Polycyclic aromatic hydrocarbons (PAHs) in surface water from the coastal area of Bangladesh

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Abstract. Sixteen USEPA priority polycyclic aromatic hydrocarbons (PAHs) in the surface water from the coastal areas of Bangladesh were analyzed by GC-MS/MS. Samples were collected in winter and summer, 2015. The total concentration of PAHs ($\sum$PAHs) showed a slight variation in the two seasons, which varied from 855.4 to 9653.7 ng/L in winter and 679.4 to 12639.3 ng/L in summer, respectively. The levels of $\sum$PAHs were comparable to or relatively higher than other coastal areas around the world. The areas with recent urbanization and industrialization (Chittagong, Cox's Bazar and Sundarbans) were more contaminated with PAHs than the unindustrialized area (Meghna Estuary). Generally, 2–3-ring PAHs were the dominant compounds. Molecular ratios suggested that PAHs in the study areas could be originated from both pyrogenic and petrogenic sources. The risk assessment revealed the extremely high ecological risk of PAHs, indicating an intense attention should be paid to PAHs pollution in the coastal areas of Bangladesh.

Keywords: polycyclic aromatic hydrocarbons (PAHs); surface water; risk assessment; coastal area; Bangladesh

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of persistent organic pollutants. These compounds are composed of two or more fused aromatic rings of carbon and hydrogen atoms. There are two predominant sources of environmental PAHs such as, petrogenic source comprising of PAHs associated with crude oil spills, and pyrogenic source including PAHs derived from fossil fuel combustion, biomass burning, waste incineration, and asphalt production (Zheng *et al.* 2016).

They are ubiquitous in the environment due to their persistence, long range transport, bioaccumulation and known to be very toxic to the biological systems (Lotufo and Fleeger 1997, Gu *et al.* 2013).

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Based on the evidences of their high toxicological risk, the United States Environmental Protection Agency (USEPA) has identified 16 PAHs as high priority pollutants. PAHs are widely distributed in marine aquatic environments, such as estuaries, coastal areas, wetlands, off-shore areas and the deep sea due to anthropogenic processes and their comparatively long half-life (Yancheshmeh *et al.* 2014, Zhang *et al.* 2016 and references therein). Bangladesh is an agricultural country that has an irregular 580 kilometers 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 are being suffered 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 semi-treated 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, it is an urgent need to monitor their pollution levels and to evaluate their potential toxicity in the environment.

In the last few decades, environmental PAHs and their distribution, sources and potential risk to ecological systems have been extensively studied in the coastal regions worldwide (Lim *et al.* 2007, Ren *et al.* 2010, Amoako *et al.* 2011, Montuori and Triassi 2012, Jaward *et al.* 2012, Sun *et al.* 2016, Zheng *et al.* 2016, Li *et al.* 2016). However, there are few studies monitoring concentrations of PAHs in the Bangladeshi environments (Zuloaga *et al.* 2013, Nøst *et al.* 2015). The present study is the first comprehensive investigation of the current status of PAHs contamination in the surface water of the coastal area of the Bay of Bengal coast of Bangladesh. The main objectives of this study were to quantity the present levels, identify the seasonal-spatial distribution, potential sources and to assess the ecological risk of water borne PAHs in the coastal areas of Bangladesh. The data from this study may assist to have an insight of the present status of PAHs contamination, and for policy making related to ecological restoration and sustainable coastal zone management in the Bay of Bengal. This study may also give a direction for the future researches regarding the distribution and source-occurance relationship of PAHs in other coastal areas of the world.

2. Materials and methods

2.1 Study area and collection of samples

In order to explore the influence of the potential pollution sources, we investigated four coastal sites with fourteen 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 water samples were collected in winter (January-February) and summer (August-September) 2015. The sampling times represent two distinct seasons, winter (dry season) and summer (rainy season), respectively. Sampling was performed during low tide. Approximately 2 L of surface water were collected from each site in polypropylene (PP) bottles pre-cleaned with deionized water, methanol, acetone, and water from the particular site of sampling. The collected
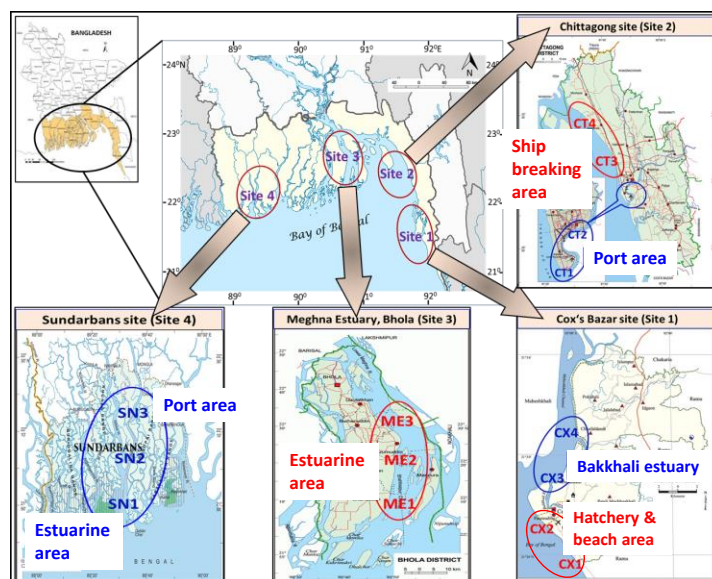
Fig. 1 Maps showing 4 sampling sites with 14 sampling locations in the coastal area of Bangladesh

samples were then filtered through 0.45 µm membranes to remove debris and transferred to new PP bottles. Samples were transported in ice-filled airtight insulating box to the laboratory of Fisheries Department of Dhaka University and stored at -8°C.

2.2 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-D_{10} (Ace-D_{10}) and Benzo[a]pyrene-D_{12} (BaP-D_{12})) 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, USA). All solvents (n-hexane, acetone, methanol and dichloromethane) used for sample processing and analysis were analytical grade and purchased from Wako Chemical (Osaka, Japan). Milli-Q (>18.2 MΩ) water generated from an ultrapure water purification system (Millipore, Billerica, MA, USA) was used throughout the experiment. Filter membranes (0.45 µm, 47 mm i.d.) were purchased from ADVANTEC® (Tokyo, Japan).

2.3 Sample pretreatment

Water samples were pretreated by solid phase extraction (SPE) followed by dispersive-SPE (d-

SPE) clean-up system. Before enrichment, ENVI-18 SPE cartridges were conditioned twice by 10 mL of dichloromethane, then twice by 10 mL of methanol and then 10 mL Milli-Q. One liter of filtered water was trapped through the SPE tubes at 10 mL/min flow rate under vacuum. The cartridges were then dried under vacuum for 10 min and kept in dark air-tight containers and transported to Yokohama National University in Japan for further analysis. The cartridges were eluted with 100 mL dichloromethane:n-hexane (1:1) followed by spiking with 100 µL of 500 ng/mL of Ace-D_{10} and BaP-D_{12} as an internal standards for quantification. The elution was concentrated to approximately 8 mL with a rotary evaporator. Afterwards, the concentrated elution was transferred to a d-SPE clean-up tube (15 mL) containing 0.9 g of anhydrous magnesium sulfate ($MgSO_4$), 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 vortexed for 1 minute and centrifuged at 3500 rpm for 5 minutes. A 5 mL aliquot of the supernatant was transferred into a glass test tube, and then the extract was evaporated to near dryness under a gentle stream of high-purity nitrogen gas, and the residue was re-dissolved in 1 mL n-hexane prior to its injection into the GC-MS/MS system.

2.4 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 m × 0.25 mm ID, 0.25 µm 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°C for 1 min, increased to 300°C for 4 min at 10°C min^{-1}, and finally held at 310°C for 2 min (total run time 31 min). The injection volume was set to one microliter (1 µL) 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°C, 300°C and 150°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. Prior to 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-D_{10} was used for Nap, Acel, Ace, Flu, Phe, Ant and BaP-D_{12} 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 liter (ng/L).

2.5 Quality assurance and quality control (QA/QC)

Strict quality control procedure was maintained during the experiments. The containers and equipment used during the whole procedure were pre-cleaned with methanol followed by acetone.

A (signal to noise) S/N ratio equal to or greater than 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 0.04 to 0.59 ng/L. 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 less than 10 (< 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/L, followed by similar extraction and analysis procedure as described in earlier sections. The mean recoveries of PAHs spiked into the water samples were 76%-114%. The detailed QA/QC data are given in Table S3.

2.7 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 $p = 0.05$ and concentrations less than LODs were set to LOD/2 (Succop *et al.* 2004). 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 water were shown by using MapViewer™ software (Version 8, Golden Software Inc., CO, USA).

