

The criteria for PAH ratios as indicators of PAH sources are listed in Table S6 and the plotted isomeric ratios of: Ant/(Ant + Phe) versus Flt/(Flt + Pyr), and BaA/(BaA + Chr) versus IP/(IP + BghiP) used in this study are shown in Fig. 4. The values of Ant/(Ant + Phe) were 0.02–0.83, 86% of which were higher than 0.1; the ratios of Flt/(Flt + Pyr) were less than 0.4 in 14%, 0.4–0.5 in 29%, and greater than 0.5 in 57% of samples (Fig. 4a). The values of BaA/(BaA + Chr) were less than 0.2 in 25%, 0.2–0.35 in 32%, and greater than 0.35 in 43% of samples; the ratios of IP/(IP + BghiP) were less than 0.2 in 4%, 0.2–0.5 in 21%, and higher than 0.5 in 75% of samples (Fig. 4b). The above data demonstrated that the PAHs in sediments from the Bangladeshi coastal area originated from both the petrogenic and pyrogenic sources, including crude petroleum (e.g., gasoline/diesel), petroleum combustion, and combustion of grass, wood, and coal. However, more than 60% of the values calculated from the PAH molecular ratios skewed towards the values for indication of pyrolytic origin.

However, PAH contamination in this area from petrogenic origin should not be ignored as well. Interestingly, it was seen from Fig. 4 that the ratios were more scattered in summer season compared with winter, indicating a considerable input of PAH through atmospheric wet and dry deposition and/or land runoff during times of heavy rainfall, and that are most probably originated from non-point diffuse sources.

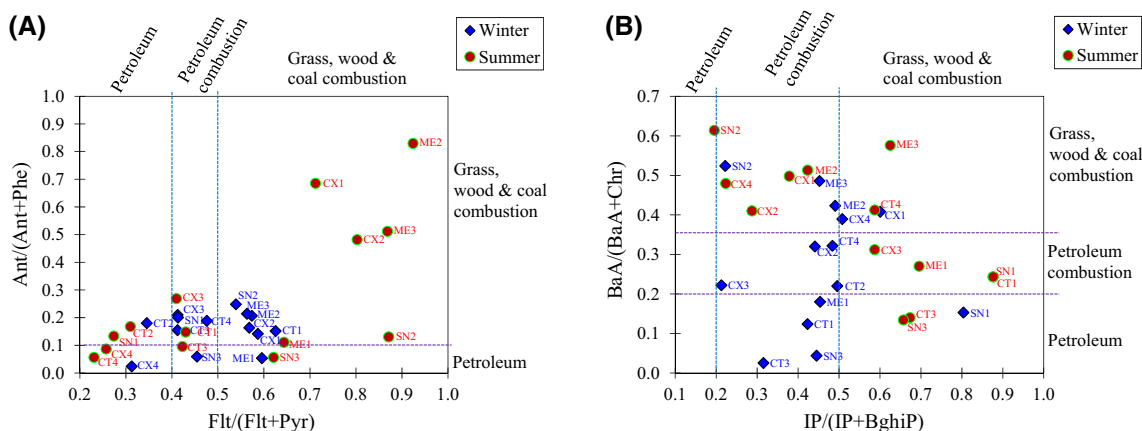


Fig. 4 PAH cross-correlations for the ratios of **a** Ant/(Ant + Phe) and Flt/(Flt + Pyr), **b** BaA/(BaA + Chr) and IP/(IP + BghiP)

