Doctoral Dissertation

博士論文

MONITORING AND ASSESSMENT OF PERSISTENT ORGANIC POLLUTANTS (POPs) IN WATER, SEDIMENT, AND SEAFOOD FROM THE COASTAL AREAS OF BANGLADESH

バングラデシュ沿岸で採取した海水、堆積物、および海産食品中の残留性有機汚染物質（POPs）のモニタリングと評価

By

MD. HABIBULLAH-AL-MAMUN

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Yokohama National University

国立大学法人　横浜国立大学
大学院環境情報学府

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Declaration

I hereby declare that except for references to other peoples' work, which have been duly acknowledged, this work is original and was carried out by me, Md. Habibullah-Al-Mamun, under the supervision of Prof. Shigeki Masunaga.

MD. HABIBULLAH-AL-MAMUN Date

(PhD candidate)

PROF. SHIGEKI MASUNAGA Date

(Supervisor)

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Md. Habibullah-Al-Mamun
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List of Abbreviations and Acronyms

AA-QS	Annual Average Quality Standard
AF	Assessment factor
AFFFs	Aqueous film forming foams
ANOVA	Analysis of variance
AWV	Avian wildlife value
BC	Black carbon
BH	Bhola
$BMDL_{10}$	Benchmark dose lower confidence limit for a 10% response
BoB	Bay of Bengal
BOBLM	Bay of Bengal Large Marine Ecosystem
C18EC	Octadecyl (endcapped) functionalized silica
CCC	Criteria continuous concentration
CCME	Canadian Council of Ministers of the Environment
CMC	Criteria maximum concentration
CT	Chittagong
CX	Cox’s Bazar
DoE	Department of Environment
d-SPE	Dispersive solid phase extraction
E/LC_{50}	Median effect/lethal concentration
EC	European Commission
EDI	Estimated daily intake
EMR-Lipid	Enhanced Matrix Removal-Lipid
EOCs	Emerging organic contaminants
EqP	Equilibrium partitioning
EQS	Environmental Quality Standards
ERA	Ecological or environmental risk assessment
ERL	Effect range low
ERM	Effect range median
ESI	Electrospray ionization
E-waste	Electronic waste
FDA	Food and Drug Administration
GC-MS/MS	Gas chromatography tandem-mass spectrometry
HMW	High molecular weight
HQ	Hazard quotient
HRGC	High resolution gas chromatography
HRMS	High resolution mass spectrometry
IARC	International Agency for Research on Cancer
ICES	International Council for the Exploration of the Sea
ILCR	Incremental lifetime cancer risk
IS	Internal Standard
ISQG	Interim sediment quality guideline
LC-MS/MS	Liquid chromatography tandem-mass spectrometry
LELs	Low effect limits
LMW	Low molecular weight
LOD	Limit of detection
LOQ	Limit of quantification
ME	Meghna Estuary

MEC	Measured environmental concentration
MPCs	Maximum permissible concentrations
MRM	Multiple Reaction Monitoring
MT	Metric ton
NCs	Negligible concentrations
NIP	National Implementation Plan
NOAEL	No-observed-adverse-effect level
NOEL	No observed effect concentration
OC	Organic compound
OCPs	Organochlorine pesticides
PAHs	Polycyclic aromatic hydrocarbons
PBDEs	Polybrominated diphenyl ethers
PCA	Principal Component Analysis
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo-*p*-dioxins
PCDFs	Polychlorinated dibenzo-*p*-furans
PCNs	Polychlorinated naphthalenes
PEC	Potency equivalent concentration
PEL	Probable effect level
PFAAs	Perfluoroalkyl acids
PFCAs	Perfluoroalkyl carboxylates
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFSAs	Perfluoroalkyl sulfonates
PNEC	Predicted no-effect concentration
POPs	Persistent Organic Pollutants
ppb	Parts per billion
ppm	Parts per million
PSA	Primary Secondary Amine
QuEChERS	Quick, Easy, Cheap, Effective, Rugged and Safe
QV	Quality values
RfD	Reference dose
rpm	Revolutions per minute
RQ	Risk quotient
S/N	Signal to noise
SELs	Serious effect limits
SN	Sundarbans
SPSS	Statistical Package for the Social Science
SQG	Sediment quality guideline
SV	Screening value
TDI	Tolerable daily intake
TEF	Toxic Equivalency Factor
TEQ	Toxic Equivalents
TN	Total nitrogen
TOC	Total organic carbon
TSS	Total suspended solids
USEPA	United States Environmental Protection Agency
WHO	World Health Organization
YNU	Yokohama National University

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Abstract of thesis

Persistent organic pollutants (POPs) have triggered an increasing concern globally for several decades because of their high level of persistence, toxicity, and ability to travel long distance far from their sources of usage, release and emission. These chemicals tend to bioconcentrate and biomagnify in the food chains, representing a definite health hazard for both wildlife and humans. Consequently, they are subject to international regulation under the Stockholm Convention. Emerging evidences suggest that emission sources of a number of POPs is gradually shifting from industrialized and developed countries to developing and/or least developed countries in tropical and sub-tropical regions. In addition, coastal environmental compartments, such as water, sediment, and biota particularly seafood are the ultimate recipients and final reservoirs of these persistent and toxic chemicals. Seafood constitutes a major portion of the daily diet for the coastal people. Subsequently, consumption of contaminated seafood is one of the most significant route of human exposure to POPs.

Bangladesh is a developing country that is undergoing rapid urbanization and industrialization in recent years particularly in the coastal belt. The country has a highly irregular deltaic marshy coastline of 580 kilometers, divided by many rivers and streams that enter the Bay of Bengal. About 42 million people (30 % of the total population) live in the coastal area (47,211 km^2; 32 % of the total land area) who consume seafood frequently in their daily diet. There are several suspected local emission sources of POPs in Bangladesh and the situation is even worse in the coastal regions. The accumulation of these toxic chemicals in this area is thus assumed to be responsible for the diminishing or damaging coastal/marine ecosystem integrity and obviously a matter of public health concern in terms of seafood safety. Unfortunately, the concerned authorities and general people have not been much aware of the existing situation; however, there are few studies monitoring concentrations of certain POPs in the environment in this country, and no comprehensive studies have been conducted so far in the coastal area of Bangladesh. Therefore, this research was initiated to provide the baseline information on the levels, distribution, and possible sources and origins of certain POPs in this area. A preliminary ecological and human health risk assessment was also carried out. The basic purpose of this study was to figure out broadly the present situation, existing problems and challenges in terms of POPs pollution in the Bangladeshi coastal area.

In this thesis, the burdens of three classes of POPs – Polychlorinated biphenyls (PCBs), Perfluoroalkyl acids (PFAAs), and Polycyclic aromatic hydrocarbons (PAHs) – were investigated in the surface water, sediment and commonly consumed seafood from the coastal area of Bangladesh. A wide-scale monitoring survey was commenced concomitantly in the four sampling sites (Cox's Bazar, Chittagong, Bhola and Sundarbans) with fourteen different locations in the southeast and southwest coastal area of Bangladesh. Sampling sites were chosen to show the influence of the potential pollution sources (cities, industrial areas, rivers and estuaries). The sampling was conducted considering two distinct seasons, winter (dry period) and summer (wet period). The winter samples were collected on early January to early February, 2015 and the summer samples were collected during the transition of summer and rainy season starting from early August to early September in 2015. The reasonable and adequate ground for the selection of these periods was to evaluate the influence of seasons on the distribution of POPs in the Bangladeshi coastal environments.

The research activities carried out and the findings are described in details in chapters 3 to 6 of this thesis. Chapter 3 are devoted to a full congener analysis of PCBs in the surface water, sediment and seafood along with the distribution, source characterization, ecological and human health risk assessment. The results of the occurrences of PFAAs are presented in chapters 4 and 5. Specifically, chapter 4 presents a detailed analysis on monitoring and assessment of PFAAs in surface water and sediment. An analysis of PFAAs contamination in the commonly consumed seafood and exposure assessment regarding human health risk are presented in chapter 5. Chapter 6 takes a detailed look at contaminations from PAHs in the Bangladeshi coastal area including their levels, distribution, seasonal variation and ecological and human health risk implications. However, the data are briefly interpreted here as follows.

The total concentrations of all 209 PCBs (∑PCBs) in surface water, sediment, and seafood were 32.17–160.7 ng/L, 5.27–92.21 ng/g dry weight (dw) and 5.16–79.62 ng/g wet weight (ww) in winter, and 46.45–199.4 ng/L, 4.61–105.3 ng/g dw and 3.82–86.18 ng/g ww in summer, respectively. PCB profiles were dominated by moderately chlorinated (4–6 Cl) homologs. Our analyses elucidated that the prominent sources of PCBs in the Bangladeshi coastal areas were derived as related to PCB technical mixtures, pigments/dyes, and combustion. The top congeners based on dominance by both occurrence and abundance were identified as potential markers of ∑PCBs, which can be used for future selective monitoring in case of reasonable constraints on full congener approach. In addition, the sum of these most abundant congeners comprising up to 46−82% of ∑PCBs by sites or species, and highly correlated with ∑PCBs (Pearson correlation, $r = 0.976-0.996$; $p < 0.05$), well representing the environmental burdens of ∑PCBs in the Bangladeshi coastal area.

This study reports the first evidence of PFAAs in Bangladesh. Fifteen target PFAAs, including C_{4-14}-PFCAs (perfluoroalkyl carboxylates) and C_4, C_6, C_8, and C_{10}-PFSAs (perfluoroalkyl sulfonates), were measured. In general, the total concentrations of PFAAs (∑PFAAs) in the surface water, sediment and seafood were in the range of 10.6 to 45.2 ng/L, 2.48 to 8.15 ng/g dw, and 0.32 to 8.71 ng/g ww in winter, and 11.5 to 46.8 ng/L, 1.07 to 3.81 ng/g dw, and 0.80 to 14.58 ng/g ww in summer, respectively. Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) were the two most abundant PFAA compounds. This was consistent with the fact that these are the two most industrially utilized PFAA compounds that were widely used in a number of applications. However, the occurrences of other PFAAs at a reasonable rate and levels elucidated their alternative usage to the banned/restricted PFAAs in this area. Since this the very first study reporting environmental occurrences of PFAAs and there is currently no information on the usage, production, import, and volumes of PFAAs in Bangladesh, it is therefore very difficult to pinpoint precisely the specific sources of PFAAs in the study area, and thus, more researches are recommended in this field.

The total concentration of 16 USEPA priority PAHs (∑PAHs) in surface water, sediment, and seafood were 855.4–9653.7 ng/L, 349.8–11058.8 ng/g dw and 184.5–2806.6 ng/g ww in winter, and 679.4–12639.3 ng/L, 199.9–17089.1 ng/g dw and 117.9–4216.8 ng/g ww in summer, respectively. Emissions of PAHs in the Bangladeshi coastal area were traced to both the pyrogenic and petrogenic sources including crude petroleum (e.g. gasoline/diesel), petroleum combustion, and combustion of grass, wood and coal.

The compounds detected in the highest amounts were PAHs in each of the medium examined in this study. However, POPs levels were at the middle of reported global range. The water, sediment, and seafood (finfish and shellfish) were noted to be severely polluted where the levels of POPs were higher than the national and international environmental quality guidelines, suggesting potential threats to the aquatic organisms and human health as well through biomagnification. There was no specific seasonal pattern of POPs in the Bangladeshi coastal areas. Spatial distribution revealed that the Chittagong, Cox’s Bazar and Sundarbans areas were more contaminated with POPs than the Meghna Estuary (Bhola) and because of greater development, thus associating these compounds to urbanization and industrialization. The results depicted that the ship breaking and port activities, open dumping and burning of waste (mostly consumer products including e-waste), discharges of untreated industrial and municipal effluents were the major source factors of environmental burden of POPs.