3. Results and discussion

3.1 Concentration of PAHs in surface water and global comparison

All of the sixteen target PAHs were detectable in the exmined coastal surface waters of Bangladesh. The concentrations of PAHs are summarized in Table 1 and illustrated in Fig. 2, while the detailed data are presented in Table S4 and S5. The total PAH concentrations (∑PAHs; sum of 16 USEPA PAHs) ranged from 855.4 to 9653.7 ng/L (mean: 3319.6 ng/L; median: 1978.6 ng/L) in winter, and from 679.4 to 12639.3 ng/L in summer (mean: 4805.1 ng/L; median: 3306.8 ng/L) (Table 1). Among the PAHs analyzed, Nap, Flu, Phe, Ant, Flt, and Pyr were the most abundant compoundswith 100% detection frequencies in both seasons. The detection frequencies for the rest of the PAHs (Acel, Ace, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP) were in the range of 14% to 93%. The dominant PAH compounds were identified both by occurrence and abundance and the top three PAH compounds were Nap, Flu and Phe, comprising up to 36-89% (mean: 68%) of ∑PAHs. These three PAH compounds were highly correlated with ∑PAHs ($r = 0.96$; $p < 0.05$) and thus, well representing the ∑PAHs in the surface water of the Bangladeshi coastal area.

In particular, Phe was the most abundant PAH compound in winter with a contribution of 16-56% of ∑PAHs, whereas Nap was the most abundant PAH compound in summer contributing up to 10-57% of ∑PAHs. However, the dominance of these PAH compounds were also reported in the surface water from three estuaries in Hai River Basin of China (Yan *et al.* 2016), Yangpu Bay, China (Li *et al.* 2015), Danube River and its tributaries, Hungary (Nagy *et al.* 2014), Estero de Urias, estuary in Sinaloa, Mexico (Jaward *et al.* 2012).

Five of the seven carcinogenic PAHs (BaA, Chr, BbF, BkF, and BaP) with relatively high toxicity were detected in >50% of the samples in both seasons. In general, the total concentration
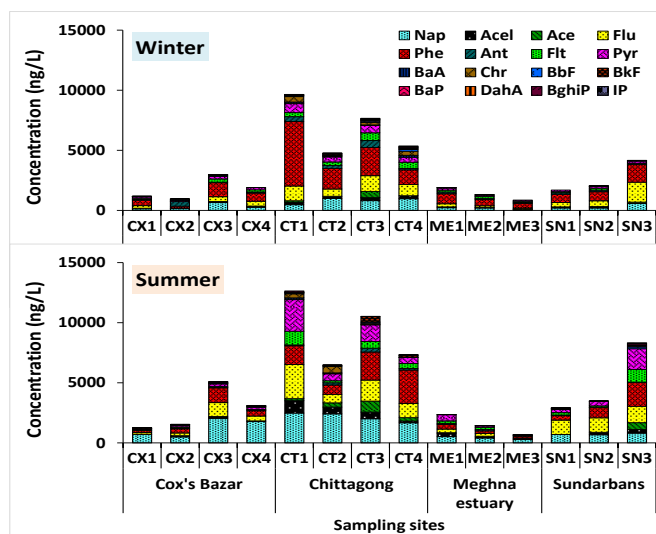
Fig. 2 Concentrations of PAHs in the surface water of the Bangladeshi coastal area in winter and summer

Table 1 Range, mean, median concentrations and detection frequencies of PAHs in water (ng/L) in two seasons (winter and summer) in the coastal area of Bangladesh

PAHs	Winter				Summer			
	Range	Mean	Median	D.F.[a]	Range	Mean	Median	D.F.
Nap	69.8–990.5	424.2	243.8	100	283.7–2487.7	1215.7	754.9	100
Acel	<LOD[b]–246.3	103.2	90.4	86	<LOD–1023.0	247.7	140.1	93
Ace	<LOD–511.4	54.9	0.03	21	<LOD–920.7	173.6	28.9	50
Flu	43.4–1659.0	582.1	393.7	100	80.9–2844.9	915.4	937.3	100
Phe	158.6–5362.4	1269.5	795.1	100	169.9–2776.2	974.1	612.7	100
Ant	29.9–576.9	179.8	115.8	100	16.6–314.5	84.7	60.9	100
Flt	25.9–642.5	225.9	165.0	100	38.7–1136.7	312.9	178.1	100
Pyr	82.7–746.6	260.7	181.0	100	22.1–2630.6	625.6	329.2	100
BaA	<LOD–164.6	38.1	10.3	86	<LOD–146.1	28.1	18.2	79
Chr	<LOD–455.8	80.3	7.6	64	<LOD–533.5	81.8	15.3	79
BbF	<LOD–142.9	30.6	0.1	43	<LOD–146.2	40.0	23.2	71
BkF	<LOD–64.6	15.2	1.0	50	<LOD–452.6	54.7	10.4	57
BaP	<LOD–77.7	19.9	6.7	50	<LOD–80.9	21.0	16.8	64
DahA	<LOD–78.8	11.5	0.01	43	<LOD–31.0	4.5	0.01	21
BghiP	<LOD–90.7	12.4	0.01	43	<LOD–111.3	18.2	0.01	29
IP	<LOD–56.5	11.2	0.01	43	<LOD–70.8	7.2	0.01	14
∑C-PAHs[c]	<LOD–848.1	206.7	52.7	100	5.2–719.6	237.2	131.1	100
∑PAHs[d]	855.4–9653.7	3319.6	1978.6	100	679.4–12639.3	4805.1	3306.8	100

[a]Detection frequency (%); *n=14* for each season, [b] Limit of detection, [c] Sum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA and IP) and [d] Sum of 16 USEPA PAHs; While calculating mean and median, values for <LOD were assigned to LOD/2 (Succop *et al.* 2004); Please refer to Table S3 for the LOD values of investigated PAHs

Table 2 Concentrations of total PAHs in water (ng/L) from various estuary and coastal regions in the world

Locations	Sampling year	N^a	∑PAHs	References
East and South China Seas	2005–05	16	30.40-120.29	Ren *et al*. (2010)
Daliao River estuary, China	2013	16	71.124255.43	Zheng *et al*. (2016)
Yangpu Bay, China	2013	16	582.8–2208.3 (W)[b]	Li *et al*. (2015)
			952.4–1201.7 (S)[b]	
Daya Bay, China	1999	16	4228–29325	Zhou and Maskaoui (2003)
Singapore's coastal waters	2005	16	2.7–46.2	Lim *et al*. (2007)
Gomti River, India	2004–06	16	60–84210	Malik *et al*. (2011)
Harbour line, Mumbai, India	2008	15	8660–46740	Dhananjayan *et al*. (2012)
Soan River, Pakistan	2013	16	61–207	Aziz *et al*. (2014)
Coastal areas of the Persian Gulf	2011	16	800–18340	Sinaei and Mashinchian (2014)
Densu River Basin, Ghana	2004	16	13–80	Amoako *et al*. (2011)
Brisbane River and Moreton Bay, Australia	2001–02	14	0.106–12	Shaw *et al*. (2004)
Mediterranean Sea, Sarno, Italy	2008	16	12.4–2321.1	Montuori and Triassi (2012)
Estero de Urias, estuary in Sinaloa, Mexico	2007	11	9–347	Jaward *et al*. (2012)
Coastal area of Bangladesh	2015	16	855.4–9653.7 (W)	This study
			679.4–12639.3 (S)	

[a] Number of PAHs and [b] W: Winter, S: Summer

of carcinogenic PAHs (∑C-PAHs) were <LOD–848.1 ng/L (mean: 206.7 ng/L) and 5.2-719.6 ng/L (mean: 237.2 ng/L), accounting 0-16% and 1-11% to the ∑PAHs in winter and summer, respectively (Table 1). Particularly, BaP (the best known potentially carcinogenic PAH) was detected in 50% and 64% samples in winter and summer, respectively. Due to absence of environmental thresholds set by the Government of Bangladesh, concentration of BaP was compared with China's Surface Water Environment Standard (GB 3838-2002) for BaP (2.8 ng/L) (Guo and Fang 2012) and the concentrations of BaP in the Bangladeshi coastal water exceeded the limited value in all cases, elucidating potential carcinogenic risk to the aquatic biota in the study area.

