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ORIGINAL ARTICLE

Distribution of polycyclic aromatic hydrocarbons in recent sediments of Sundarban mangrove wetland of India and Bangladesh: a comparative approach

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Abstract The present work is the first attempt to compare the data of a comprehensive study of the origin and distribution of 16 priority pollutant polycyclic aromatic hydrocarbons (PAHs) in surface sediments (<63 μm) from 18 sampling stations, 9 from Sundarban of Bangladesh and 9 from Indian counterpart. Σ19PAHs concentration in sediments showed wide variations from 208.3 to 12,993.1 ng g^{-1} dry weight in Indian Sundarban, whereas 208.4 to 4,687.9 ng g^{-1} in the case of Bangladesh. Fluoranthene, pyrene, benzo(b)fluoranthene, benzo(a)pyrene and dibenzo(a,h)anthracene were predominant species for both the countries. The PAH diagnostic ratios indicated that the PAHs in sediments from both the countries were of mixed source of hydrocarbons of both petrogenic and pyrolytic origin. According to the numerical effect-based sediment quality guidelines, the levels of PAHs in the Sundarban wetland of Bangladesh and India should not exert adverse biological effects. The TEQ values calculated for samples from the Bangladesh and Indian Sundarban varied from 13.68 to 1,014.75 and 1.31 to 2,451 ng g^{-1} d.w. with an average of 221.02 and 358.63 ng g^{-1}, respectively. The overall contamination status of PAH was higher in India than Bangladesh.

Keywords Polycyclic aromatic hydrocarbons (PAHs) · Sediment · India · Bangladesh · Sundarban

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are by-products of incomplete combustion of organic materials and have raised severe environmental concerns, owing to their carcinogenic properties (Wang et al. 2010). Anthropogenic inputs of PAHs from oil spills, ship traffic, urban runoff, waste discharge, as well as atmospheric fallout of vehicle exhaust and industrial stack emission have caused significant accumulation of PAHs in the marine environment, which is generally considered as the ultimate pool (Simpson et al. 1996; Seruto et al. 2005). PAHs, most of which are categorized as persistent organic pollutants (POPs), are of environmental significance due to their widespread distribution in the environment and potential toxicity to organisms. These contaminants are often hydrophobic and easily accumulated in organisms through food chains and present potential threat to aquatic ecosystems and also to human health (Colborn et al. 1993). PAHs are hydrophobic compounds; therefore, sediments represent one of the most important reservoirs of PAHs in the marine environment (Perra et al. 2011). The sorptive properties of PAHs are largely controlled by their hydrophobicity and by the particle organic carbon content. In the water column, PAH tend to associate with suspended particulate matter and are

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eventually deposited in sediments (Santschi et al. 2001). Since the deposition of sediments continues over time, sediments can act as geochronometers of the contaminant deposition in the environment as well as of general environmental change over time (Hites et al. 1977; Yamashita et al. 2000; Luo et al. 2004; Kannan et al. 2005; Van Metre and Mahler 2005). Studies concerning PAHs in water column and sediments can lead to better understanding of PAH cycles in top-to-bottom processes and their historical trend. Recent studies on the pollution status of Indian Sundarban wetland have revealed the presence of high concentrations of heavy metals (Zn and Cu), as well as persistent organic pollutants in sediments (Chatterjee et al. 2007; Sarkar et al. 2007; Sharif et al. 1991, 1993a, b). These findings have initiated our present collaborative work to evaluate the occurrence of PAHs in surface sediment samples from the Sundarban wetland belonging to both India and Bangladesh. The present paper aims to identify and quantify the potential sources of the PAH contamination in sediments to get a coherent picture of the two neighboring countries. Through the comparison of levels and distributions for PAHs, this paper aims at studying the coherence of results issued from Sundarban wetland of two neighboring countries structured by the same objectives and, therefore, by the same type of sampling strategies.

Methodology

Geographical setting

The funneled mouth of the Ganga–Brahmaputra–Meghna Rivers stretches for an east–west length of more than 380 km at the Bay of Bengal sea face, of which the Hugli estuary, the westernmost distributory of the Ganges occurs in the westernmost part of the Teri River, a distributory system of the Meghna river occurs in the easternmost direction. Despite a huge amount of sediment supply by the former two major rivers (~500 × 106 tons/year by the Ganges and ~700 × 106 tons/year by the Brahmaputra), the delta has not shown any appreciable accretion since the last 250 years due to subsidence along the north–northwest axis of the present river mouths (Eisma 1998). In this huge deltaic complex, the southerly flowing rivers are quite active in the eastern (Bangladesh) part, whereas tidal flows dominate over riverine flows in the western part (India). In the Indian part, the tidal flows surpass the fluvial fluxes for about 8 months of the year and river Ganges receives freshwater only during the four monsoon months of the year (Goodbred 2003) together with some surface runoff from the reclaimed catchment areas. Tides at the major river and estuary mouths are macrotidal (4.2–6 m) and semi-diurnal. The tidal amplitude may reach as high as 7 m or above during monsoon. In the southernmost portion of the delta, i.e., complex, the tidal creeks and large channels are generally closed by embankments except Khulna district of Bangladesh and the adjacent areas in India. The impact of reclamation with partly or fully cut mangroves is however more on the western part (Mukherjee 1969; Snead 1985).

Study area

The present study areas and the sampling locations are bounded by longitudes 88°10′E and 89°30′E at the surface. In the Indian Territory, the sampling stations have been chosen along the Hugli estuary including and adjoining areas belonging to Indian Sundarban. The nine sampling locations, e.g., Lot 8 (S_1) Gangadharpur (S_2), Gangasagar (S_3), Fresargunge (S_4), Jharkhali (S_5), Gosaba (S_6), Canning (S_7), Sandeshkhali (S_8) and Gushighata (S_9) belong to the different wave tidal environments and anthropogenic stresses (as in Fig. 1a).

The sampling stations in Bangladesh territory follow a north to south direction covering nine separate locations within the Bangladesh Sundarban. Some of these stations have included the points emergence of some distributory channels with the main north to south flowing Martaja River estuary opening in the Bay of Bengal. The sampling stations are Khalispur (S_1), Labanchara (S_2), Mongla port (S_3), Dhangmari (S_4), Karamjal (S_5), Chandpy (S_6), Estuary (S_7), Hiron Point (S_8) and Sea (S_9). All the sampling locations of Bangladesh territory also have different and distinctive wave-tide climate together with the impacts of anthropogenic stresses exerted by the Mongla Port and other activities like agriculture, industries and reclamation. The present paper is the first attempt to get a comprehensive comparative account of the concentrations of PAH congeners in sediments of the estuarine and deltaic systems of the two countries. The distance between the Hugli estuary and the Martaja estuary at the sea face is ~140 km. The study stations belong to the same chunk of 6,000 km^2 area of mangrove forest- the Sundarban, a UNESCO World Heritage Site and a well-known mangrove wetland in the world.