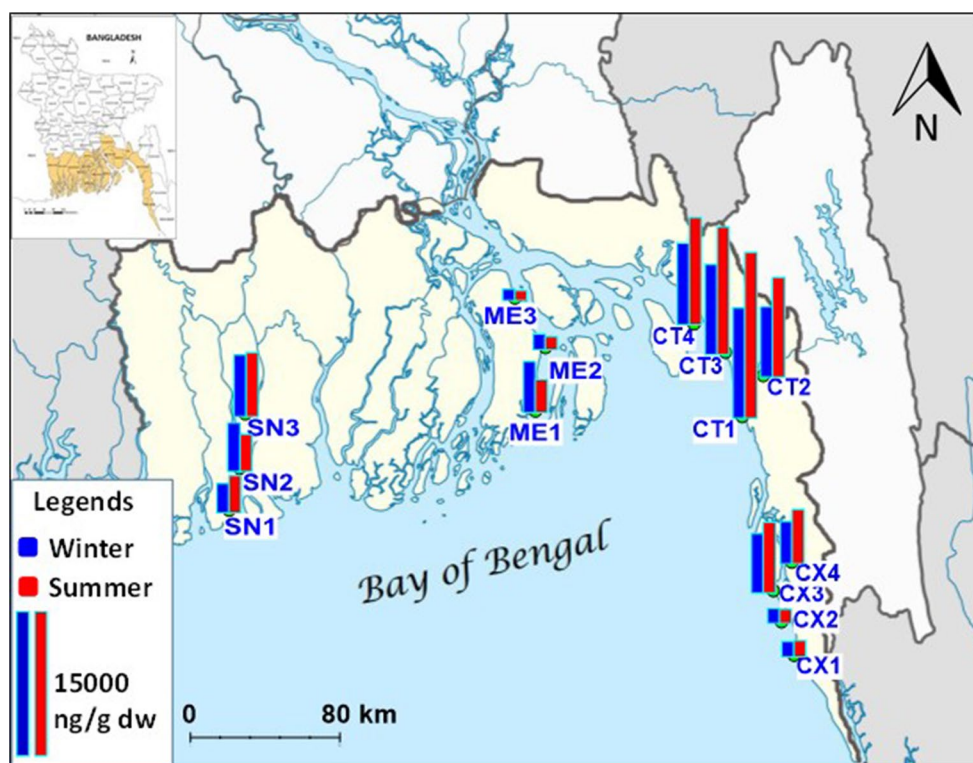
Seasonal and Spatial Distribution of Sedimentary PAHs

The levels of Σ PAHs between the two seasons did not differ significantly ($p > 0.05$). However, seasonal variations to an extent were noticeable depending on the sites. For example, CX1, CX3, CX4, CT1, CT2, CT3, CT4, SN1, and SN2 showed slightly higher concentrations in summer, whereas the rest of the sites exhibited the opposite trends. In summer (wet period), PAHs previously buried in the surface soil of heavily contaminated sites and accumulated in dry weather were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. Abdel-Shafy and Mansour (2016) reported that the concentrations of PAHs in the gas phase increased in summer in tropical regions that could be redeposited through the atmospheric wet and dry deposition. Additionally, increased shipping and fishing activities during summer in the study area might also be another crucial factor for the seasonal variations. On the contrary, elevated levels of PAHs in winter samples were probably due to either co-evaporation of PAHs with water or intense sedimentation from less mixing effects due to comparatively weaker tidal movement or wave action and lower inflow from upstream rivers in the dry season as well. Overall, the pattern of seasonal variation of Σ PAHs concentrations in this study was in agreement with other studies (Zhang et al. 2012; Tian et al. 2014), but contrary to the seasonal variations of Σ PAHs concentrations in sediment from

Chinese coastal zones (Li et al. 2016). These discrepancies might be attributed to the impact of regional hydrological conditions and local pollution sources among the study areas. However, in terms of individual PAHs, concentrations of 12 PAHs (Nap, AceI, Ace, Flu, Phe, Ant, Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP) were found to be higher in summer, whereas the other 4 PAHs (Phe, BbF, BaP, and BghiP) showed the reverse trends, although the differences were statistically significant only for Nap (average of 69.3 and 330.5 ng/g dw in winter and summer, respectively) and Ace (average of 26.8 and 170.2 ng/g dw in winter and summer, respectively) ($p < 0.05$). The seasonal variations of these particular PAHs might be related to its seasonal emissions from land-based or sea-based sources or due to impact of anthropogenic activities during this particular time. This variation might also be influenced by hydrological conditions of the site, the physico-chemical properties of the deposited sediments, absorption–desorption mechanism, degradation of PAH compounds along with their other properties. However, the exact reasons are yet to be investigated in the study area. In general, regardless of sampling sites the concentration of Σ PAHs in summer was slightly higher than that in winter.

The spatial distribution of PAHs in surface sediment samples is shown in Fig. 5. Levels of PAHs in surface sediment differed significantly between the four coastal regions ($p < 0.05$), indicating the PAHs contamination mainly influenced by the local/regional source inputs in the study areas.