Finally, a preliminary dietary exposure assessment was evaluated to elucidate the potential health effects resulting from the consumption of contaminated seafood. In this study, two general population sub-groups, adults (≥18 years) and children (6−17 years) were considered as target POPs exposed subjects. The seafood consumption data were extracted from the questionnaire surveys during our sampling campaigns. Our results revealed that the estimated daily intake (EDI) of PCBs, and potency equivalent concentration (PEC) and incremental lifetime cancer risk (ILCR) of PAHs were several times higher than the international guideline values for the protection of human health. However, the EDI of PFAAs were far below the level of safety concern. Therefore, the potential health risk for the Bangladeshi coastal people from exposure to the dietary POPs, particularly PCBs and PAHs through seafood consumption at the present levels should not be ignored. Additionally, people can be exposed to these chemicals including other POPs, such as the organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCNs), etc. through consumption of seafood and other foodstuffs (e.g. rice, meat, vegetables, etc.) and via other routs of exposure (e.g. inhalation and dermal contact), which were not encompassed in this study, and these will further increase the POPs-induced health effects on the exposed coastal residents. We should concentrate our views to solve this problem with an integrated approaches on an urgent basis as well. It is thus recommended that continuous monitoring of these toxic chemicals in all foodstuffs and environmental compartments should be carried out to elucidate a complete scenario of the ecological and human health risk implications in the coastal areas of Bangladesh.

Chapter 1

GENERAL INTRODUCTION

General introduction

1.1 Persistent organic pollutants (POPs)

Persistent organic pollutants (POPs) are a group of organic pollutants posing a global concern. POPs are intentionally or unintentionally produced lipophilic chemicals (UNEP, 2003) which have toxic properties, resist bio- and photochemical degradation in the environment, bio-accumulate through food chains and are transported over long distances from their primary emission locations (by hundreds or thousands of kilometers) via the atmosphere, rivers and marine environments (Buccini, 2003; Wong et al., 2005). In the past few decades, POPs have got global attention due to their bioaccumulation properties, high toxicity, and ubiquitous exposure of humans and wildlife (UNEP, 2007).

Since the 1970s, there has been an increasingly wider scientific field dealing with environmental risks associated with the occurrence and fate of POPs. To acknowledge the global issue of POPs and to protect human and environmental health the UNEP Stockholm Convention on POPs entered into force in 2001 regulating or banning a preliminary list of twelve chemicals (including polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans−PCDD/Fs, polychlorinated biphenyls−PCBs, hexachlorobenzene−HCB, several organochlorines used as pesticides: dichloro-diphenyl-trichloroethane−DDT, chlordane, toxaphene, dieldrin, aldrin, endrin, heptachlor and mirex) which fulfilled all the criteria of persistence, bioaccumulation, toxicity and potential for long range transport setting the definition of POPs. There are, however, numerous other POPs which are also environmental contaminants and are of great concern and these are commonly known as 'candidate POPs' or 'POPs like' chemicals. Some of them are both persistent and toxic, and still in widespread production and use, in both industrialized and less industrialized developing countries. These include polycyclic aromatic hydrocarbons (PAHs), perfluoroalkyl acids (PFAAs), hexachlorohexane (HCH) isomers, organotin compounds (for example, used as anti-fouling agents for ships), organic mercury compounds, some other pesticides—pentachlorophenol, endosulfan and atrazine, chlorinated paraffins (for example, used in cutting oils and lubricants), polybrominated diphenyl ethers (PBDE, used as flame retardants) and certain phthalates: dibutyl phthalate (DBP) and diethyl-hexylphthalate (DEHP), which are less persistent but are not the less hazardous (mainly used as plastic softeners, especially in polyvinyl chloride (PVC)). The Government of the People's Republic of Bangladesh has signed the Stockholm Convention on Persistent Organic Pollutants on 23 May 2001, after actively taking part in the negotiation process leading to the final Convention. As a Party and signatory to the Stockholm Convention, Bangladesh will be required to take actions to generate general awareness of harmful consequences of POPs to restrict and reduce their releases, and their ultimate elimination from the environment (http://www.doe.gov.bd/old/pop/html/popsinbd.htm).

Most of the POPs are lipophilic, which means that they accumulate in the fatty tissue of living animals and human beings (http://www.unece.org/spot/s01.htm). In fatty tissue, the concentrations can become magnified by up to 70,000 times higher than the background levels (http://web.worldbank.org/). As moving up the food chain, concentrations of POPs tend to increase so that animals at the top of the food chain such as fish, predatory birds, mammals, and humans tend to have the greatest concentrations of these chemicals, and therefore are also at the highest risk from acute and chronic toxic effects (Pandit et al., 2001; Eljarrat et al., 2005; Wurl and Obbard, 2005; Mohammed et al., 2011). It is thus crucial to understand how the presence, exposure and effects of POPs impact environmental quality.

Specific effects of POPs can include cancer, allergies and hypersensitivity, damage to the central and peripheral nervous systems, reproductive disorders, and disruption of the immune system. Some POPs are also considered to be endocrine disrupters, which, by altering the hormonal system, can damage the reproductive and immune systems of exposed individuals as well as their offspring; they can also have developmental and carcinogenic effects. The evidence of detrimental effects on living organisms at the population level demonstrates that POPs are a threat to the biodiversity and even have potential for disruption at the ecosystem level (http://chm.pops.int/).

The evaluation of POPs and candidate POPs and the restriction of their use are difficult processes. All pros and cons of a substance should be carefully balanced. Clearly, on the one hand, emissions should be reduced as much as possible and production may have to be terminated. On the other hand, some of these chemicals help to save lives or have other important functionalities that cannot at once be replaced. Although the use of POPs compounds has significantly declined in past two decades but some of them are still in use in developing countries because of their low cost and versatility in industry, agriculture and public health. Consequently, environmental problems associated with toxic contaminants in these countries are of great concern. In order to understand the role of tropical developing countries as possible sources of POPs, it is necessary to elucidate the distribution, behavior and fate of these compounds in the environmental compartments (Tariq et al., 2007; Eqani et al., 2011).

1.2 The specific POPs investigated by this thesis

Three classes of the POPs – polychlorinated biphenyls (PCBs), perfluoroalkyl acids (PFAAs), and polycyclic aromatic hydrocarbons (PAHs) – constitute the focus of this thesis.

1.2.1 Polychlorinated biphenyls (PCBs)

PCBs are purely man-made products, either synthesized as industrial chemicals for commercial purposes or generated unintentionally by combustion processes, especially during incineration of waste. They are among the so-called dirty-dozen, i.e. the first group of chemicals listed for elimination by the Stockholm Convention. PCBs are chemically inert and thermally very stable. Hence, they became very useful as insulating material in electric equipment such as transformers and capacitors and also in heat transfer fluids and in lubricants. They have also been used in wide range of products such as plasticizers, surface coatings, ink, adhesives, flame retardants, paints, and carbonless duplicating paper. PCBs were manufactured under various trade names in different countries. Examples of countries where PCBs were produced, with trade names indicated in bracket, are: Japan (Kanechlor), the United States (Aroclor), the United Kingdom (Pyroclor), Germany (Clophen), France (Phenoclor), Italy (Fenclor), former Czechoslovakia (Delor), and former USSR (Sovol) (Breivik et al, 2002; USEPA, 2004).

PCBs belong to a group of synthetic organic compounds called chlorinated hydrocarbons and have the chemical formula $C_{12}H_{(10-n)}Cl_n$, where n is the number of chlorine atoms and ranges between 1 and 10. They consist of two benzene rings linked together by a carbon-carbon bond (a biphenyl structure), with varying degree of chlorination (**Figure 1-1**). The two rings in a PCB molecule can rotate around the bond connecting them. The shape is further influenced by repulsion between nearby chlorine atoms so that the rings of a specific PCB will either lie approximately in the same plane (called co-planner) or in different, more perpendicular planes (called non-planner). The co-planner PCBs are “dioxin-like” and more toxic. Theoretically, there are 209 PCB mono- to deca-congeners (structural isomers) based on the different number of possibilities of chlorination of the carbon atoms. Congeners containing the same number of chlorine atoms constitute a homologue.

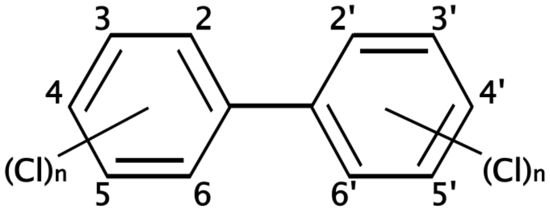

Figure 1-1. PCB molecular structure.

PCBs were first manufactured commercially in 1929, and serious concerns about the distribution of PCBs in the global environment were raised in the 1960s when PCBs were found in soil, water, and animals (Jensen, 1966). The production/usage of PCBs have been banned/restricted worldwide since the early 1970s (Harrad et al., 1994), but a large amount of PCBs was already produced. A recent study pegged the total global production of PCBs at about 1.3 million MT (Breivik et al., 2002). In addition to industrial syntheses, PCBs are also byproducts from a number of combustion processes such as municipal solid waste incineration, domestic burning of coal and wood for residential heating, and domestic and public bonfires (Farrar et al., 2004). PCBs were also present as impurities in other chemicals such as chlorinated phenols (Mai et al., 2005). It has even been hypothesized that there has been a shift in primary emission regions of PCBs on a global scale with high emission continuing in some sub-tropical and tropical regions implicated as recipients of wastes, paralleled by significant reductions in environmental burdens within former use regions (Breivik et al., 2011).

1.2.2 Perfluoroalkyl acids (PFAAs)

PFAAs are organic substances in which all of the hydrogens of the hydrocarbon backbones are substituted with fluorine atoms. The fluorine-carbon bonds are extremely stabile conferring these substances with very high thermal and chemical stability. PFAAs are persistent, and some of the substances bioaccumulate in the environment. They can be divided into the groups of perfluorinated sulfonic acids (PFSA), perfluorinated carboxylic acids (PFCA), fluorotelomer alcohols (FTOH), high-molecular weight fluoropolymers and low-molecular weight perfluoroalkanamides. Perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), often referred to as reference or key substances for the first two groups, have been most intensively studied from a toxicological standpoint. PFAAs have been synthesized for more than 50 years and are used in numerous industrial and consumer products (Stahl et al., 2011).

Most of these compounds are present in the environment as a result of human manufacture and use. Release of fluorochemicals into the environment can occur at each stage of the fluorochemical product's life cycle. They can be released when the fluorochemical is synthesized, during incorporation of the fluorochemical into a product, during the distribution of the products to consumers, during the use of the product by the consumers, and during disposal (3M, 2000).

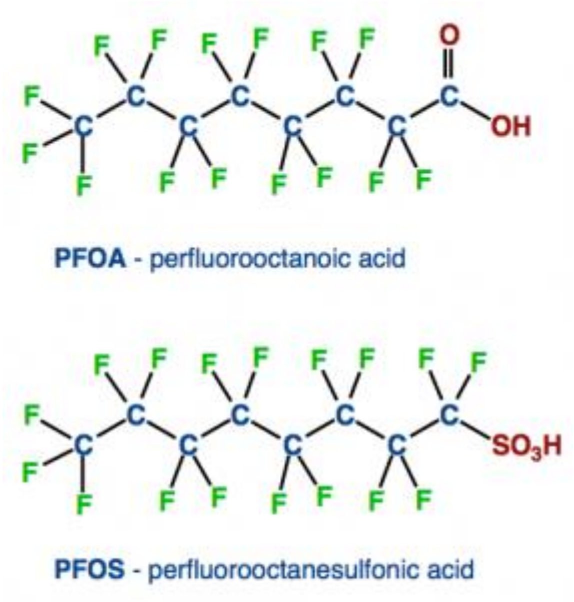

Figure 1-2. Molecular structure of PFOA and PFOS.

All industrial PFOS derivatives are prepared from perfluorooctanesulfonyl fluoride, which, in turn, is obtained by electrochemical fluorination of octanesulfonyl fluoride. Base-catalyzed hydrolysis of the fluoride results in PFOS or the respective salt. Sulfonamide derivatives have also been obtained from fluoride (Kissa, 2001). PFOS and related substances (e.g., PFHxS, PFBS) have received worldwide attention over the last few years. The term "related substances" is used to refer to any or all substances which contain the PFOS moiety (defined as the $C_8F_{17}SO_2$ group) and may break down in the environment to give PFOS. They are also referred to as the "PCBs of the twenty-first century" (Taniyasu et al., 2003). Many of the degradation products of PFAAs have been found in the environment throughout the world, but PFOS and PFOA are the two compounds most widely detected. Because of the strong carbon–fluorine (C–F) bond associated with PFAAs, PFOS and PFOA are environmentally persistent substances that have been detected worldwide in water, soils, sediments, air and biota including human (Verònica Maria Vieira, 2005).