Sampling and analytical procedure

Samples were collected with the help of a stainless steel spade, stored in polyethylene bags, and transported to the laboratory. Once in the laboratory, samples were oven-dried at 50 °C (Sarkar et al. 2012; Zuloaga et al. 2009) and were divided into two parts: one for physicochemical analysis and the other for PAH analysis. The sediment samples were stored in labeled polyethylene bags kept in iceboxes and

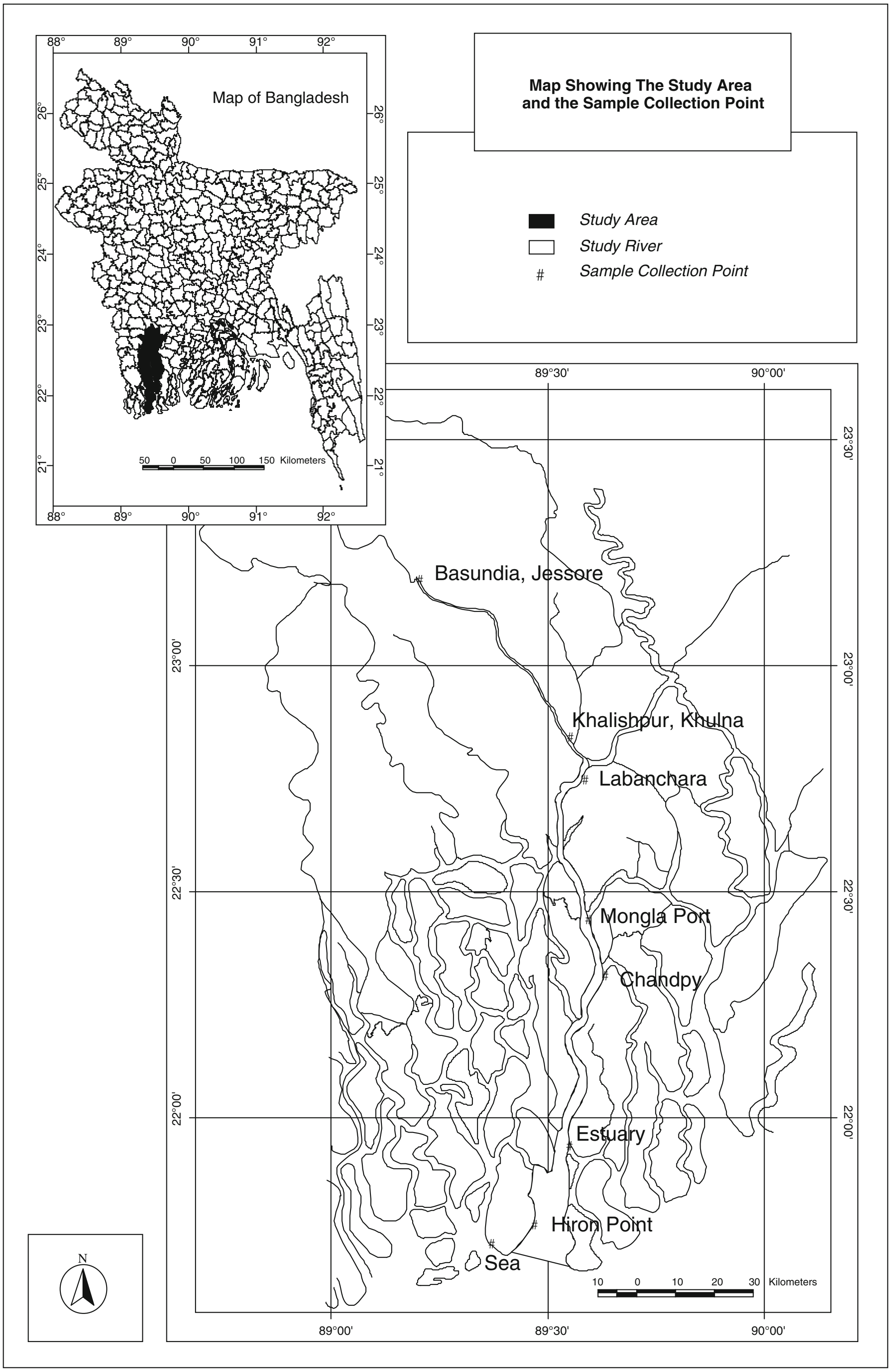

Fig. 1 **a** Map of Bangladesh showing location of the nine sampling stations (S_1–S_9). **b** Map of Hugli River estuary including Indian Sundarban showing location of the nine sampling stations (S_1–S_9)

transported to the laboratory where they were frozen to −20 °C. Within 2 days, a portion of each sample was placed in a ventilated oven at a very mild temperature (max. 40 °C). Dried samples were then disaggregated using an agate mortar and pestle and divided into two aliquots. One aliquot was sieved through 63 μm metallic sieve and stored in hermetic plastic bags until elemental analysis. All visible marine organisms and coarse shell fragments, sea grass leaves and roots when present were removed manually. The other aliquot was unsieved and used for sediment quality parameters (organic carbon, pH, % of silt, clay and sand). Organic carbon (C_{org}) content of the soil was determined