Table 2 compares the level of ∑PAHs in the surface water of the Bangladeshi coast of the Bay of Bengal with that in other riverine, estuarine and coastal areas around the world. In fact, the scientific literature of PAH levels in the coastal surface water is still scarce. In general, the ∑PAHs concentrations in the present study were comparable or lower than those measured in the coastal areas of the Persian Gulf (Sinaei and Mashinchian 2014), Gomti River Basin (Malik *et al*. 2011), Mumbai harbor line, India (Dhananjayan *et al*. 2012) and Daya Bay, China (Zhou and Maskaoui 2003). However, measured ∑PAHs concentrations in the Bangladeshi coastal area were far higher than those reported in surface water from the coastal areas of China (Ren *et al*. 2010, Li *et al*. 2015, Zheng *et al*. 2016), Singapore (Lim *et al*. 2007), Pakistan (Aziz *et al*. 2014), Ghana (Amoako *et al*. 2011), Australia (Shaw *et al*. 2004), Italy (Montuori and Triassi 2012), and Mexico (Jaward *et al*. 2012) (Table 2). Furthermore, the contamination of ∑PAHs in dissolved phases

could be classified into four grades: micro-polluted (10-50 ng/L); lightly polluted (50-250 ng/L); moderately polluted (250-1000 ng/L) and heavily polluted (>1000 ng/L) (Chen 2008, Li *et al*. 2015, Cao *et al*. 2010). Regardless of season, the concentrations of ∑PAHs in the Bangladeshi coastal surface water ranged from 679.4 to 12639.3 ng/L. Therefore, based on the global comparison along with the proposed contamination grades the surface water of the Bangladeshi coastal area could be classified as moderately to heavily polluted by PAHs.

3.2 PAH composition and source identification

The composition patterns and relative abundance of PAHs by the number of aromatic rings in the Bangladeshi coastal water in winter and summer are illustrated in Fig. 3. Interestingly, the compositions of PAHs in the investigated coastal sites are almost similar and did not varied significantly between seasons ($p > 0.05$). In particular, 2-3-ring PAHs were the dominant compounds accounting 65-92% and 61-90% of ∑PAHs in winter and summer, respectively, followed by 4-ring PAHs (8-27% in winter and 7-34% in summer). Five- and 6-ring PAHs contributed lesser percentages of the ∑PAHs and in some cases these PAHs were not detected (Table S4 and S5). The identified compositional pattern of dissolved phase PAHs is an indication of the presence of a relatively recent local source of PAHs in the study area (Fernandes *et al*. 1997, Liu *et al*. 2008, Song *et al*. 2013).

In general, regardless of source and season, the pattern of PAHs contamination on the basis of ring number were in order of 3-ring > 2-ring > 4-ring > 5-ring > 6-ring. The results are consistent with the typical PAHs composition in the surface water reported in other studies (Cao *et al*. 2010, Liu *et al*. 2013, Aziz *et al*. 2014, Sinaei and Mashinchian 2014, Li *et al*. 2015, Yan *et al*. 2016). The dominance of the low molecular weights (LMW) PAHs (2-3 rings) was attributed to their tendency of long range transport and high aqueous solubility (Aziz *et al*. 2014). On the contrary, high molecular weight (HMW) PAHs are resistant to degradation and with lower aqueous solubility they associate with particulate matter and eventually deposit in sediments (Nagy *et al*. 2014, Sinaei and Mashinchian 2014, Yun *et al*. 2016). Similar composition may be an indication of similar sources of PAH emission in the Bangladeshi coastal waters. In general, the LMW PAHs may originate from petrogenic sources such as, incomplete combustion of fossil fuel, petroleum products, and biomass, whereas HMW PAHs are mainly derived from pyrogenic sources, high temperature combustion, for example (Fernandes *et al*. 1997). The abundance of LMW PAHs in surface water signifies the importance of petrogenic sources in the Bangladeshi coastal area.

The ratio of the parent PAHs in the environmental samples is often used to identify the potential sources of PAHs (Zhang *et al*. 2003, Yunker *et al*. 2002, Katsoyiannis and Breivik 2014). In this study, the ratios of Flt/(Flt+Pyr) and Ant/(Ant+Phe) were used to diagnose the source of PAHs in the surface water (Fig. 4) and that have been effectively used to infer the sources of PAHs in the environment(Jiang *et al*. 2009, Xue *et al*. 2013, Martins *et al*. 2010). The ratios of Flt/(Flt+Pyr) ranged from 0.23 to 0.60 in winter and from 0.20 to 0.73 in summer. Irrespective to season, the ratios of Flt/(Flt+Pyr) were <0.4 (petroleum contamination) in 43%, 04-0.5 (combustion of petroleum and its by-products) in 25% and ≥0.5 (biomass combustion) in 32% of all samples. The values of Ant/(Ant+Phe) (0.02–0.71 in winter and 0.01-0.34 in summer) were ≥0.1 in 64% of the samples, suggesting that PAHs at these sites were mainly from pyrogenic source, whereas the rest of the samples (36%) had a value <0.1 indicating the petrogenic sources. The above data demonstrated that the mixed-type inputs from both combustion (pyrogenic) and petroleum (petrogenic) contributed to the PAHs pollution in the surface water of the Bangladeshi
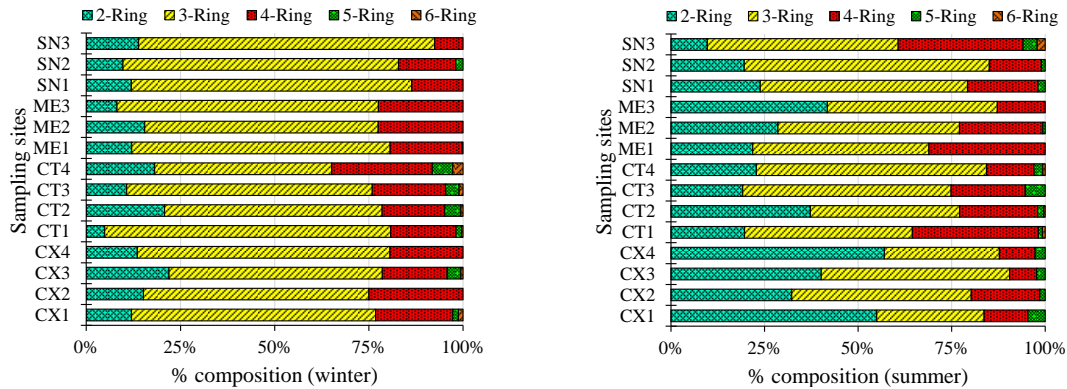
Fig. 3 Compositional profiles of PAH compounds in surface waters taken in winter and summer
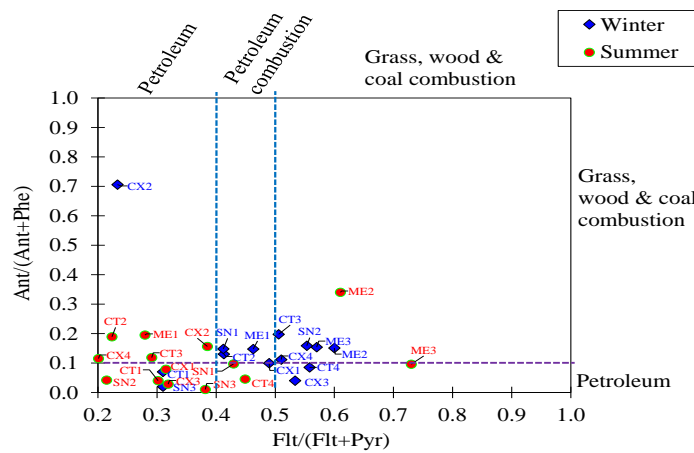
Fig. 4 PAH cross-correlations for the ratios of Flt/(Flt+Pyr) and Ant/(Ant+Phe)

coastal area.

3.3 Seasonal variations and spatial distributions of PAHs in the surface water

The seasonal variations and spatial distributions of PAHs in the surface water are presented in Fig. 5. Small variation but statistically not significant ($p > 0.05$) was recorded in the levels of water phase PAH between the two seasons. The concentration of ∑PAHs in summer samples was slightly higher than that in winter, suggesting a smaller amount of inputs during dry period.