Fig. 5 Distribution of total PAHs in the surface sediment of the coastal areas of Bangladesh. Colored area in the inset map represents the coastal areas of Bangladesh



However, the pattern of \sum PAHs distribution between sites were nearly similar in winter and summer. Concentrations of \sum PAHs were higher in the coastal sediments collected from Chittagong area (average of 8676.5 and 12,693.6 ng/g dw in winter and summer, respectively), followed by that from Sundarbans (average of 4171.1 and 4138.9 ng/g dw in winter and summer, respectively) and Cox's Bazar (average of 2734.3 and 3374.5 ng/g dw in winter and summer, respectively) compared with the sediments from Meghna estuary (average of 1946.0 and 1172.5 ng/g dw in winter and summer, respectively), which indicates that the economic development seems to be associated with the amount of PAH emissions in the study area.

In particular, sediment samples collected from the sites adjacent to the seaport and fish landing harbors (e.g., CT1, SN3, CX3) and ship breaking yards (e.g., CT3) showed elevated levels of PAH contamination. Thousands of boats and ships are travelling in the Chittagong port (the largest seaport in Bangladesh) and Mongla port areas for multipurpose operations including export–import of petroleum products. Unintentional or accidental spill of oil during the operation of oil loading and unloading might be a potential source of PAHs (Zhou and Maskaoui 2003). Diesel leakage from ships and boats may have contributed to the high PAH levels in this area and that are in agreement with other studies (Dudhagara et al. 2016; Wang et al. 2016). Ship breaking activities along the coast of Chittagong may contribute to the PAHs pollution in the adjacent areas, because dismantling ships produces various types of inorganic and organic pollutants, including PAHs (Hossain 2006; World Bank 2010; Neşer et al. 2012; Siddiquee et al. 2012; Dudhagara et al. 2016). Nøst et al. (2015) found an elevated PAHs levels in air at sites near the ship breaking activities in Chittagong which might be redeposited from air to the adjacent aquatic environment through wet deposition and air/water exchange processes. Besides, intensive boating and fishing in the areas of fish landing harbors might be potential contributing factors to the PAHs contamination in the coastal environment (Zhou and Maskaoui 2003; Li et al. 2016; Wang et al. 2016). Previous studies have identified that the ship breaking activities, leakage of diesel fuel, crude oil used in ship engines, and coal from ship and heavy transport vehicles and oil seepage from industrial activities lead to high concentrations of total PAHs in the coastal sediments (Dhananjayan et al. 2012; Dudhagara et al. 2016). In addition, establishments of numerous multipurpose industries along the coast line may contribute PAHs contamination in the surface sediments of this area.

The levels of \sum PAHs in sediment taken from the Meghna estuary were significantly lower than that from other areas ($p < 0.05$). It is to be noted that the Meghna estuary is an exclusively unindustrialized area less or not affected by cities, which led to the lower contamination by PAHs.

However, comparatively lower but detected PAHs concentrations in sediments from this site suggest that there are non-point sources of PAHs, such as surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc. Overall, the coastal sediments of the Chittagong area were the most contaminated by PAHs followed by Cox's Bazar and Sundarbans, whereas Meghna Estuary was the least contaminated area.

Impacts of Sediment Properties on PAHs Distribution

The physicochemical properties of sediment (e.g., grain size/texture, organic matter) have been recognized as a major influential factor in determining the distribution and transport of hydrophobic organic compounds (OC) like PAHs in the aquatic environments (Wang et al. 2001; Sánchez-García et al. 2010; Chen and Chen 2011; Chen et al. 2013; Li et al. 2016). Among them total organic carbon (TOC) is crucial for their sorption, a key process in trapping and transporting PAHs in sediments (Oen et al. 2006; Chen and Chen 2011). Within the total organic moieties, black carbon (BC) with the highest sorption capacity due to its condense and aromatic structure has been shown to large extent affect the distribution of OCs in sediments (Accardi-Dey and Gschwend 2002; Lohmann et al. 2005; Wang et al. 2014). The correlations between environmental contaminants might be indicative to find out the similar or multiple pollution sources in a specific study area. To identify such correlations and influence of sediment properties on the distribution of PAHs in the present study area, we performed a Spearman rank correlation analysis and the results are shown in Table S7.