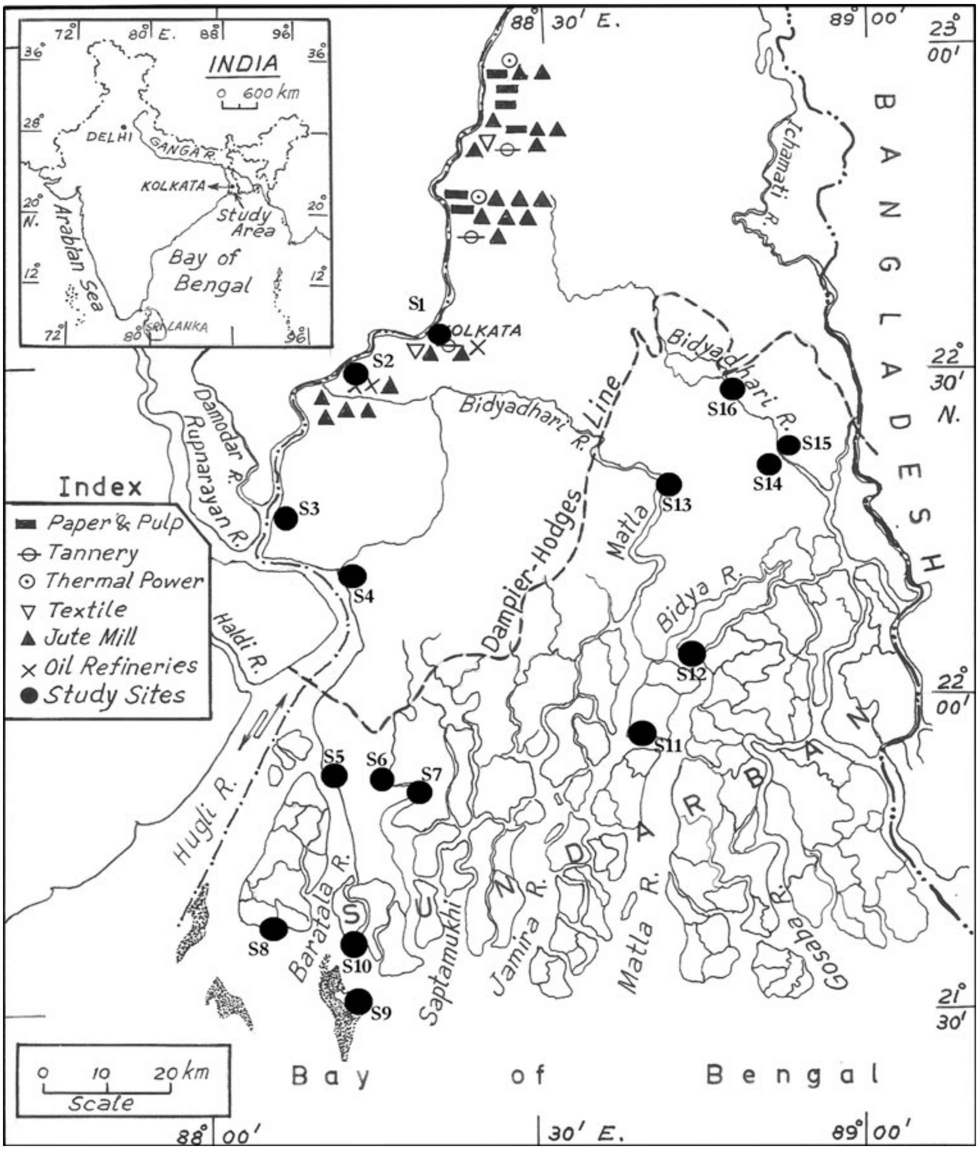

Fig. 1 continued

following a rapid titration method (Walkey and Black 1934) and pH with the help of a deluxe pH meter (model no. 101E) using combination glass electrode manufactured by M.S. Electronics (India) Pvt. Ltd. Mechanical analyses of sediment were done by sieving in a Ro-Tap Shaker (Krumbein and Pettijohn 1938) manufactured by W.S. Tyler Company, Cleveland, Ohio, and statistical computation of textural parameters was done by using the formulae of Folk and Ward (1957). Approximately 1.0 g of dry sediment sample was weighed and submitted to microwave-assisted extraction in 15 ml of acetone at 21 psi during 15 min and at 80 % of microwave power in a MDS-2000 microwave (CEM, Matthews, NC, USA), according to Bartolomé et al. (2005). 25 µl of a mixture of acenaphthene-d10, chrysene-d12 and phenanthrene-d10 at 20 µg ml^{-1} in acetone was added as surrogates. The extract was then concentrated under a gentle stream of nitrogen and loaded onto a 1-g Florisil® cartridges (Supelco, Walton-on-Thames, UK) for further clean-up of the extract. PAHs were eluted in a (4:1) *n*-hexane:toluene mixture. The elutes were concentrated to dryness, re-dissolved in 500 µl of isooctane and kept in the dark at −18 °C until analysis.

The extracted compounds were analyzed on a 6890 N chromatograph coupled to a 5973 mass spectrometer with a 7683 autosampler (Agilent Technologies, Avondale, PA, USA). Two microliters of the sample were injected in the splitless mode (2 min) at 270 °C into a 30 m × 0.25 mm × 0.25 µm HP-5 capillary column. For the separation of PAHs, the temperature program was: 60 °C for 2 min, increase at 10 °C min^{-1} to 290 °C with a final hold for 10 min. The carrier gas was helium (C-50) and was kept at a constant flux of 1.5 ml min^{-1}. The mass spectrometer was operated in the electron impact ionization mode at 70 eV. The interface was kept at 300 °C and the

Table 1 Details of the measurements performed in the SIM mode for each PAH congener

PAHs	*m*/*z*	LOD (ng g^{-1})	LOQ (ng g^{-1})	RSD sample (%)
Naphthalene (Nap)	128, 129	19.84	32.73	5.6–7.6
Acenaphthylene (Acy)	154, 154	0.49	1.17	12.8–35.0
Acenaphthene (Ace)	153, 154	2.15	4.48	18.0–30.0
Fluorene (Flu)	165, 166	2.44	6.12	5.0–13.6
Phenanthrene (Phen)	178, 179	0.42	0.99	5.0–11.2
Anthracene (Anth)	178, 179	0.17	0.34	8.9–14.1
Fluoranthene (Flr)	202, 203	0.32	0.62	8.5–24.5
Pyrene (Pyr)	202, 203	3.62	7.35	10.0–27.8
Benzo(a)anthracene [B(a)A]	228, 229	0.08	0.17	7.3–25.6
Chrysene (Chr)	228, 229	0.21	0.33	6.3–17.2
Benzo(b)fluoranthene [B(b)F]	252, 253	0.09	0.15	15.5
Benzo(k)fluoranthene [B(k)F]	252, 253	0.08	0.13	22.7
Benzo(a)pyrene [B(a)P]	252, 253	0.1	0.15	8.2–14.5

Table 2 Sediment quality parameters of Bangladesh samples

Parameters	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9
pH	7.9	7.9	7.9	7.9	7.9	7.9	7.5	7.5	7.5
Organic carbon (%)	1.5	1.39	1.46	1.42	1.39	1.33	1.51	1.55	0.52
Sand (%)	5.1	10.1	2.55	7.7	11.3	10.1	6.6	5.1	75.1
Silt (%)	57.75	60.25	68.87	61.55	54.45	52.75	45.15	47.75	2.75
Clay (%)	37.15	29.65	28.58	30.75	34.25	37.15	48.25	47.15	22.15
Texture	Silty Clay Loam	Silty Clay Loam	Silty Clay Loam	Silty Clay Loam	Silty Clay Loam	Silty Clay Loam	Silty Clay	Silty Clay	Sandy Clay Loam