Sources of pollution and precipitation might be the influential factors causing fluctuations in water quality. Previously accumulated PAHs in the surface soil from the contaminated sites in dry season can be flushed into the estuary and/or river with the surface runoff from heavy rains and floods in wet season. Furthermore, PAHs accumulation in the gas phase increase in summer, particularly in tropical regions (Abdel-Shafy *et al*. 2016) that could be redeposited to the surface water through the atmospheric wet and dry deposition. Our results, to some extent, were in agreement with that the warm water during summer can enhance the water solubility of PAHs (Shen *et al*. 2007, Song *et al*. 2013). Overall, the pattern of seasonal variation of PAHs
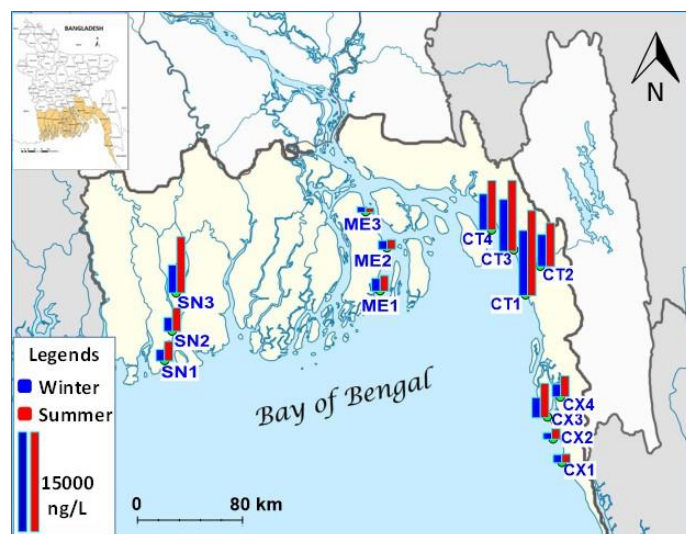
Fig. 5 Distribution of total PAHs in surface water of the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh

concentrations in this study was consistent with the seasonal variations in surface water from Tongzhou River Basin (Shen *et al*. 2007) and Taizi River Basin (Song *et al*. 2013), but contrary to the temporal variations of ∑PAHs concentrations in water from Yangpu Bay (Li *et al*. 2015) and Yellow River Estuary (Lang *et al*. 2008). These discrepancies might be attributed to the impact of regional hydrological conditions and local pollution sources among the study areas. However, the majority of the monitored PAHs in the Bangladeshi coastal area did not show clear seasonal variation with the exception of Nap, the level of which differed significantly between the two seasons ($p < 0.05$). The concentration of Nap was higher in summer (283.7-2487.7 ng/L) than in winter (69.8-990.5 ng/L). The high vapor pressure of Nap might be an influential factor to its association with the air phase (Abdel-Shafy *et al*. 2016). Therefore, Nap in the Bangladeshi coastal waters mainly come from atmospheric deposition during times of heavy rainfall, particularly in summer weather. However, in the present study, seasonal emissions from land based sources, such as seasonally operated industries or activities (e.g., Brick kiln, food and beverage factories, metal processing industries, etc.) may also influence the seasonal variations of this particular PAH, although the exact reason is still unknown.

Fig. 5 shows the **s**patial distribution of water phase PAHs in the Bay of Bengal coast of Bangladesh. Levels of PAHs in surface water 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. However, the distribution pattern of ∑PAHs between sites were more or less similar in winter and summer. Concentrations of ∑PAHs were higher in the coastal waters from Chittagong (average of 6862.2 and 9249.4 ng/L in winter and summer, respectively), Sundarbans (average of 2636.7 and 4916.8 ng/L in winter and summer, respectively) and Cox's Bazar (average of 1762.3 and 2760.6 ng/L in winter and summer, respectively) compared to that from Meghna Estuary (average of 1355.3 and 1493.9 ng/L in winter and summer, respectively). Therefore, the potential sources of PAHs are mainly located in these industrialized regions and that the amount of PAH emissions are associated with economic developments.

In particular, the highest levels of PAHs were recorded in water samples at location CT1

(9653.7 and 12639.3 ng/L in winter and summer, respectively) followed by CT3 (7662.5 and 10524.0 ng/L in winter and summer, respectively). Other two sites from Chittagong area, CT2 and CT4 also exhibited elevated PAH levels compared to other areas, indicating the existence of point source(s) in the area of Chittagong. CT1 and CT2 are located within the Chittagong port area which is the largest seaport in Bangladesh. Predominantly, thousands of boats and ships are travelling in this area 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 PAH (Zhou and Maskaoui 2003). Diesel leakage from ships and boats may have contributed to the high PAH levels in this area (Wang *et al*. 2016). CT3 and CT4 are located very close to Chittagong ship breaking yard. The existing mismanaged process of dismantling old and/or obsolete ships may produce various types of toxic and persistent pollutants, including PAHs (Neşer *et al*. 2012, Siddiquee *et al*. 2012). Various activities in ship breaking operations including asbestos removal, burning of electrical cables and plastic materials, blasting, discharges of ballast water are examples of PAH pollution sources (Hossain and Islam 2006, Sarraf *et al*. 2010). Nøst *et al*. (2015) found an elevated PAH levels in air at sites near the ship breaking activities in Chittagong which might be re-deposited from air to water through wet deposition and air/water exchange processes. Furthermore, there are hundreds of multipurpose industries located along the Chittagong coast producing, for example, fertilizers, rubber and plastic, paint, paper and pulp, pharmaceuticals, tobacco, printing and dyeing, steel products, automobile engines and electronics, jute and textiles, petroleum products, beverages, fish and tannery products, jewelry and plating. The discharge of untreated or semi-treated effluents from these industries including oil refinery factories may pose a significant contribution to the PAH contamination into the nearby environment. Dhananjayan *et al*. (2012) reported that the ship breaking activities and oil leaching from nearby industrial facilities lead to severe PAH contamination in Mumbai harbor in the Indian coast of the Arabian Sea.

Within Cox's Bazar, water samples from CX3 (Bakkhali Estuary) showed elevated concentration of ∑PAH (2988.5 and 5099.8 ng/L in winter and summer, respectively). It was expected because this site receives residential and industrial waste from the surrounding area, and the water is affected by activities such as intensive boating and fishing, which were identified as some potential contributing factors to the PAH contamination in the environment (Zhou and Maskaoui 2003, Li *et al.* 2016, Wang *et al*. 2016). Combustion or incomplete combustion of wood and wooden materials, and coal that are used by several small factories (one of the most prominent examples is brick kiln) and in some cases at households in nearby areas might also be a potential emission source of PAH. In addition, PAHs might also be emitted from the main municipal garbage dump located very close to the site CX3. Mostly the unusable consumer products including obsolete electronics are openly burnt at this site which may release substantial amount of PAHs in the adjacent areas.

In Sundarbans area, the highest concentration of ∑PAHs was recognized at SN3 (4161.4 and 8332.9 ng/L in winter and summer, respectively) which is located very close to Mongla port and fish landing center. There is high density of shipping and fishing activities in and around this area and hence high PAH levels in water are related to potential discharges from the ships and boats. Higher PAH levels might be attributed to the huge discharge of improperly treated effluents from numerous multipurpose industries such as cement, paint, paper, printing and dyeing, ship and boat repairing, plastics, etc. In addition, intense dredging operations in this area along with the dumping and burning of household wastes and resulted surface runoff and atmospheric depositions further aggravate the PAH pollution.

The levels of ∑PAHs in water taken from the Meghna estuary showed a downward increasing trend following to the bay. It is to be noted that the Meghna estuary is an exclusively unindustrialized area. Therefore, lower in concentration but detection of PAH in water from this site are related to non-point sources (e.g., surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc.). Moreover, the flow of a huge volume of water from the Ganges River of India to the Bay of Bengal through the Meghna Estuary may carry a substantial amount of PAHs (Chakraborty *et al.* 2014). In general, the levels of PAHs were higher in water samples from industrialized coastal sites (Chittagong, Cox's Bazar and Sundarbans) than those from the unindustrialized remote site (Meghna Estuary), and thus these compounds are associated to recent urbanization and industrialization.

3.4 Ecological risk assessment of water-borne PAHs

Risk quotient (RQ) approach was employed in this study to assess the potential ecological risk of PAHs on aquatic biota. The RQ was calculated by the ratio of PAH levels in water to their corresponding quality values (QV), which was displayed as follows

$$RQ = \frac{C_{PAHs}}{C_{QV}} \tag{1}$$

where C_{PAHs} is the concentration of certain PAHs in water samples and C_{QV} is the corresponding quality values of PAHs in surface water. In Bangladesh, no data regarding quality values exists for PAHs in surface water, so the negligible concentrations (NCs) and the maximum permissible concentrations (MPCs) of selected 10 PAHs (Nap, Flu, Phe, AntBaA, Chr, BkF, BaP, BghiP, and IP) in water reported by Kalf (1997) and other 6 PAHs (Acel, Ace, Flt, Pyr, BbF, and DahA) reported by Cao *et al.* (2010) were used as the quality values. Therefore, RQ_{NCs} and RQ_{MPCs} can be defined as follows

$$RQ_{NCs} = \frac{C_{PAHs}}{C_{QV(NCs)}} \tag{2}$$

$$RQ_{MPCs} = \frac{C_{PAHs}}{C_{QV(MPCs)}} \tag{3}$$

where $C_{QV(NCs)}$ is the quality values of the NCs of PAHs in water and $C_{QV(MPCs)}$ is the quality values of the MPCs of PAHs in water. This approach for the ecological risk assessment of water-borne PAHs were followed and recommended in several studies (Sun *et al.* 2009, Cao *et al.* 2010, Aziz *et al.* 2014, Zheng *et al.* 2016).