The concentrations of sedimentary TOC (%) and BC (%) were recorded as 0.06–1.28 and 0.06–0.53 in winter and 0.15–0.69 and 0.06–0.28 in summer, respectively (Table S8). No significant variation was observed in sediment physico-chemical properties between the two seasons (t test, $p > 0.05$). However, the correlation results revealed a significant positive association between the \sum PAHs and BC ($r = 0.56$, $p < 0.01$), whereas the correlations between \sum PAHs and TOC were positive but not significant ($r = 0.30$, $p > 0.05$). In terms of individual PAHs, none of them showed significant association with TOC ($p > 0.05$). On the contrary, Nap, Ace, Flu, Phe, Flt, Pyr, Chr, BbF, BkF, and IP were positively ($r = 0.43$ – 0.71) and strongly ($p < 0.01$) correlated with the BC content. Sediment texture is one of the important factors affecting the accumulation profile of PAHs in sediment. In the present study, sediment quality was characterized and we found the texture varied between seasons (Table S8). A significant correlation was found between \sum PAHs and clay particles ($r = 0.50$, $p < 0.001$). It is implied that the fine-grained particles that are enriched with soot

carbon (e.g. BC) play a significant role in the transportation and redistribution of PAHs in the coastal area of Bangladesh.

Ecological Risk Assessment of Sedimentary PAHs

Sediment Potential Toxicity Based on Carcinogenic PAHs (C-PAHs)

IARC (1987) has identified Chr, BaA, BkF, BbF, BaP, DahA, and IP as possible carcinogens. The total concentration of carcinogenic PAHs (\sum C-PAHs) were 84.5–3638.2 ng/g dw (mean: 1381.2 ng/g dw) and 76.9–4908.8 ng/g dw (mean: 1393.7 ng/g dw), accounting 8–57% and 11–48% to the \sum PAHs in winter and summer, respectively (Table 1).

The potential toxicity of sediment was evaluated using the total toxic BaP equivalent (TEQ^{carc}) (Chen and Chen 2011; Chen et al. 2013). The TEQ^{carc} for all C-PAHs was calculated using the following equation:

$$TEQ^{carc} = (C_i \times TEF_i^{carc})$$

where C_i is the C-PAH concentration (ng/g dw) and TEF_i^{carc} (toxic equivalency factors) is the toxic factor of carcinogenic PAHs relative to BaP. Among all known potentially carcinogenic PAHs, BaP is the only PAH for which toxicological data are sufficient to derive a carcinogenic potency factor (Peters et al. 1999). According to the USEPA (1993), TEFs for BaA, CH, BbF, BkF, BaP, DBA, and IP are 0.1, 0.001, 0.1, 0.01, 1, 1, and 0.1, respectively. In this study, the total TEQ^{carc} values of sediment samples varied from 15.2 to 717.7 ng TEQ/g dw, with a mean value of 239.9 ng TEQ/g dw in winter and 27.8 to 835.5 ng TEQ/g dw, with a mean value of 228.4 ng TEQ/g dw in summer. Regardless of seasons, the higher total TEQ^{carc} values were found at sites CT1, CT3, CX3, SN2, and SN3 near the port area and industrial zones (Fig. 6). It suggests that the shipping and fishing activities in the port area along with other industrial activities play an important role in the leaching of C-PAHs into the environment. The TEQ^{carc} found in the Bangladeshi coastal sediments is comparable to that in sediments from Meiliang Bay, China (Qiao et al. 2006), Barents Sea, Russia

(Savinov et al. 2003) but lower than in sediment from Naples harbor (Italy) (Sprovieri et al. 2007) and Kaohsiung Harbor, Taiwan (Chen and Chen 2011; Chen et al. 2013).

Assessment of Sediment Quality Using Biological Thresholds

Sedimentary PAHs can affect benthic and benthic-pelagic organisms. Therefore, considering the toxicity and bioaccumulation property of PAHs, as well as diverse ecological significance of the coastal area of Bangladesh, it is of great importance to evaluate the potential risk of PAHs in this area. Since no environmental quality guideline for PAHs has been established yet in Bangladesh, we compare our results with those proposed by other nations and/or international organizations.