Table 3 Sediment quality parameters of sediment samples collected from India

Parameters	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9
pH	8.5	6.5	8.4	7.6	6.5	7.9	8.0	7.9	8.3
Organic carbon (%)	0.69	0.85	0.22	0.56	0.81	0.73	0.83	0.66	0.85
Sand (%)	9.88	0.48	32.85	70.6	13.04	2.4	2.10	20.2	12.2
Silt (%)	47.36	40.1	58.45	26.2	68.36	66.56	67.63	52.9	31.8
Clay (%)	42.76	59.42	8.70	3.4	18.6	31.04	30.27	26.8	55.9
Texture	Silty Clay	Silty Clay	Silty Clay	Sandy	Silty Clay	Silty Clay	Silty Clay	Fine loamy	Silty Clay

ionization source and the quadrupole at 230 and 150 °C, respectively. Measurements were performed in the SIM mode; the *m*/*z* values for each congener are included in Table 1. Seven-point internal calibration curves were built in the range of 0.05–16 μg ml^{-1} for the target compounds. The proposed analysis method was validated after application to NIST 1944 certified reference sediment. The recoveries obtained were in good agreement with the certified values (Bartolomé et al. 2005) and relative standard deviations were lower than 17 % for all the analytes, except for naphthalene (35 %). Reagent blanks were performed for every ten samples and limits of detection (LOD) were estimated as three times the signal of the blanks plus three times the standard deviation and were in the 0.5–11 ng (Bartolomé et al. 2005). For every ten samples, replicates of the NIST 1944 were also performed in order to guarantee that the analyses were under control during the processing of the samples.

Results and discussion

Sediment geochemistry and PAH accumulation

The sediment quality parameters (pH, organic carbon and textural properties) of the different samples vary among

Table 4 Concentration (ng g^{-1}) of PAH congeners and diagnostic ratios of sediments from Bangladesh

PAHs (ng g^{-1})	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9
Naphthalene (Nap)	49.3	55.95	0	0	58.9	52.5	55.3	64.6	0
Acenaphthylene (Acy)	0	55.2	35.2	2.9	27.9	4.1	4.2	67.96	31.1
Acenaphthene (Ace)	13.8	50.6	60.3	20.6	19.8	19.5	28.6	146.6	97.1
Fluorene (Flu)	21.1	33.97	11.1	0	12.8	10.9	19.7	17.8	116.5
Phenanthrene (Phen)	190.3	164	47.9	34.9	44.7	34.2	36.9	29.7	287.9
Anthracene (Anth)	33.1	28.8	0	0	0	0	0	0	40.2
Fluoranthene (Flr)	507.3	**641.9**	51.9	40.3	38.5	29.7	64.5	34.5	363.3
Pyrene (Pyr)	411.1	546.8	42.7	30.5	32.9	20.9	48.7	35.1	276.1
Benzo(a)anthracene [B(a)A]	266.4	319.8	10.7	8.6	8.5	6.7	14.2	11.7	99.9
Chrysene (Chr)	323.3	440.3	16.1	11.8	11.6	10.2	18.5	13.8	101.6
Benzo(b)fluoranthene [B(b)F]	302.97	544.9	18.6	10.7	5.5	8.6	17.1	15.5	81
Benzo(k)fluoranthene [B(k)F]	243.3	388.8	14.8	8.3	5.3	7.9	11.8	12.3	74.6
Benzo(a)pyrene [B(a)P]	281.2	451.3	9.8	10.6	4.96	6.5	14.6	8.8	95.5
Dibenzo(a,h)anthracene [D(ah)A]	257.5	462	7.5	8.4	5.7	6.1	11.6	12.3	79.5
Benzo(ghi)perylene [B(ghi)P]	36.9	396.9	4.4	4.2	2.98	4.9	4.6	5.4	23.5
Indeno pyrene (Ind)	375.8	106.5	19.4	16.6	15.6	14.3	18.7	23.3	125.9
Diagnostic ratios									
Phe/Pyr	0.46	0.29	**1.12**	**1.14**	**1.36**	**1.64**	0.76	0.84	**1.04**
Flu/Pyr	0.051	0.06	0.26	0	0.39	0.52	0.40	0.51	0.42
Flu/(Pyr + Flu)	0.049	0.06	0.21	0	0.28	0.34	0.29	0.34	0.29
Pyr/BaP	**1.46**	**1.21**	**4.37**	**2.89**	**6.63**	**3.19**	**3.35**	**3.98**	**2.89**
Chr/BaA	**1.21**	**1.38**	**1.50**	**1.37**	**1.36**	**1.53**	**1.31**	**1.18**	**1.02**
LMW/HMW	0.10	0.09	0.79	0.39	**1.25**	**1.05**	0.65	**1.89**	0.43

them, as depicted in Tables 2 (for sediment samples collected from Martaja estuary, Bangladesh) and 3 (for sediment samples collected from Hugli estuary, India).Values of pH range from slightly acidic to basic (pH 6.5–8.5) for the samples from Hugli River estuary and adjoining areas of India but mainly basic for Martaja estuary samples, Bangladesh (pH 7.5–7.9); the acidic nature is partly due to the oxidation of FeS_2 and FeS to $SO4_2^-$ and partly results from the decomposition of mangrove litter and hydrolysis of tannin in mangrove plants releasing various kinds of organic acids (Liao 1990). Organic carbon (OC) values were below 1 % in all stations for Hugli estuary samples but were above 1 % for Martaja estuary samples except at S_9. Very low organic carbon values in the intertidal zone sediments of Sundarban wetland, India, were also recorded in previous studies (Sarkar et al. 2004; Chatterjee et al. 2007). These low values of organic carbon are probably related to the poor absorbability of organics on negatively charged quartz grains, which predominate in the intertidal sediments of this estuarine environment (Bhattacharya and Sarkar 2003; Sarkar et al. 2004). Regarding textural composition, all the studied stations at the two countries also exhibit wide variations with differences in the proportions of silt and clay content. Textural differences might be attributed to vigorous estuarine mixing, suspension–resuspension and flocculation–deflocculation processes. These variations might influence the PAH accumulation in the sediments (Tables 4, 5), where the processes involved are quite complex in nature. Other important factors are the chemical properties of the compounds, especially their water solubility, sediment composition and organic carbon content.