Fluorinated surfactants are known to be extremely resistant to chemical attack and so “can be used in media where conventional surfactants do not survive” (Kissa, 1994). They are stable to heat, acids and bases, and to reducing and oxidizing agents. Used as components in products, they repel both water and oil, reduce surface tension to much lower levels than other surfactants, act as catalysts for oligomerization and polymerization, and function where other compounds would rapidly degrade (in high temperatures, strongly acidic conditions, etc.) (3M, 2000). This extraordinary stability leads to special uses, e.g., in fire-fighting foams to extinguish fires burning at high temperatures (Moody et al., 2002). In addition, PFAAs are utilized in pesticides, cosmetics, adhesives, greases and lubricants, where they exhibit unique properties that make fluorinated surfactants irreplaceable in many of these applications. However, these fluorinated surfactants are not only stable against chemical and physical attacks; persistence against biochemical attack has also been reported (Schröder, 2003). Hence, fluorinated organic compounds are environmentally persistent substances which also exhibit bioaccumulative properties and toxicity to mammalian species (Schultz et al., 2003). After application, these fluorinated surfactants reach the aquatic environment either through their release into rivers or via wastewater discharge into receiving waters. Predominantly, however, they are adsorbed to sewage sludge (Schröder, 2003). The use of sludge for land treatment or its disposal on dump sites leads to a remobilization of these recalcitrant compounds. Also, their polarity and mobility in water and soil allow them to reach the sea or ground water in unaffected or undegraded conditions. As a consequence, these compounds have been shown to be distributed globally (Schröder, 2003). They have been detected in surface waters at concentrations in the range of ng–mg/L, in ground waters (at lower levels, μg/L), in wastewaters, sediments and sewage sludge, in the blood (ng–μg/mL) and liver (ng/g) of humans, and in the liver and fat (ng–μg/g) of several species of wildlife (including Arctic species), such as wild birds and fish (Schultz et al., 2003).

1.2.3 Polycyclic aromatic hydrocarbons (PAHs)

PAHs are included in the European Union and USEPA priority pollutant lists because they represent a large group of compounds that are mutagenic, carcinogenic, and teratogenic (Sverdrup et al., 2002; Qiao et al., 2006). They are one of the most important classes of organic contaminants in marine environments. The major sources of PAHs are both natural and anthropogenic. For examples, the terrestrial deposit of coals, atmospheric input from incomplete combustion (wood burning, forest fire, fossil fuel, and coke oven), oil leaks or spills, and exhaust emission from vehicles are the major sources PAHs in the environments (van Metre et al., 2000). The effect of PAHs is usually widespread and permanent in environmental media. Most PAHs have high hydrophobicity, and can be sorbed strongly by water-borne organic and inorganic particles. They may eventually sink down to the bottom sediment of an aquatic system. The PAHs found in the sediment are resistant to bacterial degradation in an anoxic environment. Even under favorable conditions, sorbed PAHs are released into the water as an extended source that threatens the aquatic ecosystem through bioaccumulation in the food chain (Zhang and Zheng, 2003; Chen et al., 2013).

Polycyclic aromatic hydrocarbons (also known as polynuclear aromatic hydrocarbons) are composed of two or more aromatic (benzene) rings which are fused together when a pair of carbon atoms is shared between them (**Figure 1-3**). The resulting structure is a molecule where all carbon and hydrogen atoms lie in one plane.

Physical and chemical characteristics of PAHs vary with molecular weight. For instance, PAH resistance to oxidation, reduction, and vaporization increases with increasing molecular weight, whereas the aqueous solubility of these compounds decreases. As a result, PAHs differ in their behavior, distribution in the environment, and their effects on biological systems. PAHs can be divided into two groups based on their physical, chemical, and biological characteristics. The lower molecular weight PAHs (e.g., 2 to 3 ring group of PAHs such as naphthalenes, fluorenes, phenanthrenes, and anthracenes) have significant acute toxicity to aquatic organisms, whereas the high molecular weight PAHs, 4 to 6 ring (from chrysenes to indeno(1,2,3-cd)pyrene) do not. However, several members of the high molecular weight PAHs have been known to be carcinogenic.

Although the health effects of individual PAHs are not exactly alike, these 16 PAHs have been identified as being of greatest concern with regard to potential exposure and adverse health effects on humans and are thus considered as a group: Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)-fluoranthene, Benzo(a)pyrene, Dibenz(a,h)anthracene, Benzo(g,h,i)perylene, and Indeno(1,2,3-cd)pyrene. However, International Agency for Research on Cancer (IARC) has classified seven PAH compounds as probable human carcinogens: benz(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene, dibenz(ah)anthracene, and indeno(1,2,3-cd)pyrene (IARC, 1987).

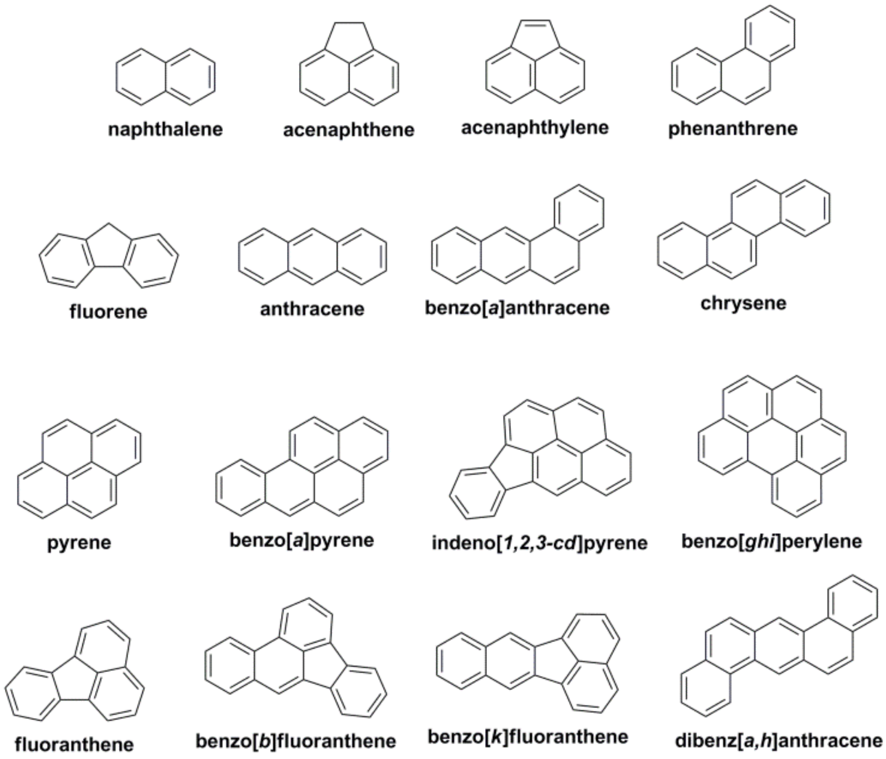

Figure 1-3. Molecular structure of 16 USEPA priority PAHs.

1.3 Background of this study

The POPs are ubiquitous in the environment with some of volatile, semi-volatile and non-volatile characteristics. Once they are produced, these pollutants can be emitted and discharged into the environment through their production and application processes (Chen et al., 2007). Upon emitting into the atmosphere, these compounds transport hundred to thousand miles which can be redeposited into the aquatic and terrestrial ecosystems far from their sources (**Figure 1-4**). Moreover, unutilized portion of POPs from their industrial and/or household applications find their way into the environment which can be temporarily deposited in river drainage basins, and subsequently transported down-river over time, to end up, in ports, estuarine and coastal areas (**Figure 1-4**).

The coastal area is the triple interface of air, land and sea. As a result, the coastal area is highly influenced by the industrial and agricultural activities, urban facilities, some sea-based activities like shipping and transportation, ship breaking and repairing, etc. The coastal ecosystems are also influenced by the wave and tidal fluxes of the sea. Waves, water currents, and wind move the oil onto shore with the surf and tide. Oil waste reaches the shoreline or coast; it interacts with sediments such as beach sand and gravel, rocks and boulders, vegetation, and terrestrial habitats of wildlife and humans, causing erosion as well as contamination (Vikas and Dwarakish, 2015). All the above mentioned coastal activities, directly or indirectly, influence the fate and distribution of POPs in the coastal ecosystems. Moreover, POPs present in the atmosphere may participate in exchange or deposition mechanisms that will determine their inputs to open waters and coastal areas of the sea (Agrell et al., 2002; Jurado et al., 2005). Upon entering into the coastal ecosystems they interacts with various types of materials and undergoes many transitions between different compartments such as water, suspended matter, sediments and organisms and are considered potent toxicants, capable of producing a wide spectrum of adverse health effects in biota and humans (Safe, 1984, 1990; Vreugdenhil et al., 2002). Therefore, determination of those compounds existing in coastal water, sediment and biota may indicate the extent of coastal organic pollution and the accumulation characteristics in the coastal ecosystems. However, the multi-media monitoring of the environmental samples has been proved to be a very suitable concept to address various important issues of coastal pollution by persistent organic pollutants (Tanabe and Subramanian, 2006).

In particular, coastal water is an important medium influencing the fate, transport and bioavailability of hydrophobic organic contaminants in coastal regions (Xie et al., 2007; Gioia et al., 2008; Lohmann et al., 2013). In coastal water, hydrophobic organic compounds can be freely dissolved or bound to particulate phases, with phase distribution being determined by the chemical hydrophobicity. Freely dissolved concentrations are more relevant for ecotoxicology studies, as this represents the bioavailability to aquatic organism, while particulate-bound concentrations are important for fate and transport investigations (Bergen et al., 1993, Mackay and Fraser, 2000; Vrana et al., 2005).

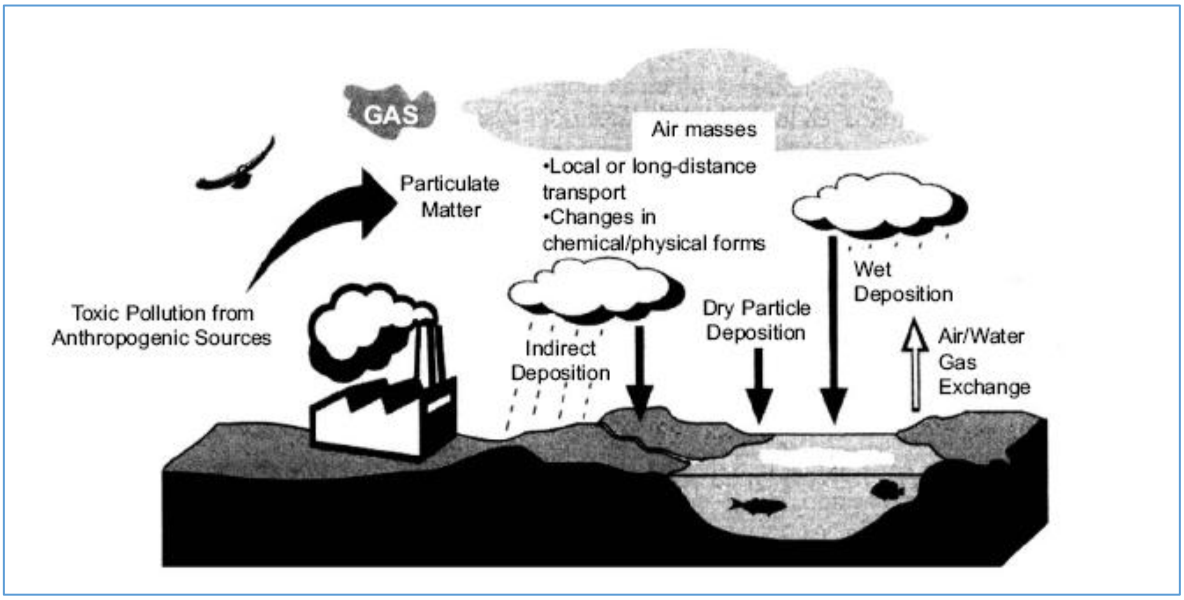

Figure 1-4. Possible pathways showing how POPs can be released into the environment, transported, and redeposited in water and on land far from their sources (Source: USEPA, 1994).