Risk classification of individual PAHs and ∑PAHs is presented in Table 3 (Cao *et al.* 2010). The mean values of RQ_{NCs} and RQ_{MPCs} of PAHs in the Bangladeshi coastal surface water in winter and summer are shown in Table 4 and the detailed data are provided in Table S6. The mean values of RQ_{MPCs} of Acel, Flu, Phe, Ant, Pyr, BaA, and BbF in winter, Nap, Acel, Ace, Flu, Phe, Ant, Flt, Pyr, BaA, BbF, and BkF in summer were all higher than 1, indicating that the biota in this ecosystem was at high risk and suffered from severe toxicity. The mean values of RQ_{MPCs} of other individual PAHs, such as Nap, Ace, Flt, Chr, BkF, BaP, DahA, BghiP, and IP in winter, Chr, BaP,

Table 3 Risk classification of individual PAHs and ∑PAHs (Cao *et al*. 2010)

Individual PAHs			∑PAHs		
	RQ_{NCs}	RQ_{MPCs}		$RQ_{\sum PAHs(NCs)}$	$RQ_{\sum PAHs(MPCs)}$
Risk-free	0		Risk-free	= 0	
			Low-risk	≥ 1; < 800	= 0
Moderate-risk	≥ 1	< 1	Moderate-risk$_1$	≥ 800	= 0
			Moderate-risk$_2$	< 800	≥ 1
High-risk		≥ 1	High-risk	≥ 800	≥ 1

Table 4 Mean values of RQ_{NCs} and RQ_{MPCs} of individual PAHs and total PAHs in surface water in the Bangladeshi coastal area

PAHs	QVs (ng/L)		Winter		Summer	
	NCs	MPCs	RQ_{NCs}	RQ_{MPCs}	RQ_{NCs}	RQ_{MPCs}
Nap	12	1200	35.3	0.4	101.3	1.1
Acel	0.7	70.0	147.5	1.5	353.9	3.5
Ace	0.7	70.0	78.5	0.8	248.0	2.5
Flu	0.7	70.0	831.6	8.3	1307.7	13.1
Phe	3.0	300.0	423.2	4.2	324.7	3.2
Ant	0.7	70.0	256.9	2.6	121.0	1.2
Flt	3.0	300.0	75.3	0.8	104.3	1.0
Pyr	0.7	70.0	372.5	3.7	893.7	8.9
BaA	0.1	10.0	381.5	3.8	281.2	2.8
Chr	3.4	340.0	23.6	0.2	24.0	0.2
BbF	0.1	10.0	305.2	3.1	399.4	4.0
BkF	0.4	40.0	38.0	0.4	136.7	1.4
BaP	0.5	50.0	39.7	0.4	42.0	0.4
DahA	0.5	50.0	23.0	0.2	9.0	0.1
BghiP	0.3	30.0	41.5	0.4	60.6	0.6
IP	0.4	40.0	27.9	0.3	18.0	0.2
∑PAHs			3101.2	31.0	4425.5	44.3

DahA, BghiP, and IP in summer were < 1 and RQ_{NCs} > 1, showing moderate risk to the ecosystems which should not be ignored as well.

In particular, Flu showed the highest mean RQ_{NCs} and RQ_{MPCs} both in winter and summer, suggesting a high ecological concern for this particular PAH compound in the study area. Besides, for all sites $RQ_{\sum PAHs(NCs)}$ >800 and $RQ_{\sum PAHs(MPCs)}$ > 1 except at ME3 (Table S6). Site ME3 exhibited the lowest concentrations of ∑PAHs in both seasons (855.4 ng/L in winter and 679.4 ng/L in summer). Therefore, the risk associated with ∑PAHs at all sites is of high level except for ME3 which is under moderate risk$_2$. Regarding the season, the ecological risk of ∑PAHs in summer was higher than that in winter. Overall, the results from the ecological risk assessment revealed that the aquatic ecosystem risk posed by the water-borne PAHs in the coastal area of Bangladesh is extremely high. Therefore, long term intensive water quality monitoring is suggested to develop

effective management strategies and that should be utilized effectively to control the contamination of PAHs in Bangladesh.

4. Conclusions

This study investigated 16 USEPA priority PAHs in the surface water from the coastal areas of Bangladesh and the level of contamination was assessed. The levels of PAHs ranged from moderate to a relatively high level of contamination compared to other coastal areas. The most abundant PAHs by ring structures were 2- to 3- ring, suggesting low to moderate molecular weight PAHs were prevalent in this area. Although slightly higher concentration of ∑PAHs was recorded in summer waters than those in winter, the variation was not statistically significant, suggesting inputs of PAHs from almost similar sources in the two distinct seasons. The source identification indicated that PAHs contaminations in the Bangladeshi coastal area were caused by 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, and thus relating these compounds with the recent urbanization and industrialization. The calculated risk quotient values indicate an extremely high ecological risk of PAH contamination in the surface waters of the Bay of Bengal coast of Bangladesh.

Acknowledgements

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Appendix

Description of sampling sites

Sampling sites were chosen in coastal areas to show the influence of the potential pollution sources (i.e., cities, industrial areas, rivers). The first site, Cox's Bazaar (Site 1), is a seaside tourist town with an unbroken 125 km world's longest natural sandy sea beach. Considering two diverse ecological aspects, it was divided into two sub sites: hatchery area (CX1-CX2) and Bakkhali estuary (CX3-CX4). More than 53 shrimp hatcheries and aquafarms, fish landing centers, huge hotels for the amusement of tourists and some industries are located in the hatchery site. Likewise, the Bakkhali estuary, regarded as an important economic part of Cox's Bazar district with a harbor and imperative local fishery, is about 0.5 km wide and >10 m deep at its mid-point and directly influenced by semi-diurnal tides. Moreover, Maheshkhali Island is one of the important tourist attracting economic zone incorporating to Bakkhali estuary through 9 to 11 kilometer long Maheshkhali channel to Cox's Bazaar central island. Consequently, these sampling areas are mostly influenced by the municipal sewage and industrial wastage discharged from these unplanned industries, hatcheries, and aquaculture farms. The second site, Chittagong (Site 2), is located near the Chittagong port (CT1-CT2) and ship breaking area (CT3-CT4). This is the southeastern principal seaport region of the country, where a significant ecological change is pronounced due to huge discharges of untreated or semi treated domestic and municipal sewage as well as effluents from multifarious industries (namely, paper and pulp, tanneries, textile, chemicals, pharmaceuticals, rubber and plastic, oil refinery, steal rerolling, leather, jute, tobacco, fish processing plants, paint, coal-based thermal power plants, fertilizer, rechargeable batteries, jewelry, plating, automobile engine and electronics industries, etc.,) as well as contaminated mud disposal from harbor dredging. Besides, Chittagong ship breaking yard is world's second largest ship breaking area confined to 18 km2 area along the coast of Sitakunda Upazilla, particularly Bhatiary to Kumira in Chittagong division. While dismantling the ships, the industry generates a huge organic and inorganic wastes which is discharged into the nearby area and thus polluting the coastal water and sediment. The third site (ME1-ME3), Meghna Estuary (Site 3), is an estuarine area where the main rivers mix together to the Bay of Bengal. This site is influenced mainly by the domestic and industrial effluents carried by the inland rivers from the country and trans-boundary countries. The fourth site, Sundarbans (Site 4), is located near the southwest part of the coastal area which is regarded as a large mangrove ecosystem in Bangladesh. This sampling area (SN1-SN3) is also mostly influenced by different anthropogenic and industrial activities like cement factories, export processing zone, sea port, paper industries, oil refinery industries, steal rerolling, fertilizer industry, hatcheries and aquafarms, fish processing industries, leather industries, dyeing industries, paint industries etc. in Khulna and Mongla area. However, in Bangladesh context, no reliable data is available regarding how much the industries contribute to the coastal pollution.