Distribution of PAHs in sediments

Concentrations of 16 PAHs measured in surface sediments of 9 sampling sites in Martaja river estuary, Bangladesh, and 9 sampling sites in Hugli river estuary, India, have been summarized in Tables 4 and 5, respectively. PAH concentrations ranged from 0 to 641 ng g^{-1} in Martaja River, Bangladesh, sediments and 0 to 1,839.5 ng g^{-1} for Hugli River sediment samples, India. The prevalent level of PAH concentration is lower than that of Mediterranean coastal sediments of Egypt (Barakat et al. 2011), Huaihe River sediments, China (Fu et al. 2011) and Liaoning, Jilin Heilongjiang provinces, northeast China (Lin et al. 2012). Figure 2a and b show the distribution pattern of two-, three-, four-, five- and six-ring PAH compounds in the individual stations of Bangladesh and India, respectively. For Martaja sediments, as it can be seen in Fig. 2a, there is

Table 5 Concentration (ng g^{-1}) of PAH congeners and diagnostic ratios of sediments from India

PAHs (ng g^{-1})	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9
Naphthalene (Nap)	418.5	86.1	216.8	42.9	46.9	142	198.4	121.9	108.6
Acenaphthylene (Acy)	157.1	14.9	2.9	1.7	6.7	8.6	11.7	10.7	12.3
Acenaphthene (Ace)	38.6	23.9	25.9	32.2	20.4	29.99	26.8	26.2	26.3
Fluorene (Flu)	28	36.7	23.3	11.7	6	22.7	22.4	21.3	20.97
Phenanthrene (Phen)	426.2	107.7	178.9	56.1	157.5	234.5	106.7	60.3	104.97
Anthracene (Anth)	224.2	10.3	5.9	3.2	8.9	14.99	5.6	3.6	5.95
Fluoranthene (Flr)	1,839.5	125.5	201.3	27.8	152.2	555.2	140.2	58.6	82.6
Pyrene (Pyr)	1,572.1	86.3	89.5	24.2	101.7	475.6	138.9	54.7	74.2
Benzo(a)anthracene [B(a)A]	845.8	28.7	10	3.3	25.3	126.9	13.6	12.2	19.6
Chrysene (Chr)	1,090.1	40.3	21.95	2.7	37.7	203.6	25.97	17.5	24.4
Benzo(b)fluoranthene [B(b)F]	1,123.7	50	13.6	<LOD	35.3	234.8	21.4	19.3	22.4
Benzo(k)fluoranthene [B(k)F]	1,284.2	46.5	20.2	<LOD	37	186.5	24.6	17	23
Benzo(a)pyrene [B(a)P]	1,140.9	39.6	43.1	0.4	41.96	187.9	13.98	11.2	30
Dibenzo(a,h)anthracene [D(ah)A]	952.8	39.8	25.8	0.5	28	130.5	22.7	20.8	29.6
Benzo(ghi)perylene [B(ghi)P]	387.9	20.7	35.8	0.7	18.8	67.5	6.8	12.3	10.7
Indeno pyrene (Ind)	1,463.6	57.6	53.7	0.8	44.6	172.95	26.95	33.98	42.8
Diagnostic ratios									
Phe/Pyr	0.27	**1.25**	**2**	**2.31**	**1.55**	0.49	0.77	**1.1**	**1.41**
Flu/Pyr	0.02	0.43	0.26	0.48	0.06	0.05	0.16	0.39	0.28
Flu/(Pyr + Flu)	0.02	0.29	0.21	0.32	0.06	0.05	0.14	0.28	0.22
Pyr/BaP	**1.38**	**2.18**	**2.08**	**60.6**	**2.42**	**2.53**	**9.94**	**4.89**	**2.47**
Chr/BaA	**1.29**	**1.41**	**2.19**	**0.82**	**1.49**	**1.6**	**1.92**	**1.44**	**1.25**
LMW/HMW	0.11	0.5	0.9	2.5	0.5	0.2	0.9	0.95	0.8

a dominance (60–70 % of the total PAHs) of high molecular weight PAH compounds (HPAHs) (i.e., 3–4, 5–6 rings) at stations S_1, S_2, S_3, S_4 and S_9. It can be referred here that the low molecular weight PAH compounds (LPAHs) were degraded more rapidly in the sediments (Jones et al. 1986) in these five sampling sites. In comparison, the three-, four-, five- and six-ringed PAHs and their alkyl groups have been found to be more refractory in a natural environment which is essential in preserving the source features (Sauer and Boehm 1991). The predominance of HMW PAHs at majority of the study sites in Martaja estuary, Bangladesh, was the evidence of the strong influence of pyrogenic sources for these contaminants (Kimbrough and Dickhut 2006).

This conclusion was supported by a previous research, which found automobile emissions and coal-dominated PAH sources in the southern Chesapeake Bay region, USA (Dickhut et al. 2000). On the other hand, the distribution of two-, three-, four-, five- and six-ringed PAHs in the individual stations of Hugli River estuary, India, is completely different from that of Bangladesh. Different trends of PAH distribution were observed as evidenced in Fig. 2b. For example, stations S_1, S_2 and S_5, S_6 show low contribution of two-ring PAH compounds compared to three-, four-, five- and six-ring PAH compounds, i.e., dominance of high molecular PAH molecules. Dominance of two-ring PAH compounds could be found in station S_8, while station S_4 shows very low contribution of five- and six-ring PAH compounds. The dominance of high molecular PAHs, i.e., three-, four-, five- and six-ring PAH compounds was pronounced at stations S_1, S_2, S_3, S_5, S_6, S_7 and S_9. Thus, it could be stated that the source of PAHs generating different ring structures are various in case of Hugli River sediments. These mixed sources were also found in the previous observations of the same area (Sarkar et al. 2007; Binelli et al. 2008, etc.). Coal is the dominant source of PAHs as in other river sediments (Walker and Dickhut 2001). However, automotive sources of PAHs also dominate (Dickhut et al. 2000). Coal has distinctly high levels of fluoranthene and pyrene with lower level of other HMW PAHs. In contrast, auto-emissions have similar levels of fluoranthene and pyrene, relative to benz(ghi)perylene and only moderately lower levels of both benzo(a)anthracene and benzo(a) pyrene (Kimbrough and Dickhut 2006).