Sediments generally consist of detritus, inorganic and organic particles, and are generally spatially heterogeneous in terms of physical-biogeochemical characteristics (Hakanson, 1992) and this heterogeneity can result in large spatial variability in POPs levels. Sediment is an important component in water system which is regarded as a sink and reservoir for many particle-bound persistent organic pollutants and has a large impact on their distribution, transportation, and fate in the aquatic environment (Ahrens et al., 2009; Yang et al., 2011; Jonsson et al., 2003; Liu et al., 2008). Reversibility of the contaminants exchange between sediment and the overlaying water column results in sediments playing as key secondary sources for the benthic and pelagic water ecosystem, effectively controlling biota exposure (Li et al., 2000; Zeng and Venkatesan, 1999). Through the interaction between sediments and water, the transfer of POPs from sediments to organisms is now regarded as a major route of exposure for many species (Zoumis et al., 2001).

Several factors are involved in affecting the occurrence of persistent organic contaminants in the coastal sediments including composition of sediments (grain size fractions, e.g. sand, silt, clay), water content, organic matter content (Loss on Ignition, LOI), total organic carbon (TOC), and black carbon (BC), partitioning processes and pattern of deposition of contaminants in the sediments (Sánchez-García et al., 2010). Sorption is one of the key phenomena that affects the fate and behavior of contaminants in the sediments.

Amorphous organic matter relatively homogenous, lipophilic and gel like matrix (Cornelissen et al., 2005) is one of the responsible agents for the absorption of contaminants in sediments (Accardi-Dey and Gschwend, 2002). Another form of organic matter of glassy and condensed type, generally termed as carbonaceous geosorbents (CG), is involved in the adsorption of hydrophobic contaminants (Semple et al., 2013). Recently, Hung et al. (2006, 2007) showed that TOC and BC concentrations were strongly, positively correlated with concentrations of total PCBs and selected pesticides in aquatic sediments, suggesting that these carbonaceous geo-sorbents can significantly influence the distributions of organic pollutants through either post-depositional adsorption or sequestration, or by co-transport of similarly sourced materials. These field results contribute to the growing body of literature which demonstrates that TOC and BC plays an important role in the distributions of aromatic hydrocarbons such as PAHs, PCBs and some common pesticides (Hung et al., 2010). Moreover, since organic matter is a primary source of food for benthic organisms, it is important in maintaining a viable ecosystem.

In particular, black carbon (BC) is an important type of CG involved in affecting the overall behavior of OCs in sediments, partially due to its porous nature (Sun et al., 2008). Highly carbonaceous BC acts like activated carbon and shows very high affinities for organic compounds including POPs (Ahmad et al 2001; Allen-King et al. 2002; Jonker and Koelmans, 2002). Understanding the relationship between BC and OCs in sediments is essential for determining the fate and behavior of OCs in coastal environments (**Figure 1-5**). BC influences the retention of contaminants in sediments (Hung et al., 2006; Jonker and Koelmans, 2002). Primarily, BC deposits near the source of emission but it can travel hundreds to thousands of kilometers in the atmosphere reaching even remote sites. In sediments, their residence time could reach up to several millions of years (Masiello and Druffel, 1998). Organic pollutants strongly bind to BC particles through the occlusion into the carbon condensed structure, conditioning their transport to that of BC particles and limiting their bioavailability (Ahrens and Depree, 2004; Huang et al., 2003; Jonker and Koelmans, 2002; Koelmans et al., 2006).

However, BC and other carbonaceous material has attracted considerable interest from environmental chemists, because its sorption capacity for some hydrophobic organic compounds (HOCs) may be at least 10–100 times higher than that of AOM; it is considered a ‘super-sorbent’, and believed to be responsible for a large part of the sorption/partitioning of some HOCs in soils and sediments (Accardi-Dey and Gschwend, 2002; Cornelissen and Gustafsson, 2005; Cornelissen et al., 2005; Lohmann et al., 2005; Nam et al., 2008). Since the sorption can limit the degradation of pollutants as well as influence the rate and extent that bioaccumulation occurs, a quantitative understanding of sorption is necessary to evaluate the hazards that pollutants pose (Accardi-Dey and Gschwend, 2002).

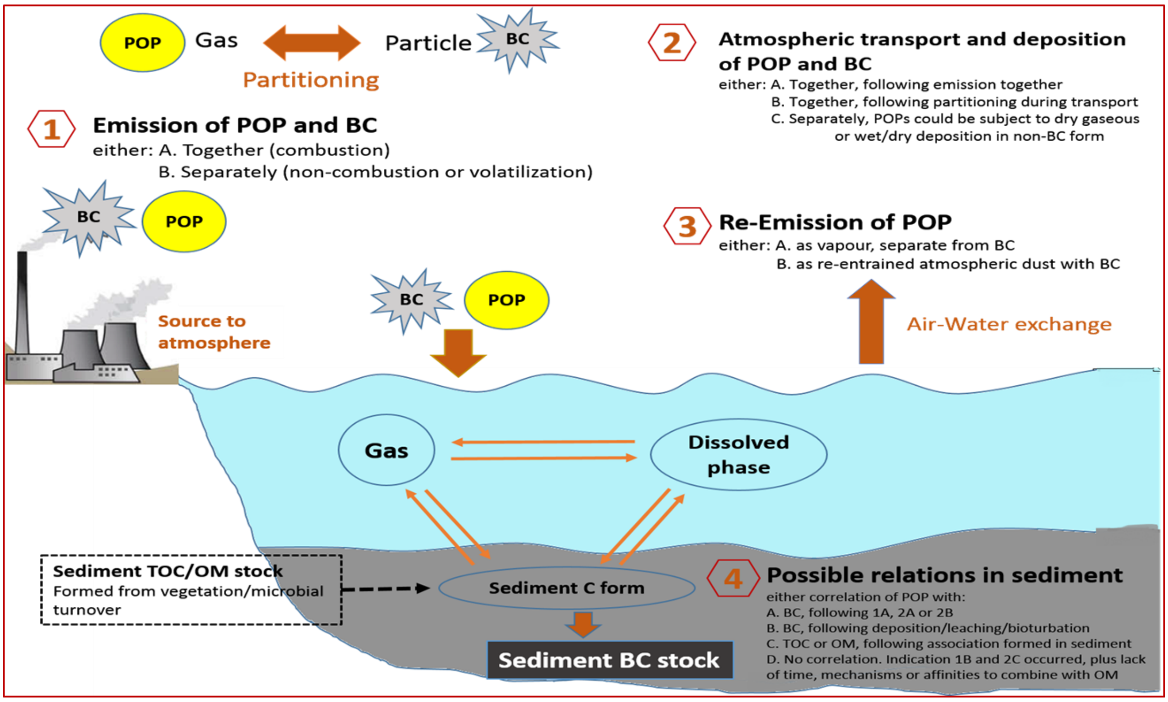

Figure 1-5. Possible scenarios for the relationship between POPs and sediment OM (with a focus on BC) and the inferences which can be made about their sources, fate, and transport (adapted from Nam et al., 2008).

Eventually, some of the POPs which will end up in the coastal ecosystem are taken up by the biota including phytoplankton, algae, microorganisms, and plants. These species are in turn consumed by fish and crustaceans, resulting in their biomagnification within food chains. Since POPs have a high affinity for lipids and a low elimination rate, they tend to bioaccumulate in fatty tissue (Holoubek, 2001). For finfish and shellfish (e.g. crustaceans), POPs can also be taken up via gill respiration, depending on how lipophilic the contaminants are (AMAP, 1998). Fishes and crustaceans are often used as bioindicators of aquatic pollution to elucidate the contamination status and distribution of POPs (Ueno et al., 2003; Salem et al., 2014), because they occupy high trophic level and are important food source of human population (Abdel-Baki et al., 2011). The consumption of fish has increased in importance among the health conscious due to their high protein supply, low saturated fat and omega-3 fatty acids content that are known to contribute to good health (Copat et al., 2012). However, humans are on top of the food chains and are therefore at risk of being exposed to high levels of contaminants (Middaugh et al., 2001; Braune et al., 2005; Chiu et al., 2004), with an increase exposure risk to those consuming the contaminated fish and other seafood that eat higher on the food chain. Thus, it is of utmost importance to investigate the status of POPs in fish and crustaceans from coastal water bodies since the contaminants levels in fish and seafood would reflect bioaccumulation potential from exposure through consumption (Hardell et al., 2010).

Ecological or environmental risk assessment (ERA) is defined as the procedure by which the likely or actual adverse effects of pollutants and other anthropogenic activities on ecosystems and their components are estimated with a known degree of certainty using scientific methodologies (Depledge and Fossi, 1994). ERA has become increasingly important since environmental scientists as well as the general public have learned that POPs are ubiquitous environmental contaminants which persist in the environment, bioaccumulate through the food chain, and pose a risk of causing adverse effects to human health and wildlife. The risk assessment process can be divided into a scientifically oriented risk analysis and a more politically oriented risk management. Risk analysis is a process, which comprises some or all of the following elements: hazard identification, effect assessment, exposure assessment and risk characterization (Van Leeuwen and Hermens, 1995). Environmental risk management deals with regulatory measures based on risk assessment (Van Leeuwen and Hermens, 1995). Risk management and risk analysis, are closely related but different processes: in risk analysis the risk of a certain situation is determined, whereas risk management examines solutions to the problem. Although ERA is generally performed by predictive methods, the interest in the assessment of pollution that began in the past and may have ongoing consequences in the future is increasing. These so-called retrospective ERAs are primarily concerned with establishing the potential relationship between a pollutant source and an ecological effect caused by exposure of organisms to the pollutant (Suter, 1993; van der Oost et al., 2003).

Since human exposure to POPs occurs through dietary intake more commonly than through inhalation or dermal contact (Darnerud et al., 2006, Wang et al., 2013). Ingestion of contaminated food, especially of animal origin with high fat content, were the main dietary source of human exposure to POPs (Li et al., 2008; Roosens et al., 2010). Information on the residual concentrations in ingredient (e.g., seafood including finfish and shellfish), or the estimation based on aqueous concentration and bioaccumulation factor (sediment-biota accumulation) is essential for human health risk assessment of POPs (Asante et al., 2013). Recent studies on distribution of POPs around the globe suggest that developing nations are at higher risk of exposure to POPs as the POP emission sources have shifted to developing nations (Bogdal et al., 2013; Chakraborty et al., 2013; Zhang et al., 2008). In Asian developing countries, however, less information are available on human exposure to POPs, although it is suspected that the residents have been exposed to relatively high levels of these contaminants (Tanabe and Kunisue, 2007; Ali et al., 2014).

The presence of POPs in terrestrial and aquatic food chains is relevant to those concerned with both human health and environmental protection because of the many common exposure pathways and biological effects among different species. In the past, some chemicals with health risks for humans have been identified following reports of adverse effects in wildlife. Integrating human and ecological risk assessments may improve society's ability to manage the design, manufacture, use and disposal of chemicals in a safe and efficient manner. Integrated human and ecological risk assessment represents a new direction for characterizing the risks which anthropogenic contaminants present to the environment, within which humans are an integral part (Ross and Birnbaum, 2003).

Coastal pollution, originating from both land and sea, impacts all trophic levels, from primary producers to apex predators, and thus interferes with the structure of coastal communities and consequently ecosystem functioning (Todd et al., 2010). Most of the coastal areas of the world have been reported to be damaged from pollution, significantly affecting commercial coastal fisheries. Therefore, control of aquatic pollution has been identified as an immediate need for sustained management and conservation of the existing fisheries and aquatic resource (Islam and Tanaka, 2004). Apart from the ecosystem approach, consuming fish and seafood containing POPs is an important pathway through which humans gain exposure to these pollutants (**Figure 1-6**) (Bocio et al., 2007; Storelli et al., 2007). Therefore, investigating organic pollutants in different matrices of coastal ecosystem (e.g. water, sediment, and seafood) is an essential indicator in determining the level of contamination of coastal resources and identifying the risks to ecosystem and human health as well (Yim et al., 2005). However, recognition is increasing that both humans and the environment can be more effectively protected by more integrated approaches to research, risk assessment, and decision making (Cirone and Duncan, 2000; Di Giulio and Benson, 2002; Munns et al., 2003).