Table S1 Basic information of sampling sites of the present study

Sampling sites	Location ID	Location		Site description
		Latitude	Longitude	
Cox's Bazar (Site 1)	CX1	21°13'22"N	92°01'58"E	Hatchery area; Many small and big industries
	CX2	21°22'06"N	92°00'18"E	Beach area; Tourism
	CX3	21°28'50"N	91°58'17"E	Bakhhali estuary, Near airport, Fish landing center

Table S1 Continued

Sampling sites	Location ID	Location		Site description
		Latitude	Longitude	
Cox's Bazar (Site 1)	CX4	21°30'57"N	91°58'55"E	Maheshkhali channel
Chittagong (Site 2)	CT1	22°13'22"N	91°48'08"E	Port area; Karnaphuli river estuary
	CT2	22°19'48"N	91°51'48"E	Port activities; Karnaphuli river flow
	CT3	22°26'22"N	91°43'39"E	Ship breaking area
	CT4	22°34'30"N	91°37'59"E	Ship breaking area
Meghna Estuary, Bhola (Site 3)	ME1	22°06'41"N	90°48'24"E	Mouth of estuary
	ME2	22°24'12"N	90°51'31"E	Mid estuary
	ME3	22°33'37"N	90°45'12"E	Upper estuary
Sundarbans (Site 4)	SN1	21°48'47"N	89°28'44"E	Estuarine area
	SN2	21°59'39"N	89°31'27"E	Upper estuary; Industrial zone
	SN3	22°13'52"N	89°34'05"E	Industrial area; Port area

Table S2 Instrumental characteristics and parameters used for the analytical determination of PAHs

GC analysis conditions				
GC	Agilent 7890A GC			
Column)	DB-5MS 30 m×0.25 mm I.D, Thickness 0.25 μm			
Oven Temperature	70°C(1 min)-10°C/min-300°C(4min)-10°C/min-310°C(2min)			
Inlet Temperature	300°C			
Carrier gas and flow rate	Helium 1.2 mL/min (constant flow)			
Injection	1 μL, Splitless			
MS/MS analysis conditions				
MS/MS	7000C			
Collision gas and flow rate	Nitrogen, 1.5mL/min			
Quenching gas and flow rate	Helium, 2.25mL/min			
Ionization method	EI 70eV			
Transfer line temperature	320°C			
Ion source temperature	300°C			
Quadrupole temperature	150°C			
Tuning	Auto Tune			
Mode of operation	Multiple reactions monitoring (MRM), Gain 10			
MRM parameters				
Compounds	MS1(m/z)	MS2(m/z)	CE(eV)	Dwell Time (msec)
Nap	128	102, 78	20, 20	25
Acel	152	151, 126	20, 30	25

Table S2 Continued

MRM parameters				
Compounds	MS1(m/z)	MS2(m/z)	CE(eV)	Dwell Time (msec)
Ace	154	152, 162	25, 20	25
Ace-D$_{10}$	164	162, 160	30, 30	25
Flu	166	165, 164	20, 35	15
Phe	178	152, 151	15, 40	15
Ant	178	152, 151	15, 40	15
Flt	202	201, 200	30, 45	15
Pyr	202	201, 200	30, 45	15
BaA	228	226, 202	30, 30	40
Chr	228	226, 202	30, 30	40
BbF	252	250, 226	30, 47	75
BkF	252	250, 226	30, 47	75
BaP-D$_{12}$	264	260, 236	30, 30	75
BaP	252	250, 226	30, 47	75
DahA	278	276, 274	35, 55	75
BghiP	276	274, 248	40, 50	75
IP	276	274, 250	40, 50	75

Table S3 Recoveries*, LODs, LOQs and linearity of calibration curves for the analytical determination of PAHs

PAHs	% Recoveries [mean (RSD)] (n=3)	LOD[a] (ng/L)	LOQ[b] (ng/L)	Linearity (r^2)[c]
Nap	97 (6)	0.05	0.15	0.9986
Acel	114 (4)	0.1	0.29	0.9999
Ace	98 (1)	0.05	0.14	0.9999
Flu	103 (4)	0.11	0.34	0.9999
Phe	94 (4)	0.16	0.48	0.9997
Ant	84 (5)	0.1	0.32	0.9999
Flt	86 (4)	0.04	0.12	0.9992
Pyr	107 (6)	0.05	0.15	0.9993
BaA	93 (5)	0.03	0.09	0.9999
Chr	103 (4)	0.02	0.06	0.9999
BbF	94 (2)	0.12	0.36	0.9998
BkF	111 (7)	0.12	0.36	0.9992
BaP	88 (9)	0.19	0.59	0.9996
DahA	103 (4)	0.01	0.04	0.9996
BghiP	76 (4)	0.02	0.07	0.9998
IP	109 (7)	0.01	0.04	0.9998

[a] Limit of detection; [b] Limit of quantification; [c] Calibration curves (1-10000 µg/L for each compound); * The

recovery of spiked PAHs was calculated using the following equation

$$\text{Spike recovery rate (\%)} = (C_{sample + spiked} - C_{sample}) / C_{spiked} \times 100$$

where $C_{sample + spiked}$ is the concentration of PAHs in a spiked sample, C_{sample} is the concentration of PAHs in the sample (same as above without spiking target compounds, C_{spiked} is the concentration of the spiked target PAHs (10 ng/L).

Table S4 Concentrations (ng/L) of 16 USEPA priority PAHs in surface water from the coastal area of Bangladesh in winter 2015

PAHs	PAH Ring	Sites													
		CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
Nap	2	141.9	147.9	655.8	257.7	470.3	990.5	819.3	973.3	229.9	202.3	69.8	203.5	199.6	576.9
Acel		45.2	<LOD[a]	93.5	102.1	194.0	181.3	246.3	242.3	61.6	36.6	38.2	87.4	116.7	<LOD
Ace		<LOD	<LOD	<LOD	<LOD	143.0	<LOD	511.4	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	114.6
Flu	3	208.8	43.4	391.9	395.6	1225.4	617.7	1311.4	951.9	260.4	141.1	82.0	369.9	491.1	1659.0
Phe		461.3	158.6	1158.2	692.3	5362.4	1701.5	2348.4	1211.6	841.2	534.4	400.4	687.9	749.0	1466.2
Ant		50.9	380.4	48.3	86.8	403.6	255.0	576.9	112.5	145.3	94.9	72.8	119.2	141.2	29.9
Flt		114.5	25.9	272.7	187.2	335.4	288.5	642.5	519.4	156.8	170.3	109.6	95.4	159.7	84.4
Pyr	4	119.3	85.3	238.4	179.9	746.6	410.0	628.0	411.3	182.1	113.5	82.7	136.2	128.8	187.8
BaA		4.3	94.0	3.1	2.4	136.2	52.0	29.0	164.6	14.3	6.3	<LOD	<LOD	22.7	5.1
Chr		4.0	38.4	<LOD	<LOD	455.8	43.3	196.0	334.3	11.3	3.1	<LOD	<LOD	<LOD	37.4
BbF		<LOD	<LOD	49.5	<LOD	64.7	31.0	120.1	142.9	<LOD	<LOD	<LOD	<LOD	19.1	<LOD
BkF	5	2.0	<LOD	17.1	<LOD	23.8	44.4	52.8	64.6	<LOD	<LOD	<LOD	<LOD	7.9	<LOD
BaP		15.9	<LOD	27.2	<LOD	32.9	47.4	63.9	77.7	<LOD	<LOD	<LOD	<LOD	13.3	<LOD
DahA		<LOD	<LOD	14.1	<LOD	18.7	78.8	37.1	7.4	5.0	<LOD	<LOD	<LOD	<LOD	<LOD
BghiP	6	8.1	<LOD	0.0	<LOD	26.1	15.2	34.0	90.7	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
IP		6.6	<LOD	18.6	<LOD	14.8	14.6	45.2	56.5	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑2-Ring PAHs		141.9	147.9	655.8	257.7	470.3	990.5	819.3	973.3	229.9	202.3	69.8	203.5	199.6	576.9
∑3-Ring PAHs		766.3	582.5	1691.8	1276.8	7328.4	2755.5	4994.5	2518.3	1308.6	807.1	593.3	1264.4	1498.1	3269.8
∑4-Ring PAHs		242.1	243.6	514.2	369.5	1674.0	793.8	1495.5	1429.7	364.5	293.2	192.3	231.6	311.2	314.7
∑5-Ring PAHs		17.9	<LOD	108.0	<LOD	140.1	201.6	274.0	292.6	5.0	<LOD	<LOD	<LOD	40.3	<LOD
∑6-Ring PAHs		14.7	<LOD	18.6	<LOD	40.9	29.9	79.2	147.3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑LMW-PAHs[b]		908.1	730.4	2347.7	1534.5	7798.7	3746.1	5813.8	3491.7	1538.5	1009.3	663.1	1467.9	1697.6	3846.7
∑HMW-PAHs[c]		274.6	243.6	640.8	369.5	1855.0	1025.2	1848.7	1869.6	369.6	293.2	192.3	231.6	351.5	314.7
∑C-PAHs[d]		32.8	132.4	129.7	2.4	746.8	311.5	544.1	848.1	30.6	9.3	<LOD	<LOD	63.0	42.5
∑PAHs[e]		1182.7	973.9	2988.5	1904.0	9653.7	4771.3	7662.5	5361.3	1908.1	1302.5	855.4	1699.5	2049.1	4161.4