Assessment of diagnostic ratios

PAHs can be generated by three main processes: combustion at a very high temperature of organic matter, release of petroleum or diagenetic processes (degradation

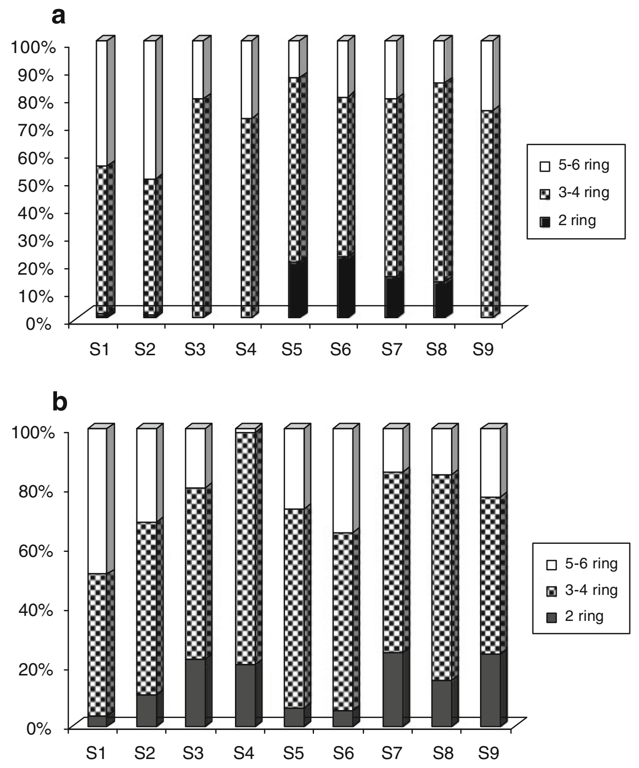

Fig. 2 **a** Percentage distribution of two-, three-, four-, five- and six-ring PAH congeners in the nine sampling stations of Bangladesh (S_1–S_9). **b** Percentage distributions of two-, three-, four-, five- and six-ring PAH congeners in the 16 sampling stations of the Hugli River estuary (S_1–S_9), India

of the organic matter) (Neff 1979). However, for PAHs contamination, it is difficult to discriminate between pyrolytic products and petroleum-derived ones, due to the parameters complexity governing PAHs distribution in the marine sediments. In general, petrogenic PAHs are more available than the pyrolytic ones, because the first ones are introduced directly into the marine environment in dissolved, colloidal forms or fairly associated with suspended matter, whereas the last ones are probably strongly associated with the particles issued from pyrolytic sources. Besides, pyrogenic PAHs are characterized by the dominance of the high molecular weight (HMW) (PAHs 5–6 rings) (Muel and Saguem 1985), while petroleum hydrocarbons are dominated by the low molecular weight (LMW) (PAHs 2–4 rings) (Neff 1979; Wise et al. 1988; Berner et al. 1990).

In the coastal area of West Bengal, the industrial and anthropogenic activities constitute the main source of PAHs. Several methods have been used to identify the origin and the sources of PAHs in marine sediments, such as the characteristic of PAHs profiles or the characteristic ratios between PAH compounds. The use of PAH ratios is not feasible, due to the variety of processes contributing to the distribution of PAHs in marine sediments. In spite of it, information on major pollution sources for a particular area can help to infer the primary factors, which cause the appearance of these compounds in the environment. Diagnostic ratios provide only qualitative information about the contribution of various sources with regard to PAH contaminations (Wang et al. 2010). A first step to discriminate between petrogenic and pyrolytic contamination consisted in comparing LPAHs and HPAHs compound abundances (Perra et al. 2011). In order to estimate the origin of the contamination, the LMW/HMW ratio was the first index calculated (Tables 4, 5), where LMW = Naph + Acy + Ace + Flu + Phen + Anth + Flu + Pyr + B[a] A + Chry and HMW = B[b + k]Fl + B[a]P + DB[ah]A + B[ghi]P + IndP. It was revealed that most values of the LMW/HMW ratio are lower than 1 indicating a pyrolytic origin of PAHs excepting S_5, S_6 and S_8 for Martaja river samples (LMW/HMW = 1.25, 1.05 and 1.89, respectively) and S_4 for Hugli River samples (LMW/HMW = 2.5), where PAHs contamination could arise from a petrogenic source. Another method to estimate the origin of the PAHs in sediments uses the ratios Phen/Anth and Flu/Pyr. Phenanthrene and anthracene are the two structural isomers. Because of their different physico-chemical properties, they could behave differently in the environment and could lead to different values for their Phen/Anth ratio (Gschwend and Hites 1981). Pyrolysis of organic matter at very high temperature generates PAHs, characterized by a low Phen/Anth ratio (<10), while the slow maturation of petroleum at lower temperatures leads to much larger values of the Phen/Anth ratio (>25) (Soclo 1986; Raoux 1991). In conclusion, phenanthrene is thermodynamically more stable than anthracene, so a Phen/Anth ratio <10 is characteristic of a pyrolytic pollution (Baumard et al. 1999; Budzinski et al. 1997; Sicre et al. 1987). In the same way, the Flu/Pyr ratio is also discriminating. Fluoranthene is less thermodynamically stable than pyrene and a predominance of fluoranthene over pyrene is characteristic of pyrolytic products, while in petroleum-derived PAHs, pyrene is more abundant than fluoranthene (Budzinski et al. 1997). During the combustion process, pyrolytic products are usually characterized by a fluoranthene/pyrene ratios >1 (Baumard et al. 1999). The ratios of phenanthrene to anthracene (Phen/Anth) and fluoranthene to pyrene (Flu/Pyr) have been applied to assess the contribution of petroleum or combustion sources of PAHs. A Phen/Anth ratio of <10 and Flu/Pyr ratio of >1 have also been evidenced in the majority of the cases, which strongly support pyrogenic origins of PAHs (Baumard et al. 1998). Sediment samples with Phen/Anth <10 and Flu/Pyr <1 were characteristic of a mixed pattern of pyrogenic and petrogenic contamination. It is probably prudent to observe that all the samples display a dominance of the pyrolytic PAHs. The likely sources of these are a combination of local point source anthropogenic inputs and the ubiquitous pyrolytic PAHs deposition that results from global Aeolian transport (Jaward et al. 2004). The latter will be present in greater proportion in the samples with low, compared to those higher, total PAH concentrations. The distribution of individual PAHs may be used to differentiate between pyrolytic and petrogenic sources. These are characterized by certain molecular indices based on the concentration ratios of selected PAHs (Colombo et al. 1989). However, one difficulty in identifying the PAH sources is the possible coexistence of several of them and the transformation processes that PAHs can undergo before deposition in the sediments (Butler and Crossley 1981). The wide range of PAH ratios found in sediments of both Martaja (Bangladesh) and Hugli estuary (India) may suggest a mixed source of hydrocarbons of both petrogenic and pyrolytic origin, although some post-depositional alterations within the sediment cannot be ruled out (Bicego et al. 2006). In order to provide a better estimate of PAH sources, a series of ratios are worked out. PAH sources to wetland sediments were similar. Moreover, it can be seen that they reflect predominant combustion sources and particularly, originated from grass, wood and coal.