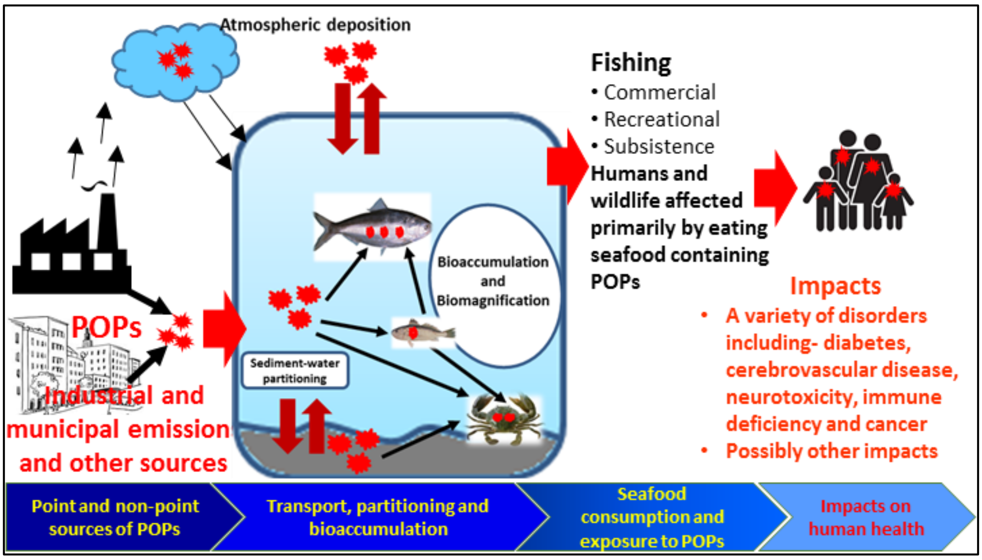

Figure 1-6. Schematic pathways of human exposure to POPs through seafood consumption.

Bangladesh is an exclusively riverine agricultural country that is undergoing rapid industrialization, urbanization and economic development in recent years, particularly in the coastal regions. The country has a highly irregular deltaic marshy coastline of 580 kilometers, divided by many rivers and streams that enter the Bay of Bengal, which is characterized by a tropical climate. The coastal areas of Bangladesh suffer from environmental degradation due to rapid human settlement, the development of industrial hubs, tourism and transportation, extensive ship breaking and port activities, illegal and mismanaged dumping of e-waste, the operation of an excessive number of mechanized boats, deforestation, and increasing agriculture and aquaculture activity and large discharges of untreated and semi-treated domestic and municipal sewage. There are also effluents containing heavy loads of organic and inorganic pollutants from many large and small local industries. **Figure 1-7** shows some representative photographs showing several potential sources of POPs pollution in the Bangladeshi coastal area. With the population and industrial pressures increasing along the rivers, the estuarine and coastal areas of Bangladesh face threats to their coastal ecosystems from contaminants such as the ascertained POPs (e.g. PCBs, PFAAs, and PAHs), which can accumulate in the coastal or marine food chains. Local populations can be exposed to these chemicals via contaminated seafood consumption (**Figure 1-6**). About 42 million people (30 % of the total population) live in the coastal area (47,211 km^2; 32 % of the total land area) of Bangladesh, of which about 5 million are engaged directly in commercial fishing

(BOBLM, 2011). Seafood is the most important and one of the major dietary components of the coastal populations. But, in the Bangladesh context, the concerned authorities and general people have not been aware and no complete study has been carried out so far regarding this issue.

Figure 1-7. Representative photographs (A–G) showing some potential sources of POPs pollution in the Bangladeshi coastal area. A: Ship breaking and dismantling; B: E-waste dumping; C: Open burning of waste including E-waste; D: Discharging of untreated industrial effluents; E: Discharging channel of untreated municipal effluents into the Bay; F: Dumping of municipal and household garbage; G: Painting of boat and ships.

1.4 Research question(s)

The present study was initiated to address the following three basic research questions:

I. Is the Bangladeshi coastal area polluted by persistent organic pollutants (POPs)?
II. Is the environmental concentrations of POPs threat to the aquatic organisms and other wildlife?
III. Is the Bangladeshi coastal population exposed to health risk through consumption of seafood?

1.5 Aim of the study

This study was initiated and intended to provide baseline information of ambient level along with seasonal trends of certain POPs including PCBs, PFAAs, and PAHs in the coastal areas of Bangladesh. The surface water, sediment, and seafood (finfish and shellfish) are being analyzed to address the present status of coastal pollution by POPs. Physical-chemical properties of water and sediment that are responsible for the distribution of pollutants in the coastal environment will also be evaluated. To assess the seasonal variation in the level of contamination, 4 sites (14 locations) are being monitored in two successive sampling campaigns; one of which is winter season (January–February, 2015) and other one is summer season (August–September, 2015). Coastal pollution by POPs might be a potential threat to the aquatic organisms including fish and other seafood. Fishes are good bioindicators to elucidate the contamination status and distribution of POPs in the aquatic ecosystems. Furthermore, the contaminated seafood from coastal environment may become a public health concern in near future. Hence, it is important to investigate the concentrations of POPs in commonly consumed or popular seafood in order to evaluate the possible risk from consumption of these seafood. Unfortunately, the accumulation of POPs in Bangladeshi coastal area has been paid less attention and no complete study has been carried out so far. Thus, it is vital to identify the legacy and contemporary issues impacting POPs concentration and their distribution in the coastal ecosystem. Therefore, the aim of this research work is to determine the concentrations of certain POPs in surface water, sediment, some commonly consumed seafood giving emphasis on ecological and public health risk assessment in the coastal area of Bangladesh.

1.5.1 Specific Objectives

The study is being undertaken with the following specific objectives in mind:

- To determine the concentrations of POPs (PCBs, PFAAs, and PAHs) in surface water, sediment and commonly consumed seafood in the coastal area of Bangladesh.
- To assess the seasonal variation of POPs level among the environmental samples.
- To assess the distribution, source and ecological risk of ascertained POPs in the Bangladeshi coastal area.
- To evaluate the public health risk with respect to the levels of POPs contamination in commonly consumed seafood.

1.6 Thesis outline

This thesis is organized into seven chapters. It begins with a general introduction in chapter 1 (this chapter), which introduces the contaminants that were investigated and the background and objectives of this research. Pertinent literature regarding the occurrences and distribution of POPs in the environmental compartments, particularly in coastal ecosystems, ecotoxicological concern of these pollutants are reviewed in chapter 2.

The research carried out and its findings are described in detail in chapters 3 to 6. Chapter 3 are devoted to a full congener analysis of PCBs in the surface water, sediment and seafood along with the distribution, source characterization, ecological and human health risk assessment. The results of the occurrences of PFAAs are presented in chapters 4 and 5. Specifically, chapter 4 presents a detailed analysis on monitoring and assessment of PFAAs in surface water and sediment. An analysis of PFAAs contamination in the commonly consumed seafood and exposure assessment regarding human health risk are presented in chapter 5. Chapter 6 takes a detailed look at contaminations from PAHs in the Bangladeshi coastal area including their levels, distribution, seasonal variation and ecological and human health risk implications. A general discussion of the various findings and their implications, as well as conclusions and recommendations for further research are presented in chapter 7. Supporting information for the various chapters is included in appendixes.

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Chapter 2

LITERATURE REVIEW

Literature review

Abstract

The introductory sections of chapters 3 to 6 each contains specific reviews. The issues not covered in those specific reviews but needed to comprehend this thesis are included in this chapter. Essentially, presented in this chapter is a review of pertinent literature on POPs, focusing primarily on PCBs, PFAAs, and PAHs, their occurrences and distribution in the environmental compartments, particularly in coastal ecosystems, ecotoxicological effects and risk assessment, which form the theme of this research. Due to growing concerns regarding POPs in the environment, extensive studies and monitoring programs have been carried out in the last two decades to determine their concentrations in water, sediment, and more recently, in biota. In recent years, particular attention has been paid to the occurrence and role of various POPs in coastal/marine environmental contamination. Interest in the local, regional and global distribution of POPs in coastal environments is exemplified by the ever-increasing number of environmental surveys and monitoring programs which have been undertaken, especially since the 1970s when analytical techniques were sufficiently developed to permit detection of environmental concentrations of these compounds. The primary objectives of most monitoring surveys can be summarized as follows: (1) Comparisons of spatial changes to identify sources and so called ‘‘hot spots” containing great contaminant concentrations; (2) Comparisons of temporal changes to detect deterioration or improvement of contaminant concentrations in the environment; (3) Checks on compliance (with reference, for instance, to governmental standards and established guidelines); (4) Assessment of possible adverse effects (e.g. ecological and public health risks); (5) Provision of exposure data for more detailed risk assessments. Such studies have revealed the ubiquity of POPs in many environmental compartments, not only in sediments, air and water, but also in living organisms. The persistence of many POPs in environmental media, combined with an increasing knowledge of their toxicity, has inevitably led to grave concerns for ecosystem and public health. Furthermore, measuring and monitoring POPs per se would be of limited merit, unless such measurements are performed in the context of risk assessment.

2.1 Fate and distribution of POPs in coastal ecosystems

POPs are organic chemical compounds that are resistant to biodegradation and thus remain in the environment for a long time, can travel great distances and become widely distributed through natural processes, bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and the environment.

The distribution of POPs has been widely investigated worldwide (Iwata et al., 1993; Kannan et al., 1997; Tanabe et al., 2000; Tanabe and Subramanian, 2006; Minh et al., 2008;

Augusto et al., 2013; Zhang et al., 2013; Li et al., 2014; Peng et al., 2015). In this thesis, the fate and distribution of POPs will be reviewed giving emphasize on aquatic environment, particularly coastal ecosystem. The fate of POPs in the coastal ecosystems depends on a wide range of processes including mechanical processes, e.g., transformation with moving flows; chemical processes, e.g., chemical decay and phase change; physical processes, e.g., adsorption; and biological processes, e.g., pollutant accumulations and transport by biota (Peng et al., 2015).

Organic pollutants can enter the coastal environment by a number of processes and once introduced they are subject to biogeochemical cycling, sinks, and bio-accumulation processes. **Figure 2-1** shows the schematics of the general cycle of organic pollutants in a coastal water body. Their inputs can be due to direct spills from industrial, aquaculture, urban runoff, and other anthropogenic activities, or to indirect inputs from rivers and run-off or from atmospheric deposition. There are three mechanisms of atmospheric deposition: (1) diffusive exchange between the atmosphere (gas phase) and the dissolved marine phase, (2) dry deposition of aerosol-bound pollutants, and (3) washout of gas and aerosol-bound chemicals by rainwater (Gigliotti et al., 2005; Jurado et al., 2005). The predominance of the atmospheric deposition mechanism will depend on the physical–chemical properties of the pollutant, magnitude of aerosol deposition, and precipitation regime. Pollutants with a high affinity for aerosols, such as PAHs (Dachs and Eisenreich, 2000) will show a tendency to deposit associated to dry and wet aerosols (Gigliotti et al., 2005). In contrast, the atmosphere–seawater exchange of other pollutants mostly occurring in the gas phase, such as PCBs, will be dominated by diffusive exchanges (Jurado et al., 2005). Many organic pollutants are hydrophobic (characterized by high values of the octanol–water partition coefficient, K_{OW}) and thus have high affinity to organic matter to which they sorb (Jurado et al., 2004). The current knowledge of the environmental cycling of this subclass of chemicals is much more advanced than for polar chemicals, which have only being comprehensively studied during the last decade. The cycling of hydrophobic chemicals in the marine environment will be coupled to that of organic matter. In fact, there are numerous works that show that some pollutants such as PCBs bioconcentrate in plankton (Dachs et al., 1999a, b; Berrojalbiz et al., 2009), bacteria (Wallberg and Andersson, 2000), and bioaccumulate in higher trophic webs (Gobas et al., 1999; Restrepo et al., 2005). The bioconcentration of POPs with low aqueous solubility is rather well understood (Del Vento and Dachs, 2002; Gobas et al., 1999), and that of ionic organic chemicals, such as pharmaceuticals, has been recently described (Fu et al., 2009). In addition, lipid normalized concentrations of persistent bioaccumulative chemicals increase at higher trophic levels of food webs by a mechanism called biomagnification (Gobas et al., 1999). Biomagnification processes in fish have been explained by using fugacity-based models. Gobas et al., (1999) found that concentrations of POPs in the gastrointestinal tract were significantly higher than in the food consumed. This was due to the food (lipids) absorption in the gastrointestinal tract, which decreased the fugacity capacity of food residuals, thus increasing its concentration that results at the end to a biomagnified concentration in the fish. Because of the tendency of hydrophobic pollutants to sorb on organic matter and soot carbon, sediments are important recipients of organic pollutants in the environment (Shang et al., 1999). Sediment concentrations are usually high, much higher than what is predicted from the water levels (Jurado et al., 2007; Dueri et al., 2008). In open oceans, sediments can be considered as a final sink of POPs (Dachs et al., 2002; Lohmann et al., 2006). However, even though coastal sediments are an important reservoir of POPs globally (Jonsson et al., 2003), they are not a final sink of POPs. Coastal sediments are indeed frequently remobilized (re-suspended, refocused, and transported through canyons and along the shelf) and these remobilization events cause POPs to desorb to the dissolved phase and enter again the environmental cycle with potential volatilization, etc. (Asher et al., 2007; Yan et al., 2008; Garcia-Flor et al., 2009). This effect of sediment remobilization will be more important in shallow ecosystems. In fact, bottom seawater, sediment interstitial water, and near sediments water concentrations are dominated by diffusion of pollutants from the sediments (Jurado et al., 2007).