[a]Limit of detection, [b]Sum of low molecular weight PAHs (Nap, Acel, Ace, Flu, Phe, and Ant), [c]Sum of high molecular weight PAHs (Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP), [d]Sum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA and IP) and [e] Sum of 16 USEPA PAHs

Table S5 Concentrations (ng/L) of 16 USEPA priority PAHs in surface water from the coastal area of Bangladesh in summer 2015

PAHs	PAH Ring	Sites													
		CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
Nap	2	701.2	499.1	2045.6	1778.4	2487.7	2417.0	2015.7	1675.6	515.6	411.7	283.7	697.0	683.4	808.7
Acel		21.9	99.6	78.4	50.3	1023.0	559.9	545.8	138.8	274.5	141.5	39.7	<LOD	196.4	298.1
Ace		<LOD[a]	<LOD	64.5	<LOD	172.1	353.6	920.7	285.8	57.8	<LOD	<LOD	<LOD	<LOD	575.3
Flu	3	134.3	229.1	1169.3	411.7	2844.9	705.2	1738.6	1184.7	308.6	238.2	80.9	1196.9	1209.2	1364.2
Phe		193.7	349.3	1220.0	439.2	1554.8	786.2	2341.4	2776.2	378.5	210.5	169.9	380.0	845.8	1992.4
Ant		16.6	64.7	35.4	57.1	64.8	183.4	314.5	132.4	91.7	108.6	18.1	40.8	37.3	20.1
Flt		38.7	93.3	95.5	49.2	1136.7	164.8	565.4	412.4	200.7	191.5	59.8	207.1	104.8	1060.5
Pyr	4	84.3	149.0	204.2	195.9	2630.6	572.2	1377.6	506.3	517.3	122.4	22.1	275.1	383.2	1718.4
BaA		16.0	20.9	26.4	23.6	146.1	79.7	45.1	4.1	6.2	<LOD	5.2	20.4	<LOD	<LOD
Chr		9.8	20.2	42.0	28.3	341.0	533.5	95.9	8.3	10.3	7.1	<LOD	48.1	<LOD	<LOD
BbF		<LOD	13.5	47.7	32.5	82.8	70.9	97.2	21.9	<LOD	<LOD	<LOD	24.3	22.2	146.2
BkF	5	59.6	9.1	33.7	26.7	<LOD	<LOD	452.6	117.0	<LOD	<LOD	<LOD	11.7	<LOD	55.1
BaP		<LOD	<LOD	37.0	25.7	39.0	35.4	<LOD	33.4	<LOD	9.3	<LOD	20.9	12.6	80.9
DahA		<LOD	<LOD	<LOD	<LOD	18.4	<LOD	13.6	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	31.0
BghiP	6	<LOD	<LOD	<LOD	<LOD	97.4	26.6	<LOD	19.0	<LOD	<LOD	<LOD	<LOD	<LOD	111.3
IP		<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	29.9	<LOD	<LOD	<LOD	<LOD	<LOD	70.8
∑2-Ring PAHs		701.2	499.1	2045.6	1778.4	2487.7	2417.0	2015.7	1675.6	515.6	411.7	283.7	697.0	683.4	808.7
∑3-Ring PAHs		366.6	742.7	2567.7	958.3	5659.6	2588.2	5861.0	4517.9	1111.2	698.8	308.5	1617.7	2288.6	4250.2
∑4-Ring PAHs		148.8	283.5	368.0	297.1	4254.4	1350.3	2083.9	931.2	734.5	321.1	87.1	550.8	488.0	2778.8
∑5-Ring PAHs		59.6	22.5	118.5	84.9	140.2	106.3	563.4	172.2	<LOD	9.3	<LOD	56.9	34.8	313.1
∑6-Ring PAHs		<LOD	<LOD	<LOD	<LOD	97.4	26.6	<LOD	48.9	<LOD	<LOD	<LOD	<LOD	<LOD	182.1
∑LMW-PAHs[b]		1067.7	1241.8	4613.3	2736.7	8147.3	5005.2	7876.7	6193.5	1626.8	1110.6	592.2	2314.7	2972.1	5058.9
∑HMW-PAHs[c]		208.4	306.0	486.5	381.9	4492.0	1483.2	2647.4	1152.4	734.5	330.4	87.1	607.7	522.8	3274.1
∑C-PAHs[d]		85.4	63.7	186.8	136.8	627.4	719.6	704.4	214.6	16.4	16.4	5.2	125.5	34.8	383.9
∑PAHs[e]		1276.1	1547.8	5099.8	3118.6	12639.3	6488.4	10524.0	7345.8	2361.3	1440.9	679.4	2922.4	3494.9	8332.9

[a] Limit of detection, [b] Sum of low molecular weight PAHs (Nap, Acel, Ace, Flu, Phe, and Ant), [c] Sum of high molecular weight PAHs (Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP), [d] Sum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA and IP) and [e] Sum of 16 USEPA PAHs

Table S6 RQ_{NCs} and RQ_{MPCs} of individual and total PAHs in water

RQ	Seasons	Sites	Nap	Acel	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	IP	∑PAHs
RQ_{NCs}	Winter																		
		CX1	11.8	64.6	0.0	298.3	153.8	72.8	38.2	170.5	43.0	1.2	0.0	5.0	31.8	0.0	26.9	16.6	934.3
		CX2	12.3	0.0	0.0	62.1	52.9	543.4	8.6	121.8	940.3	11.3	0.0	0.0	0.0	0.0	0.0	0.0	1752.7
		CX3	54.7	133.6	0.0	559.9	386.1	68.9	90.9	340.6	31.0	0.0	495.4	42.9	54.4	28.1	0.0	46.6	2333.0
		CX4	21.5	145.9	0.0	565.1	230.8	124.0	62.4	257.0	24.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1430.6