The large use of coal and wood for domestic cooking, preparing molasses and husking rice by local inhabitants was reported by Chatterjee et al. (2009) from Indian Sundarban. The combustion source of PAH was also endorsed by Liu et al. (2007) from China. In addition, this also acts as a potential local atmospheric source of PAHs. A comparative account of the PAH contamination status of the two countries shows that, in general, there is a great difference in the distribution pattern and concentration range of PAH congeners in two countries. The sediment samples from Sundarban wetland, India, showed higher range of PAH concentration (208.3–12,993.1 ng g^{-1}) than that of Bangladesh samples (208.4–4,687.9 ng g^{-1}). According to the number of aromatic rings, the 16 PAH compounds were divided into four groups representing two-, three-, four-, five- and six-ring PAHs. For sediment samples of Martaja River, Bangladesh, a higher proportion of PAH species with three- and four-ring structures for majority of the stations and five to six rings for the station S_2 was detected. Compositions and relative abundance of individual PAH in sediments of the Hugli River, India, were quite similar, except for site S_1, S_3 and S_4, where the two-ring PAH were predominant. The result suggests that the PAH contamination in the two countries come essentially from same identical source and is indicative of a pyrolytic origin. Among all 9 PAHs, acenaphthene, naphthalene and fluoranthene were predominant species in the nine sampling sites of Bangladesh, which accounted approximately for 0.42–29.36, 0–22.16 and 13.69–19.34 %, respectively, of total PAHs. In addition, pyrene and benzo[a]pyrene were prevalent in samples from most of the sites. Conversely, the trend is different for sediments in Indian sites. Naphthalene predominated at the level of 0–95.73 % followed by phenanthrene (1.07–26.91 %) and fluoranthene (1.2–19.87 %).

Generally, biomass combustion process and the release of uncombusted petroleum products are the two main sources of anthropogenic PAHs found in the environment. In the present study, three typical pyrogenic PAHs (i.e., naphthalene, acenaphthalene and fluoranthene) were present at high concentrations. Furthermore, in both the cases, level of fluoranthene was higher than pyrene and that of indeno [1,2,3-cd]pyrene was higher for Bangladesh samples than benzo[ghi]perylene and lower for Indian samples.

Potential toxicological assessment

The effects range low (ERL) and the effects range median (ERM) values were used for impact assessment on aquatic sediment with a ranking of low to high impact values (Long et al. 1995). The measured concentrations of PAHs were compared with the existing ERL and ERM values. Results showed that naphthalene, anthracene, pyrene and benzo(a)pyrene concentrations were below the ERL of values for Martaja sediment samples (Table 6). But in the case of Hugli river sediment samples none of the PAH values showed below ERL values at 100 % cases (Table 7). These findings indicated that the studied sites have some potential biological impact. Long et al. (1995) refined the method but did not change the basic conceptual approach. TEL/PEL values (threshold effects level and probable effects level) were determined by the Florida Department of Environmental Protection (FDEP 1994). The method was similar to the method for deriving ERL/ERM values, but both ‘‘effect’’ and ‘‘no effect’’ data were used in calculating TEL and PEL values. Essentially, the TEL corresponds to the ERL and the PEL to the ERM. The TEL was calculated as the geometric mean of the 15th percentile concentration of the effects set and the 50th percentile concentration of the no-effects dataset. Similarly, the PEL was the geometric mean of the 50th percentile of the effects data and the 85th percentile of the no-effects data. The TEL (or ERL) and PEL (or ERM) values define three concentration ranges for a chemical, including those that were rarely (below the TEL or ERL), occasionally (above the TEL or ERL, but below a PEL or ERM) and frequently (above the PEL or ERM) associated with adverse effects. When contaminant concentrations exceed PEL or ERM, sediment samples were predicted to be toxic. The comparison of chemical concentrations of PAHs with sediment quality guidelines (SQGs) was summarized in Tables 6 and 7. As it can be seen, the evaluation of ecotoxicological risk can change according to considered SQGs. For Martaja river sediments, Bangladesh, PAH

Table 6 SQGs values for PAH congeners and relative percentage of samples amongst ranges of sediment quality guidelines for Bangladesh sediment samples

PAHs	SQG	% of stations			SQG	% of stations		
	ERL–ERM	<ERL	ERL–ERM	>ERM	TEL–PEL	<TEL	TEL–PEL	>PEL
Naph	160–2,100	100	–	–	35–391	33.34	66.66	–
Acy	44–640	22.22	77.78	–	6–128	22.22	77.78	–
Ace	16–500	44.44	55.56	–	7–89	–	77.78	22.22
Flu	19–540	44.44	55.56	–	21–144	11.11	88.89	–
Phe	240–1,500	11.11	88.89	–	87–544	66.67	33.33	–
Ant	85–1,100	100	–	–	47–245	100	–	–
Flr	600–5,100	11.11	88.89	–	113–1,494	66.67	33.33	–
Pyr	665–2,600	100	–	–	153–1,398	66.67	33.33	–
B(a)A	261–1,600	22.22	77.78	–	75–693	66.67	33.33	–
Chry	384–2,800	11.11	88.89	–	108–846	77.78	22.22	–
B(b)Fl	–	–	–	–	–	–	–	–
B(k)Fl	–	–	–	–	–	–	–	–
B(a)P	439–1,600	100	–	–	89–763	66.67	33.33	–
Per	–	–	–	–	–	–	–	–
InPyr	–	–	–	–	–	–	–	–
D(ah)A	63–260	66.66	22.22	11.11	6–135	11.11	66.67	22.22
B(ghi)P	–	–	–	–	–	–	–	–
Ind	–	–	–	–	–	–	–	–

concentration showed 11.11–100 % of cases exceeding the ERL values. ERM value was exceeded in the case of dibenzo(a,h)anthracene for 11.11 % cases, whereas PEL values exceeded for 22.22 % cases for acenaphthene and dibenzo (a,h) anthracene. For Hugli River samples, India, PAH contamination levels exceeded the ERM values of naphthalene (6.25 %), fluorene (6.25 %), pyrene (6.25 %), benzo(a)anthracene (6.25 %), chrysene (6.25 %), benzo(a)pyrene (6.25 %) and dibenzo (a,h) anthracene (18.75 %) while PEL level exceeded for naphthalene (68.75 %), acenaphthylene (12.5 %), fluoranthene (12.5 %), pyrene (6.25 %), benzo(a)anthracene (6.25 %), chrysene (6.25 %) and dibenzo(a,h) anthracene (12.5 %). In case of Hugli river samples, India, PAH concentration showed 44.44–88.87 % of cases exceeding the ERL values. For fluoranthene, benzo(a)anthracene and pyrene, 11.11 % of the cases for each congener exceeded the ERM values but PEL values for those congeners were in the range of 11.11–22.22 % . Some PAHs and especially their metabolic products are of great concern due to their documented carcinogenicity. These potentially carcinogenic PAHs (CPAHs) include BaA, BaP, BbF, BkF, DahA and InP (Savinov et al. 2003). The total concentrations of CPAHs in sediments from the Sundarban of Bangladesh and Indian subcontinent varied from 57 to 2,050.5 and 7.7 to 7,901 ng g^{-1} d.w. with an average of 658 and 1,152 ng g^{-1} and accounted for 49.9 and 50.6 % of the total PAHs concentrations, respectively. Among the CPAHs, BaP is the only PAH for which toxicological data are sufficient for the derivation of a carcinogenic potency factor (Peters et al. 1999). The toxic equivalency factors (TEFs) were used to quantify the carcinogenicity of other PAHs relative to BaP and to estimate BaP-equivalent doses (BaPeq dose) (Nadal et al. 2004). Calculated TEFs for BaA, BaP, BbF, BkF, InP, DahA and Chr are 0.1, 1, 0.1, 0.01, 0.1, 1 and 0.001, respectively, according to the USEPA. In this study, we converted the above mentioned seven PAH concentrations into one toxic concentration for each site using the corresponding TEFs.