All the environmental processes depicted in **Figure 2-1** will drive and control the concentrations of pollutants in coastal waters and sediment. They are dynamical and concentrations are often controlled by kinetics of the processes. Jurado et al. (2007) found that concentrations at the surface are dominated by atmospheric inputs such as diffusive exchange and wet deposition (the time series of wet deposition events is shown as well). Conversely, near the sediment, concentrations are high due to diffusion from the sediment. However, in shallow coastal ecosystems and estuaries the scenario will be different where the concentration is almost constant in all the water column and the concentrations are close to equilibrium with the sediment, because it is the sediment that is supporting the water column occurrence of pollutants through diffusion and sediment resuspension. This scenario is what happening in the Hudson River estuary (Asher et al., 2007; Schneider et al., 2007), the Chesapeake Bay (Ko and Baker, 1995), and other coastal areas. Indeed, since in shallow systems pollutants released from sediments will dominate as a secondary source, any legislation and regulation of riverine and atmospheric deposition inputs will be unsuccessful in controlling pollutant seawater levels. The historical pollutant burden accumulated in the sediment will remain the source controlling the water column concentrations for a long time. This is what is now observed, for example, in off-shore Barcelona in the NW Mediterranean Sea, where the concentration of some legacy POPs such as PCBs is high in seawater (Garcia-Flor et al., 2009). These high concentrations seem to be supported by the sediment, and in fact the coastal waters act as source of PCBs to the atmosphere (Garcia-Flor et al., 2009).

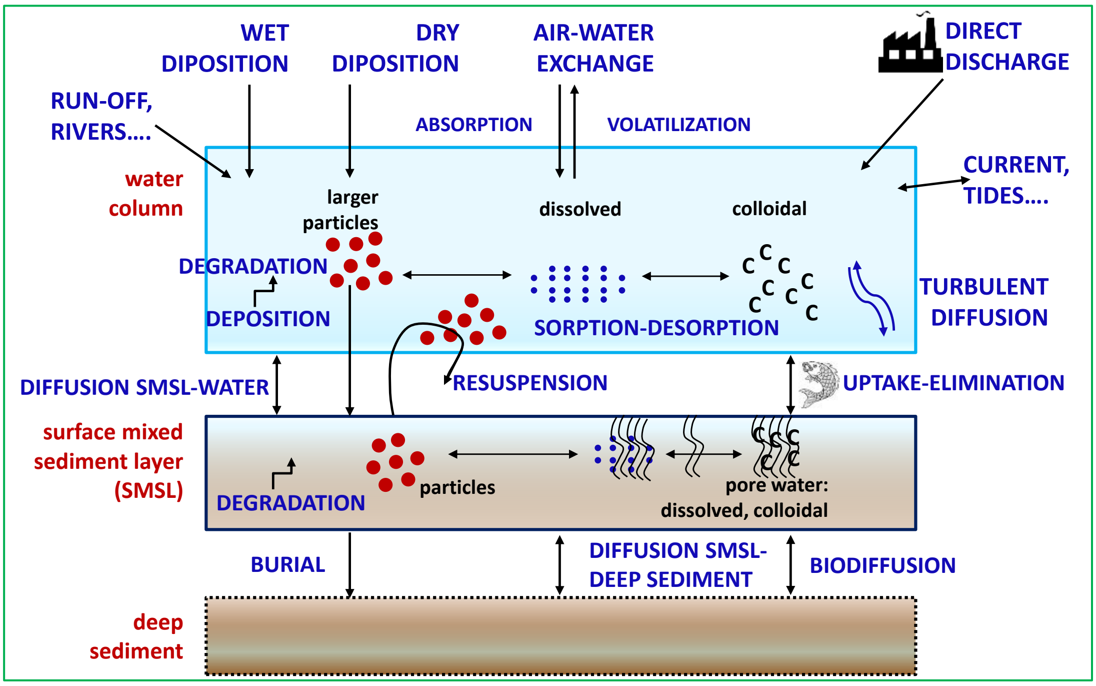

Figure 2-1. Schematics of the processes involved in the fate and transport of POPs in the coastal environment (adapted from Jurado et al., 2007).

The environmental cycling of organic pollutants are highly dependent on the compound physical–chemical properties, then except close to point sources, the environmental behavior and levels will depend on these properties. As a first classification, pollutants can be divided in swimmers, flyers, and single/multiple hoppers (Wania, 2006; Lohmann et al., 2007). Swimmers will be those chemicals that have high solubility in water and low volatility; usually these are ionic or very polar compounds such as pharmaceuticals, some herbicides, UV filters, and others. Once they have entered a water body, these chemicals follow the movement of water until they are degraded or sorbed to sediments or sinking particles. All chemicals with these properties will move similarly. The flyers are chemicals with high vapor pressures, they tend to be volatilized and remain in the atmosphere until they are degraded, The hoppers is a large group of chemicals which include most POPs, they are semivolatile, have relatively low water solubility, and high affinity for organic matter. These chemicals are those that are transported atmospherically and can enter coastal ecosystems through the different atmospheric deposition processes, even though they can also enter the marine environment through direct spills to the water body, rivers, or run-off. These chemicals, depending on their persistence and properties can undergo successive steps of volatilization/deposition, named as “grasshopping”, and thus affect all environments and ecosystems regardless of their entry vector in the environment (Wania and Mackay, 1996; Jurado and Dachs, 2008). In any case, regardless a chemical is a flyer, swimmer, or a hopper, pollutants should be viewed as travelers that will be traveling, and affecting ecosystems during their journey, until they are degraded and/or sequestered by the deep ocean. Then, pollutants, even historically deposited in estuarine or coastal sediments, will eventually be able to re-circulate in the environment, and have an effect decades or hundreds of years after their moment of introduction in the environment.

The chemicals with similar properties will move together in the environment. However, obviously, pollutants with similar physical–chemical properties can undergo fractionation during transport due to temperature effect on partitioning (Wania and Mackay, 1996), or due to selective sequestration (Jurado and Dachs, 2008). In any case, these fractionation processes will never be able to completely separate the occurrence of two similar chemicals, and only the relative occurrence of one chemical to the other can be modified to some extent during transport. Indeed, even away from point sources, concentrations of pollutants are likely correlated among them, and if a family of pollutants shows an occurrence with high concentrations, it can be predicted that high concentrations will be found of other pollutants with similar properties (see for example, Restrepo et al., 2005). In addition, even the concentrations of those pollutants with similar properties that cannot be identified due to lack of knowledge or appropriate analytical methods will also be present at high concentrations. This fact has important implications for the impact of organic pollutants in the environment and for ecotoxicological studies. Indeed, most or almost all toxicological studies have focused on the assessment of the effects of individual pollutants or simple mixtures of pollutants. In the environment, it will never happen that a given organism is exposed to a single pollutant or even a simple mixture of pollutants. In the environment, all ecosystems and organisms, including humans, are exposed to thousands of organic chemicals, most of them still unknown or never reported (Muir and Howard, 2006). There is a need for a certain “methodological revolution” in ecotoxicological studies and methodologies to incorporate these facts. In some respect, this lack of information mirrors the non-availability of a comprehensive inventory of the pollutants occurring in a water body, and the fact that the total concentration of pollutants is unknown.

Concentrations in environmental media reflect the balance of inputs, outputs, and the various environmental processes affecting them. Sediments and associated interstitial waters are among the media with higher concentrations, much higher than concentrations in suspended particles and surface waters. This is due to the important depositional fluxes of pollutants to the sediments and conversely, to less efficient fluxes in the other direction, from the sediment to the water column. This implies than during periods when a chemical is released to the environment through primary sources, the level of pollutant concentrations in sediment will increase, since deposition is higher than resuspension. If the primary source disappears due to regulatory measures or change in use-profile, the concentration of the chemical decreases in the water body and the settling flux will also decrease; then the sediment can become a source to the water column. In coastal areas with the absence of direct inputs from anthropogenic activities, the occurrence of pollutants will be dominated by atmospheric deposition (Asher et al., 2007; Van Ry et al., 2000; Yan et al., 2008; Dachs and Méjanelle, 2010).

2.2 POPs distribution coefficients

The partitioning of the POP between the water phase and particles or other sorbing phases can be expressed as a solid-water partition coefficient (K_D), which is the ratio between the concentrations in the solid phase (C_S) and the water phase (C_W) at equilibrium: $K_D = C_S/C_W$. Since organic matter is considered the main sorbing phase for POPs (Schwarzenbach, et al., 2003; Rabodonirina et al., 2015; Soubaneh et al., 2015), partitioning can be estimated from the organic carbon content of the phase (f_{OC}), and the partition coefficient between organic carbon and water (K_{OC}) for the compound: $K_D = f_{OC} \times K_{OC}$. The organic carbon-water partition coefficient (K_{OC}) can in turn be predicted from the hydrophobicity of the POP in the form of the octanol-water coefficient (K_{OW}), for instance from the relationship $K_{OC} = 0.35\ K_{OW}$ suggested by Seth et al. (1999) for soil and sediment organic matter or the relationship $K_{DOC} = 0.08\ K_{OW}$ suggested by Burkhard (2000) for dissolved organic matter. However, the degree of uncertainty is large in both suggested regressions, partly due to analytical differences but also due to the large variability in the organic matter in soils, sediment, and water (Burkhard, 2000). The variability in the organic matter quality affects the sorption of POPs. For instance, terrigenous material generally sorbs POPs better than planktonic material (Gustafsson et al., 2001), which may be due to the better sorption to the more aromatic structure in the terrestrial colloids (Gustafsson et al., 2001), and humic acids tend to display higher sorption of POPs than fulvic acids (Burkhard, 2000; Kuivikko et al., 2010). Observations of more enhanced sorption of POPs to soil and sediment than could be explained with the organic matter partitioning model led to the development of a model with dual-phase sorption domains, where the sorption can be explained with both (linear) absorption into amorphous organic matter, and (nonlinear) adsorption onto black carbon (BC) and other carbonaceous georesidues such as coke (Accardi-Dey and Gschwend, 2002; Cornelissen et al., 2005). The BC sorption is more favourable for compounds with a planar structure (Cornelissen et al., 2005; Jonker and Koelmans, 2002). The dual-mode sorption has been connected to the observation of a rapid and a slow phase in kinetic studies on sorption/desorption of organic compounds to natural particles. In the rapid phase (hours or days), the hydrophobic compounds sorb/desorb in the outer regions or the particle, but by time (months), they can migrate to/from the inner regions. The slow desorption can be caused by the energy required to break bonds between the compound and the natural particle, or the resistance to mass transfer when the compounds are diffusing through the matrix (Pignatello and Xing, 1996). Sorption to amorphous OC would correspond to the rapidly desorbing phase, while sorption to BC equals the slowly desorbing phase (Cornelissen et al., 2005). A related concept of relevance for the POP bioavailability is aging, the phenomenon that the bioavailability of POPs in soils and sediments can decrease with time (Alexander, 2000). However, as demonstrated by Birdwell and Thibodeaux (Birdwell and Thibodeaux, 2009), if the labile fraction (rapid release fraction) is removed, it will be replenished from the nonlabile (slow release) fraction (Josefsson et al., 2010, 2011).