Table S6 Continued

RQ	Seasons	Sites	Nap	Acel	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	IP	∑PAHs
RQ$_{NCs}$	Winter																		
		CT1	39.2	277.2	204.2	1750.6	1787.5	576.6	111.8	1066.6	1361.6	134.1	646.7	59.6	65.8	37.3	87.0	36.9	8242.7
		CT2	82.5	259.0	0.0	882.4	567.2	364.3	96.2	585.7	520.4	12.7	309.6	111.1	94.7	157.6	50.8	36.6	4130.9
		CT3	68.3	351.8	730.6	1873.5	782.8	824.1	214.2	897.2	289.6	57.6	1201.2	132.0	127.8	74.3	113.5	113.0	7851.4
		CT4	81.1	346.2	0.0	1359.9	403.9	160.7	173.1	587.6	1646.5	98.3	1429.1	161.5	155.3	14.9	302.4	141.4	7061.9
		ME1	19.2	88.1	0.0	372.1	280.4	207.5	52.3	260.2	143.0	3.3	0.0	0.0	0.0	10.1	0.0	0.0	1436.1
		ME2	16.9	52.3	0.0	201.6	178.1	135.5	56.8	162.2	62.7	0.9	0.0	0.0	0.0	0.0	0.0	0.0	867.1
		ME3	5.8	54.6	0.0	117.1	133.5	103.9	36.5	118.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	569.6
		SN1	17.0	124.8	0.0	528.4	229.3	170.3	31.8	194.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1296.2
		SN2	16.6	166.7	0.0	701.6	249.7	201.7	53.2	184.1	227.2	0.0	191.2	19.7	26.5	0.0	0.0	0.0	2038.2
		SN3	48.1	0.0	163.7	2370.1	488.7	42.7	28.1	268.3	51.0	11.0	0.0	0.0	0.0	0.0	0.0	0.0	3471.8
	Summer																		
		CX1	58.4	31.3	0.0	191.9	64.6	23.7	12.9	120.4	160.0	2.9	0.0	149.0	0.0	0.0	0.0	0.0	815.1
		CX2	41.6	142.2	0.0	327.3	116.4	92.5	31.1	212.9	209.1	5.9	134.6	22.7	0.0	0.0	0.0	0.0	1336.3
		CX3	170.5	112.1	92.2	1670.5	406.7	50.6	31.8	291.6	263.6	12.4	477.4	84.3	74.0	0.0	0.0	0.0	3737.6
		CX4	148.2	71.9	0.0	588.2	146.4	81.6	16.4	279.9	236.0	8.3	324.8	66.7	51.4	0.0	0.0	0.0	2019.7
		CT1	207.3	1461.4	245.9	4064.2	518.3	92.5	378.9	3758.0	1461.4	100.3	828.1	0.0	77.9	36.9	324.8	0.0	13555.9
		CT2	201.4	799.8	505.1	1007.4	262.1	262.0	54.9	817.5	797.3	156.9	708.7	0.0	70.9	0.0	88.6	0.0	5732.5
		CT3	168.0	779.7	1315.3	2483.7	780.5	449.3	188.5	1967.9	451.2	28.2	972.3	1131.4	0.0	27.2	0.0	0.0	10743.2
		CT4	139.6	198.2	408.4	1692.4	925.4	189.1	137.5	723.3	41.0	2.4	218.8	292.4	66.7	0.0	63.4	74.8	5173.6
		ME1	43.0	392.2	82.6	440.9	126.2	131.0	66.9	738.9	61.6	3.0	0.0	0.0	0.0	0.0	0.0	0.0	2086.2
		ME2	34.3	202.2	0.0	340.2	70.2	155.1	63.8	174.9	0.0	2.1	0.0	0.0	18.6	0.0	0.0	0.0	1061.5
		ME3	23.6	56.7	0.0	115.5	56.6	25.8	19.9	31.6	52.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	381.8
		SN1	58.1	0.0	0.0	1709.9	126.7	58.3	69.0	393.1	204.0	14.2	243.2	29.2	41.8	0.0	0.0	0.0	2947.5
		SN2	57.0	280.5	0.0	1727.4	281.9	53.3	34.9	547.5	0.0	0.0	221.8	0.0	25.3	0.0	0.0	0.0	3229.5
		SN3	67.4	425.9	821.9	1948.9	664.1	28.8	353.5	2454.8	0.0	0.0	1462.3	137.6	161.7	61.9	371.1	176.9	9136.9
RQ$_{MPCs}$	Winter																		
		CX1	0.1	0.6	0.0	3.0	1.5	0.7	0.4	1.7	0.4	0.0	0.0	0.0	0.3	0.0	0.3	0.2	9.3
		CX2	0.1	0.0	0.0	0.6	0.5	5.4	0.1	1.2	9.4	0.1	0.0	0.0	0.0	0.0	0.0	0.0	17.5
		CX3	0.5	1.3	0.0	5.6	3.9	0.7	0.9	3.4	0.3	0.0	5.0	0.4	0.5	0.3	0.0	0.5	23.3
		CX4	0.2	1.5	0.0	5.7	2.3	1.2	0.6	2.6	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	14.3
		CT1	0.4	2.8	2.0	17.5	17.9	5.8	1.1	10.7	13.6	1.3	6.5	0.6	0.7	0.4	0.9	0.4	82.4
		CT2	0.8	2.6	0.0	8.8	5.7	3.6	1.0	5.9	5.2	0.1	3.1	1.1	0.9	1.6	0.5	0.4	41.3
		CT3	0.7	3.5	7.3	18.7	7.8	8.2	2.1	9.0	2.9	0.6	12.0	1.3	1.3	0.7	1.1	1.1	78.5
		CT4	0.8	3.5	0.0	13.6	4.0	1.6	1.7	5.9	16.5	1.0	14.3	1.6	1.6	0.1	3.0	1.4	70.6
		ME1	0.2	0.9	0.0	3.7	2.8	2.1	0.5	2.6	1.4	0.0	0.0	0.0	0.0	0.1	0.0	0.0	14.4
		ME2	0.2	0.5	0.0	2.0	1.8	1.4	0.6	1.6	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	8.7
		ME3	0.1	0.5	0.0	1.2	1.3	1.0	0.4	1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	5.7

Table S6 Continued

RQ	Seasons	Sites	Nap	Acel	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	IP	∑PAHs
RQ$_{NCs}$	Winter																		
		SN1	0.2	1.2	0.0	5.3	2.3	1.7	0.3	1.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	13.0
		SN2	0.2	1.7	0.0	7.0	2.5	2.0	0.5	1.8	2.3	0.0	1.9	0.2	0.3	0.0	0.0	0.0	20.4
		SN3	0.5	0.0	1.6	23.7	4.9	0.4	0.3	2.7	0.5	0.1	0.0	0.0	0.0	0.0	0.0	0.0	34.7
	Summer																		
		CX1	0.6	0.3	0.0	1.9	0.6	0.2	0.1	1.2	1.6	0.0	0.0	1.5	0.0	0.0	0.0	0.0	8.2
		CX2	0.4	1.4	0.0	3.3	1.2	0.9	0.3	2.1	2.1	0.1	1.3	0.2	0.0	0.0	0.0	0.0	13.4
		CX3	1.7	1.1	0.9	16.7	4.1	0.5	0.3	2.9	2.6	0.1	4.8	0.8	0.7	0.0	0.0	0.0	37.4
		CX4	1.5	0.7	0.0	5.9	1.5	0.8	0.2	2.8	2.4	0.1	3.2	0.7	0.5	0.0	0.0	0.0	20.2
		CT1	2.1	14.6	2.5	40.6	5.2	0.9	3.8	37.6	14.6	1.0	8.3	0.0	0.8	0.4	3.2	0.0	135.6
		CT2	2.0	8.0	5.1	10.1	2.6	2.6	0.5	8.2	8.0	1.6	7.1	0.0	0.7	0.0	0.9	0.0	57.3
		CT3	1.7	7.8	13.2	24.8	7.8	4.5	1.9	19.7	4.5	0.3	9.7	11.3	0.0	0.3	0.0	0.0	107.4
		CT4	1.4	2.0	4.1	16.9	9.3	1.9	1.4	7.2	0.4	0.0	2.2	2.9	0.7	0.0	0.6	0.7	51.7
		ME1	0.4	3.9	0.8	4.4	1.3	1.3	0.7	7.4	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	20.9
		ME2	0.3	2.0	0.0	3.4	0.7	1.6	0.6	1.7	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.0	10.6
		ME3	0.2	0.6	0.0	1.2	0.6	0.3	0.2	0.3	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	3.8
		SN1	0.6	0.0	0.0	17.1	1.3	0.6	0.7	3.9	2.0	0.1	2.4	0.3	0.4	0.0	0.0	0.0	29.5
		SN2	0.6	2.8	0.0	17.3	2.8	0.5	0.3	5.5	0.0	0.0	2.2	0.0	0.3	0.0	0.0	0.0	32.3
		SN3	0.7	4.3	8.2	19.5	6.6	0.3	3.5	24.5	0.0	0.0	14.6	1.4	1.6	0.6	3.7	1.8	91.4

Table S7 Physical and chemical characteristics of surface water collected from the coastal area of Bangladesh

Season	Site ID	pH	Temperature (°C)	Salinity (‰)	TSS (mg/L)
Winter	CX1	7.3	19.5	17.3	465
	CX2	6.2	20	18.4	340
	CX3	7.2	20.5	19.6	490
	CX4	7.1	20.5	18.6	435
	CT1	6.6	19.5	21.3	280
	CT2	6.3	21	19.5	310
	CT3	6.7	22	17.4	230
	CT4	6.5	21.5	18.6	260
	ME1	7.7	19	16.8	800
	ME2	7.1	19.5	20.4	650
	ME3	7.8	22	21.6	460
	SN1	6.9	19	17.3	850
	SN2	7.6	21	18.6	730
	SN3	6.9	23	19.8	960

Table S7 Continued

Season	Site ID	pH	Temperature (°C)	Salinity (‰)	TSS (mg/L)
Summer	CX1	7.2	24.5	22.5	500
	CX2	6.5	23.3	24.0	350
	CX3	6.8	24.1	12.5	480
	CX4	7.1	23.5	16.5	460
	CT1	6.5	22.3	13.5	350
	CT2	6.2	23.8	15.5	440
	CT3	6.3	23.6	17.0	250
	CT4	6.5	23.5	18.5	290
	ME1	7.5	22	4.5	850
	ME2	6.8	24.3	3.5	630
	ME3	7.3	23.6	7.0	520
	SN1	6.5	23.5	13.5	900
	SN2	6.2	24.4	14.0	750
	SN3	5.5	24.9	16.5	850