The total BaP toxicity equivalent (TEQ) for all PAHs was calculated as: $\mathrm{TEQ}_{\mathrm{carc}} = \Sigma\ (\mathrm{Ci} \times \mathrm{TEF}_{\mathrm{carc}})$, where Ci was the concentration of individual PAHs (ng g^{-1} d.w.) and TEFi was its corresponding TEF. Total TEQ values calculated for samples from the Bangladesh and Indian Sundarban varied from 13.68 to 1,014.75 and 1.31 to 2,451 ng g^{-1} d.w. with an average of 221.02 and 358.63 ng g^{-1}, respectively. The maximum total TEQ value was found at sample location S_2 for Bangladesh and S_1 for India. Among different PAHs, contribution to the total TEQ decreased in the order: B[b]F (53.7 %) > D[ah]A (45.53 %) > BaP (44.47 %) > Chr (43.4 %) > B[k]F (38.3 %) > BaA (31.5 %) > Ind (10.5 %) in Bangladesh and Ind(59.7 %) > B[k]F (52.4 %) > BaP (46.5 %) > B[b[F (45.85 %) > Chr (44.5 %) > D[ah]A (38.87 %) > B[a]A (34.51 %) in India. In comparison with other studies (Table 8), the total TEQ values in sediments

Table 7 SQGs values for PAH congeners and relative percentage of samples amongst ranges of sediment quality guidelines for Hugli river sediment samples, India

PAHs	SQG	% of stations			SQG	% of stations		
	ERL–ERM	<ERL	ERL–ERM	>ERM	TEL–PEL	<TEL	TEL–PEL	>PEL
Naph	160–2,100	43.75	50	6.25	35–391	6.25	25	68.75
Acy	44–640	87.5	12.5	–	6–128	18.75	68.75	12.5
Ace	16–500	–	100	–	7–89	–	100	–
Flu	19–540	25	75	–	21–144	31.25	68.75	–
Phe	240–1,500	81.25	18.75	–	87–544	18.75	81.25	–
Ant	85–1,100	87.5	12.5	–	47–245	87.5	12.5	–
Flr	600–5,100	87.5	6.25	6.25	113–1,494	37.5	50	12.5
Pyr	665–2,600	87.5	6.25	6.25	153–1,398	87.5	6.25	6.25
B[a]A	261–1,600	87.5	6.25	6.25	75–693	81.25	12.5	6.25
Chry	384–2,800	87.5	6.15	6.25	108–846	81.25	12.5	6.25
B(b)Fl	–	–	–	–	–	–	–	–
B(k)Fl	–	–	–	–	–	–	–	–
B(a)P	439–1,600	87.5	6.25	6.25	89–763	81.25	12.5	–
Per	–	–	–	–	–	–	–	–
InPyr	–	–	–	–	–	–	–	–
D(ah)A	63–260	81.25	–	18.75	6–135	6.25	81.25	12.5
B(ghi)P	–	–	–	–	–	–	–	–
Ind	–	–	–	–	–	–	–	–

Table 8 A comparison of total TEQ in sediments (ng g^{-1} d.w.)

Location	Range	Mean	References
Coastal lagoons in central Vietnam	2–98	21	Giuliani et al. (2008)
Guba Pechenga, Barents Sea, Russia	11–300	<120	Savinov et al. (2003)
Gulf of Gemlik, Marmara Sea, Turkey	5.6–1,838	134	Ünlü and Alpar (2009)
Meiliang Bay, Taihu Lake, China	94–856	407	Qiao et al. (2006)
Jialu River, China	50.4–312.8	167.4	Fu et al. (2011)
Sundarban wetland, Bangladesh	13.6–1,014.75	221.03	Present study
Sundarban wetland, India	1.3–2,450.9	358.63	Present study

of the Bangladesh were lower than that detected in the Gulf of Gemlik, Marmara Sea, Turkey (Ünlü and Alpar 2009). However, the TEQ levels were higher than those of other literature reported sites, such as the Meiliang Bay, Taihu Lake, China (Qiao et al. 2006), the Guba Pechenga, Barents, Sea, Russia (Savinov et al. 2003), the coastal lagoons in central Vietnam (Giuliani et al. 2008) and Jialu River, China (Fu et al. 2011). In case of Indian Sundarban, the values of TEQs were found higher than all literature reported sites.

Conclusion

The analyses of the surface sediments from Indian as well as Bangladesh Sundarban environments provided very useful and important information for evaluating PAH contamination levels and probable potential sources. Data obtained reveal that the wetland appeared to be moderately polluted by PAHs in the Indian side and less polluted in the Bangladesh side. Based on the use of diagnostic source ratios, it can be referred that the PAH source in sediments was mixed comprising contamination by local vehicle combustion, biomass burning and domestic and industrial coal combustion. However, in some specific sites, a pyrogenic as well as petrogenic pattern was also observed, probably due to the mixture of industrial and urban discharges. The data provide background information that should be useful in designing future strategies for inter-governmental environmental protection of this vast wetland. Compared with SQGs values, it is revealed that the concentration of PAHs exceeded exclusively at the site Lot

8 of Indian Sundarban. This would exert adverse biological effects and demands immediate preventive measures to avoid the ecological risks on marine organisms especially the benthic species. Further research should be performed to investigate the effects of these compounds on the biogeochemical cycles of this wetland. Though high molecular PAHs are highly mutagenic and carcinogenic and low molecular PAHs are less mutagenic, low and moderate molecular PAHs presented much more ecosystem risk than high molecular PAHs in the present study.

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