2.3 Sources of organic pollutants in coastal ecosystem

Organic pollutants can enter the environment due to a myriad of sources and a number of processes. Sources can be divided between primary and secondary. Primary sources are those directly related to the use of chemicals. For example, PCBs can be accidentally released from an old electrical transformer. Conversely, secondary sources are occurring once the pollutant enters the environment due to its cycling in the environment through transport and transformation. For example, when PCBs are volatilized from soil and water, thus soils and surface waters become secondary sources. Secondary sources are usually diffuse sources, while primary sources can be diffuse or point sources. All regulatory efforts have focused in controlling the primary sources, while secondary sources are not regulated.

2.3.1 Point Sources Versus Diffuse Sources

Organic pollutants can have a multitude of point sources. It is important to take into account that not only the point sources resulting in direct spills or release of a pollutant to the water body are relevant to coastal pollution. In this sense, it is a misconception to consider rivers as the main source of pollutants in estuaries and coasts, and to see rivers as main point sources to the coastal system. A pollutant applied to a field as an herbicide can be volatilized and after atmospheric transport be deposited to coastal waters, maybe hundreds of kilometers from the source. For a given organic pollutant, the relative importance of these diffuse and local point sources will depend on the given ecosystem. For example, it has been shown that riverine flux of herbicides is related to watershed characteristics (Capel and Larson, 2001). Atmospheric deposition is a clear example of another diffuse source of pollution to coastal ecosystems, and the level of pollutants in a water body will never be lower than that derived from the combination of the different atmospheric deposition processes, regardless of the managing practices in the watershed or costal area. Atmospheric deposition is notably important in remote coastal areas. For example, Arctic and Antarctic coastal systems are affected by high concentrations of POPs derived from atmospheric deposition (Tomy et al., 2000; Fisk et al., 2001) being these concentrations amplified due to an effect known as global distillation of POPs through ‘grasshopping’ and sequestration in cold environments (Wania and Mackay, 1996; Jurado and Dachs, 2008). Near to point sources, these can determine the occurrence and levels of pollutants in waters, sediments, and biota. However, in the background environment, just some kilometers away from point sources, the diffuse sources are the dominant control on pollutants occurrence and levels. Atmospheric deposition processes are a conspicuous diffusive source for surface coastal waters: the concentrations of organic pollutants will never be significantly lower than those equilibrated with the atmosphere regardless of the efforts done for regulating water concentrations. This simple and well-known fact shows that water and atmosphere legislations have to be harmonized to promote the efficient control of organic pollutant levels in many ecosystems. The control of background levels by equilibration with atmosphere is not considered in the EU water directive nor by other regulations. In this respect, there is a generalized misconception in all national and international legislation related to organic pollutants.

2.3.2 Primary Versus Secondary Sources

One of the current research topics of interest is to determine the predominant sources and whether the environmental occurrence and impacts are dominated by primary and secondary sources. This is also important in terms of regulatory efforts since these can only affect, in most cases, the primary emissions of POPs. In general, the occurrence of emerging pollutants such as pharmaceutical products, or UV filters, or current use herbicides are dominated by primary sources of these pollutants in the environment. However, for legacy POPs, such as those regulated in the Stockholm Convention (UNEP, 2001) their occurrence is often dominated and controlled by secondary sources. Indeed, chemicals such as PCBs or dichlorodiphenyltrichloroethanes (DDTs) have entered already in the global environment, and their environmental fate and exposure by biota is controlled, in many ecosystems, by the release from secondary sources (surface waters, soils, and sediments) and how they cycle in the environment (Lohmann et al., 2007). Currently, secondary sources can be more important than primary sources for legacy pollutants such as PCBs, DDT, etc., and even, in some ecosystems, for current use (or new emerging) chemicals such as PFAAs, PBDEs, PAHs, etc. Regulation of emissions is a complicated task and sometimes generates no perceptible improvement at short term. Sometimes regulation of the emissions of some chemicals, even though due to primary emission, can be problematic due to their diffuse character. For example, emissions of PCDD/F as minor byproduct of combustion processes can only be regulated to a certain value due to the myriad of combustion sources present in a given region. The same comment applies to the regulation of emissions of PAHs from combustion of fuel in vehicles and/or combustion of wood or wooden materials in households.

2.4 POPs in various environmental matrices

Most of the POPs are typically `water-hating' and `fat-loving' chemicals, i.e. hydrophobic and lipophilic. In aquatic systems they partition strongly to solids, notably organic matter, avoiding the aqueous phase. They also partition into lipids in organisms rather than entering the aqueous milieu of cells and become stored in fatty tissue. This confers persistence on the chemical in biota since metabolism is slow and POPs may therefore accumulate in food chains. Humans, through ingestion of contaminated fish and shellfish, may be exposed to elevated levels of POPs (Ross and Birnbaum, 2003).

To add to the proliferation of data, many national and international monitoring programs have been developed to assess POPs in the marine/coastal environment. Such programs include, for example, the National Status and Trends assessments in the United States (US), undertaken by the National Oceanographic and Atmospheric Administration (NOAA) (since 1984); various Mussel Watch monitoring activities, especially in the US (e.g. the California Mussel Watch, since 1986); the USFDA Pesticide Residue Monitoring Program (since 1987); the USEPA Environmental Measurement and Assessment Program (since 1988); the Dioxin Monitoring Program by the US State of Maine (since 2001); and the Global POPs Monitoring Program (GMP), established to evaluate the effectiveness of the Stockholm Convention. These studies have contributed valuable information to global monitoring, but the key question to be addressed concerns their effectiveness in producing definitive results which reliably identify spatial and temporal differences in contaminants, let alone providing data which may be used to assess environmental risks or to set reference standards and guidelines.

In this section (section 2.4 of this chapter), I discussed briefly on the previously published literatures related to POPs in various coastal/marine environmental matrices (surface water, sediment and biota), particularly in Asian countries. In recent decades, with the rapid economic development in Asia, the effects of various persistent pollutants on human health and the ecosystem have caused increasing concerns among scientists and risk assessors and managers in the region and beyond. However, for decades research on POPs in Asia has lagged behind those in Europe and North America. Data are limited and vary greatly in quality. In addition, information is scattered among numerous journals and documents, many of which are in native languages.

PCBs primarily accumulate in soils and sediment as a result of spills, leaking toxic landfills, or contamination from products containing the chemicals. While PCBs do pollute the air via volatization and dispersion, the contaminants are most problematic in soils and sediments where they adhere to organics and are very slow to degrade. The primary route of exposure for humans and wildlife is through the ingestion of contaminated dietary items. A number of studies from both the developed and developing countries of Asian region showed environmentally concerned levels of PCBs in various matrices of the ecosystem, particularly in surface water, sediment and biota (Kajiwara et al., 2004; Kubota et al., 2002; Okumura et al., 2004; Honda et al., 2008; Nakata et al., 2002; Nie et al., 2005; Mai et al., 2002; Liu et al., 2003; Hong et al., 2011; Ramu et al., 2007; Jeong et al., 2001; Carvalho et al., 2008; Wurl and Obbard, 2005; Ilyas et al., 2011; Bhattacharya et al., 2003; Das et al., 2002; Babu Rajendran et al., 2004; Farooq et al., 2011; Sanpera et al., 2002; Eqani et al., 2012). PCBs are highly lipophilic and dissolve in fatty tissues and bioaccumulate over an organism's lifespan. This property is important to both human and ecological toxicology because bioaccumulation leads to biomagnification, the process by which persistent toxins increase in concentration upward through the food chain (Faroon et al., 2003). As a result, the highest concentrations of PCBs are often observed in top predators with long life-spans and high fat deposits such as humans. A positive relationship between PCB concentrations in human samples and dietary intake of fish and shellfish have been reported (Kostyniak et al., 1999; Stewart et al., 1999; Bocio et al., 2007). Furthermore, monitoring of PCBs in food has been used to assess human exposure in a number of countries (Nakata et al., 2002; Moon and Ok, 2006). Although the concentrations of PCBs in various environmental matrices have decreased dramatically since peaking in the 1970s (Jones et al., 1992; Mason, 1998; Schneider et al., 2001; Schuster et al., 2010), they continue to bioaccumulate in organisms and be categorized as major global contaminants. Jonsson et al. (2003) estimated that human exposure to PCBs is expected to continue for decades and perhaps centuries because of the very long global environmental mean residence times of these pollutants.

Due to high energy of carbon–fluorine bonds, PFAAs are resistant to hydrolysis, photolysis, microbial degradation, and metabolism by vertebrates (Kissa, 2001). PFAAs, especially perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), were first reported to be widespread in the environment (Giesy and Kannan, 2001, 2002), and they were subsequently detected in aquatic systems (Fujii et al., 2007; Rayne and Forest, 2009) and wildlife (Kannan et al., 2002; Houde et al., 2006; Suja et al., 2009). Many PFAAs can be accumulated in aquatic system, leading to bioaccumulation and biomagnification through the food chain to wildlife and humans (Loi et al., 2011; Squadrone et al., 2014). There is still long-term public concern over adverse effects of PFAAs on ecosystem and human health as well as secondary release of PFAAs from the environment (Pico et al., 2011; Lu et al., 2013). In recent years, great attention has been paid to PFOS and PFOA, two of the frequently detected predominant PFAAs in the environment (Wang et al., 2015). Given their water solubility and negligible vapor pressure when dissolved in water, most PFAAs can accumulate in aquatic systems and are readily transported by hydrological processes (Taniyasu et al., 2003; Yamashita et al., 2005). Water and sediments are considered final sinks of PFAAs and aquatic systems are an important medium for their transport (Hansen et al., 2002).

Studies about PFAAs in Asian countries have been focused primarily on the aspects including detection of PFAAs in different environmental media, risk assessment of PFAAs on environment and ecosystem, toxicology to humans and wildlife, and estimation of sources and emission (Wang et al., 2010a,b; Cai et al., 2012; Wu et al., 2012; Xie et al., 2013a,b; Yeung et al., 2009; Zhang et al., 2011; So et al., 2004; Rostkowski et al., 2006; Yoo et al., 2009; Taniyasu et al., 2003; Lein et al., 2008). However, reports regarding PFAAs in coastal environmental compartments are still scarce in Asian developing countries, particularly in Bangladesh.

PAHs with carcinogenic and mutagenic characteristics are an important class of organic pollutants. The fates of these compounds in the environment and the remediation of PAH-contaminated sites are, therefore, of high public interest (Yun et al., 2008). The incomplete combustion of materials associated with high temperature industrial processes is the main anthropogenic source of PAHs. PAHs are released into the environment through wastewater effluents, coke and petroleum refining industries, accidental oil spills and leakages, aerial fallout, rainwater runoff, forest and prairie fires, vehicle traffic, and domestic heating (Yuan et al., 2014). Due to their persistence and semi-volatile nature, PAHs are widely distributed in diverse biotic and abiotic compartments, particularly in the coastal ecosystems. The coastlines of tropical and subtropical regions, are exposed to anthropogenic contamination by PAHs from tidal water, river water and land-based sources via coastal discharges (Sarkar et al., 2012). The source identification and risk assessment of PAHs has been investigated in coastal surface water (Li et al., 2006; Chizhova et al., 2013; Wu et al., 2011; Ren et al., 2010; Li et al., 2015; Lim et al., 2007), sediment (Hu et al., 2011; Froehner et al., 2010; Zakaria et al., 2002; Boonyatumanond et al., 2006; Cho et al., 2009), and biota (Isobe et al., 2007; Nakata et al., 2003, 2014; Wan et al., 2007; Moon et al., 2004).