Many approaches have been developed to evaluate the ecotoxicological aspect of sediment-bound PAH contamination. The sediment quality guidelines (SQG) developed by the U.S. National Oceanic and Atmospheric Administration is a widely used sediment toxicity screening guideline that provides two target values to estimate potential biological effects: effects range low (ERL) and effects range median (ERM) (Long et al. 1995). The comparison of the measured concentrations of PAHs with the ERM and ERL values are shown in Table 1. Among the sediment samples analyzed, none of them exceeded the ERM of \sum PAHs. The \sum PAHs are between ERL and ERM in 50% of samples in both seasons, and below ERL in the other samples (50%). In most of the samples except ME2 and ME3 in winter and CX2 and ME3 in summer, the concentrations of the majority of the individual PAHs were higher than ERL but lower than ERM value, which indicate that biological effects might occur occasionally. Moreover, some PAHs were in at least one sample that exceeded ERM in sites CT3 and SN2 in winter and CX3, CT1, CT2, CT3, CT4, and SN2 in summer (e.g., site CX3: Phe, site CT1: Ace and Pyr, site CT2: Pyr, site CT3: Pyr and Chr, site CT4: Pyr and site SN2: Ace and DahA), which indicates that biological effect would occur frequently at these sites.

In addition to ERL and ERM, we also evaluated the SQG for high molecular weight PAHs such as BbF, BkF, BghiP, and IP based on the interim sediment quality guideline (ISQG) range recommended by the Canadian sediment guidelines for the protection of aquatic life (Long et al. 1995; Behnisch et al. 2003). The low effect limits (LELs) of BbF, BkF, BghiP, and IP are 200, 240, 200, and 170 ng/g dw, respectively, with serious effect limits (SELs) of 320,000, 1,340,000, 320,000, and 32,000 ng/g dw. None of the measured PAH in any samples exceeded the SEL. The concentrations of BbF, BkF, BghiP, and IP exceed the LEL values in 7–43% of samples irrespective to season. These findings showed that PAHs in the sediment from coastal areas of

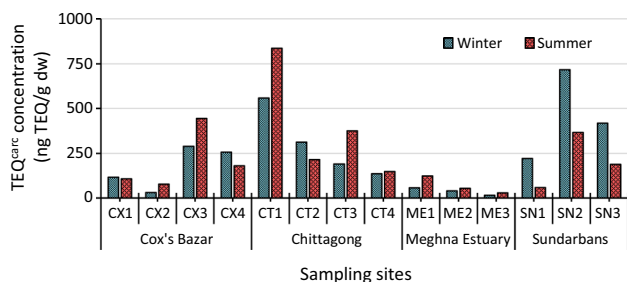


Fig. 6 Distribution of TEQ^{carc} values (ng TEQ/g dw) in the surface sediments of the Bangladeshi coastal areas

Bangladesh will cause adverse biological effects mostly occasionally but for some sites the effect would occur more frequently.

Conclusions

The concentrations of 16 USEPA priority PAHs were measured in the surface sediment from the coastal areas of Bangladesh. The samples were collected in winter (January–February) and summer (August–September), 2015. All of the 16 PAHs were detected in the sediment samples. The levels of PAHs ranged from moderate to a relatively high PAHs pollution compared to other coastal areas worldwide. The most abundant PAHs by ring structures were 4- to 5-ring, suggesting high molecular weight PAHs were prevalent in the Bangladeshi coastal sediments. The concentrations of Σ PAHs were slightly higher in summer than those in winter, but the seasonal variations were not statistically significant, thus suggesting inputs of PAHs from almost similar sources in the two seasons. Some selected molecular ratios demonstrated that the PAHs in the Bangladeshi coastal areas originated from both the petrogenic and pyrogenic sources, including crude petroleum (e.g., gasoline/diesel), petroleum combustion, and combustion of grass, wood, and coal. Spatial distribution revealed that the Chittagong, Sundarbans, and Cox's Bazar areas were more contaminated with PAHs than the Meghna Estuary because of greater development, including industrialization and urbanization activities. The concentrations of the sedimentary PAHs exceeded the environmental quality guidelines in a high percentage of samples, indicating the toxic effects of these compounds might be posed to the ecosystem, which should not be ignored and some control measures or remedial actions are recommended to lessen the PAH contamination. Furthermore, long-term continuous monitoring should be conducted to develop effective management strategies, as well as the existing legal policies should be utilized to control the PAH pollution in the coastal areas of Bangladesh.

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Compliance with Ethical Standards

Conflict of interest The authors declare that they have no conflict of interest.

Ethical Approval This article does not contain any studies with human participants or animals performed by any of the authors.

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