Due to their high hydrophobicity and strong particulate-oriented behaviors, PAHs are partitioned preferentially to sediments in aquatic ecosystems (Parolini et al., 2010; Kannan et al., 2005; Colombo et al., 2006). Therefore, sediment is the most important sink for PAHs in the aquatic environment. Once deposited in sediments, PAHs are less subjected to photochemical or biological oxidation, especially if the sediment is anoxic. Thus, sedimentary PAHs tend to be persistent and may accumulate to high concentrations (Cho et al., 2009). Elevated concentrations of PAHs (>10,000 ng g^{-1} dry weight) have been recorded in coastal mangrove sediments (Tam et al., 2001; Ke et al., 2002). Because of their potential (or, in some cases, proven) carcinogenicity, immunotoxicity, genotoxicity, and reproductive toxicity, PAHs in contaminated sediments can directly affect sediment-dwelling organisms leading to bioaccumulation and biomagnification through the food chain to wildlife and humans (Sverdrup et al., 2002; Gu et al., 2003; Sarkar et al., 2012). Therefore, the determination and monitoring of PAHs in environmental samples is necessary and important to human health. Recently, extensive input of petrogenic PAHs to the Southeast Asian coastal zone has been reported (Zakaria et al., 2002). However, few data are available for PAH contamination in the coastal environmental matrices of the South Asian countries, such as: India (Dominguez et al., 2010; Saha et al., 2009; Dhananjayan et al., 2012), Pakistan (Farooq et al., 2011; Aziz et al., 2014), Sri Lanka (Pathiratne et al., 2007). Unfortunately, no comprehensive study regarding PAH contamination has been carried out yet in the coastal environment of Bangladesh.

2.5 Ecotoxicological effects of POPs

The ecotoxicological effects of POPs in the environment have caused much concern in recent years, and this has led to the control or complete ban on the use of these chemicals in many countries. Among the wide range of organic substances contaminating the aquatic environment a major concern has so far focused on PCBs, PFAAs, and PAHs. High persistence and resistance to bio-degradation of these toxic pollutants make them continue to be yet largely present in the coastal environment. POPs are found not only in living organism as animals and plants, but also in humans (Sweetman et al., 2005; El-Shahawi et al., 2010).

Public concern about POPs contamination increased recently because several of these compounds are identified as hormone disrupters which can alter normal function of endocrine and reproductive systems in humans and wildlife. There are many risks and effects of having these chemicals in our environment and none of them are a benefit to the earth. After these pollutants are put into the environment, they are able to stay in the system for decades causing problems such as cancer, birth defects, learning disabilities, immunological, behavioral, neurological and reproductive discrepancies in human and other animal species (Sweetman et al., 2005; El-Shahawi et al., 2010).

POPs contaminate food, water and accumulate in the food chain such as eagles, polar bears, killer whales and human being. There is evidence that many people worldwide may now carry enough POPs in their body fat where POPs accumulate to cause serious health effects, including severe illness and eventually death. Laboratory investigations and environmental impact studies in the wild have implicated POPs in endocrine disruption, reproductive and immune dysfunction, neurobehavioral and disorder and cancer (Pauwels et al., 2000; Katsoyiannis and Samara, 2005).

More recently some POPs have also been implicated in reduced immunity in infants and children, and the concomitant increase in infection, also with developmental abnormalities, neurobehavioral impairment and cancer and tumor induction or promotion. Some POPs are also being considered as potentially important risk factor in the etiology of human breast cancer by some authors (Roots et al., 2005). Human exposure to POPs is carried through the food chain. Because the animals are subsisting in water contaminated with these pollutants, they are affected as well when the humans consume these animals, they have also consumed the pollutant which then is accumulated in the body (Katsoyiannis and Samara, 2005). Because children are still developing, they are much more susceptible to the effects of pollutants. Their developing cells are sensitive to contaminants and are more likely to be affected by exposure of POPs. The brain is apparently in the greatest concern because some studies have shown that children exposed to POPs during infancy had remarkably lower scores on assessments determining intelligence and ability to shut out distractions (Bouwman, 2003; Bolt and Degen, 2002). At a young POPs can have serious consequences side effects such any of the following: birth defects, certain cancer and tumors at multiple sites, immune system disorders, reproductive problems, reduced ability towards off diseases, stunted growth and permanent impairment of brain function, POPs are a suspected carcinogen, diseases such as endometriosis (a painful, chronic gynecological disorder in which uterine tissues are affected), increased incidence of diabetes and others and neurobehavioral impairment including learning disorders, reduced performance on standard tests and changes in temperament (Bolt and Degen, 2002).

2.6 Risk assessment

2.6.1 Ecological risk assessment

POPs are strongly particle-associated in aquatic ecosystems due to their hydrophobic properties, and tend to accumulate in sediments. Contaminated sediments may constitute a particular threat for the associated biota and even for other organisms throughout the marine food web (Gomez-Gutierrez et al., 2007). The importance of polluted sediments as a cause for impairment of coastal water bodies has led to the development of several methods for assessing sediment quality (USEPA, 2000). In the screening-level process, exposure estimates are based on site-specific data. The effects are often estimated by the application of specific sets of sediment toxicity benchmarks, the sediment quality guidelines (SQGs), which are usually adopted to describe the levels of contaminants in sediments associated with different categories of adverse effects (Hill et al., 2000). SQGs have been demonstrated to be useful tools for assessing the quality of freshwater, estuarine and marine sediments (USEPA, 1996; MacDonald et al., 2000; Long et al., 2006). They have been derived with a variety of theoretical methods that rely upon analyses of matching, field-collected chemistry and biological effect data (Wenning et al., 2004). Moreover, SQGs provide a basis for interpreting whole-sediment chemistry data by identifying the concentrations of chemicals of potential concern that can cause or contribute to adverse effects on sediment-dwelling organisms (Long et al., 2006). There are several primary approaches for estimation of the biological effects of contaminated sediment based on chemical data alone (Casado-Martinez et al., 2006). For example, the equilibrium partitioning model (EqP-model) proposed by Di Toro et al. (1991), the effects range approach (Long et al., 1995), the effect level approach (MacDonald et al., 1996) and logistic regression models (USEPA/NOAA, 2005) are some of these methods. Since the plethora of proposed SQGs exhibit great variability, a consensus-based approach was developed that attempted to reconcile existing values (Swartz, 1999; MacDonald et al., 2000; Binelli et al., 2008).

2.6.2 Human health risk assessment

The risks of exposure to POPs are evaluated in risk assessment processes. These typically consist of a hazard identification followed by hazard characterization, exposure assessment and, finally, a risk characterization (EC, 2000). The hazard identification typically involves the identification of the contaminant and of the effects that are considered as adverse (EC, 2000). The identification traditionally follows from in vivo (animal) experiments. However, with the aim of reducing animal experiments, other approaches gain more importance such as computational toxicology and in-vitro toxicology evaluation (Hamers et al., 2006; Harju et al., 2007). The hazard characterization describes the process of quantification of the relevant adverse effects. This is often referred to as the dose-response relationship. This results in benchmarks such as the no observed-adverse-effect-level (NOAEL), which is the level of exposure of which the effects in the treated animals do not differ significantly from those in the untreated (control) animals. The exposure assessment aims at characterization of the nature and size of the human population exposed to an emission source and the magnitude, frequency and duration of that exposure (EC, 2000). Finally, risk characterization relates to the estimation of the probability of the occurrence and the severity of adverse effects in a certain human population, based on the previous three stages by comparing the estimated exposure and the hazard characterization (EC, 2000).

2.7 Conclusion

The history of aquatic environmental pollution goes back to the very beginning of the history of human civilization. However, aquatic pollution did not receive much attention until a threshold level was reached with adverse consequences on the ecosystems and organisms. Aquatic pollution has become a global concern, but even so, most developing nations are still producing huge pollution loads and the trends are expected to increase. Knowledge of the pollution sources and impacts on ecosystems is important not only for a better understanding on the ecosystem responses to pollutants but also to formulate prevention measures. Many of the sources of aquatic pollutions are generally well known and huge effort has been devoted to the issue. Although several persistent organic compounds have been banned but still many are in use illegally, particularly in developing countries. In the South Asian context, India is the largest producer and consumer of persistent organic compounds in the region and found highly contaminated country in the region. Pakistan is found to be the second contaminated country in the context of banned chemicals. Bangladesh, Nepal and Sri Lanka are also facing such conditions of contamination but it is difficult to draw a true picture of contamination levels in these regions due to lack of sufficient information or evidence. Therefore, comprehensive researches are needed to assess the distribution, occurrence, sources and toxic effects of POPs in the region. Keeping in view a direct and pointed effort shall be made to enforce the law of the land and to strictly stop the usage of banned persistent organic compounds. More effort shall be made to scientific studies and researches in the region and a proper data shall be maintained so that along with awareness a check on contamination could be maintained effectively.

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Chapter 3

POLYCHLORINATED BIPHENYLS (PCBS) IN SURFACE WATER, SEDIMENT AND SEAFOOD FROM COASTAL AREA OF BANGLADESH AND RISK ASSESSMENT

Polychlorinated biphenyls (PCBs) in surface water, sediment and seafood from coastal area of Bangladesh and risk assessment

Abstract

A comprehensive congener specific evaluation of polychlorinated biphenyls (PCBs) was conducted for the first time in Bangladesh. All 209 congeners of PCBs in surface water, sediment and commonly consumed seafood from the coastal areas of Bangladesh were analyzed by GC-MS/MS. Samples were collected in winter and summer 2015. Regardless of season and site, the total concentrations of PCBs (∑PCBs) were 32.17–199.35 ng/L, 4.61–105.25 ng/g dw, and 3.82–86.18 ng/g ww in water, sediment and seafood samples, respectively. The levels of ∑PCBs were either at the middle or lower end of reported global range. No significant seasonal variation was observed in the levels of ∑PCBs in either medium. Spatial distribution revealed that the areas with recent urbanization and industrialization (Chittagong, Cox's Bazar and Sundarbans) were more contaminated with PCBs than the unindustrialized area (Meghna Estuary). PCB profiles were dominated by moderately chlorinated (4–6 Cl) homologs. Our analyses elucidated that the past and on-going use of PCB-containing equipment (e.g. transformers and capacitors being operated in the electrical generating sector) as well as the anthropogenic activities such as urban developments, commercial and industrial establishments (e.g. ship breaking and port activities) are likely the major sources of PCB contamination in Bangladesh. The top congeners based on dominance by both occurrence and abundance were identified as potential markers of PCBs, which can be used for future selective monitoring in case of reasonable constraints on full congener approach. From the ecotoxicological point of view, PCB concentrations in the Bangladeshi coastal area exceeded some of the existing national and international environmental quality guidelines/standards, suggesting potential threat to the aquatic organisms and human health as well through biomagnification. In addition, the coastal residents are sufficiently exposed to the dietary PCBs through seafood consumption which may cause severe health risk including dioxin like toxic effects.

3.1 Introduction

Polychlorinated biphenyls (PCBs) are a class of chlorinated organic compounds with various industrial and commercial applications due to their low flammability, chemical and thermal stability, and electric insulating properties. PCBs were used in a wide range of applications such as dielectric fluids in transformers and capacitors, coolants, flame retardants, hydraulic oils, lubricants, printing ink and dye carriers, pesticide and wax extenders, and additives in paints, plastics, adhesives, sealants, and other products (ATSDR, 2000). PCBs have up to 10 chlorine (Cl) atoms connected to two phenyl rings and the variation in number and position of the Cl atoms results in 209 possible configurations, or congeners. Some PCBs are termed "dioxin-like" for having a structure similar to 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD), recognized as the most toxic organic compound (McFarland and Clarke, 1989). Twelve dioxin-like PCBs (DL-PCBs) comprising four non-ortho (PCBs 77, 81, 126, 169) and eight mono-ortho (PCBs 105, 114, 118, 123, 156, 157, 167, 189) congeners are of particular concern due to their toxicity and are thus recommended for monitoring worldwide. Conversely, the remaining congeners, referred to as the non-dioxin-like congeners (nDL-PCB), exert weak or no effect on Ah-receptors; however, they interfere with intracellular signaling pathways that are regulated and modulated by Ca^{2+}, such as those involving ryanodine receptors, protein kinase C, inositol triphosphate or arachidonic acid, and, thus, cause neurotoxicity (Simon et al., 2007). PCBs are highly persistent due to their resistant to biological and chemical degradation. They are ubiquitous in the environment (Fowler, 1990) as a result of widespread usage since their first commercial synthesis in 1929 and being prone to large-scale dispersal through atmospheric and oceanic transport and biologically mediated mechanisms such as bioaccumulation (Goerke and Weber, 2001; Gouin et al., 2004). The PCB levels are further biomagnified up in the food chain (Porte and Albaigés, 1993). The food-chain transfer of PCBs thus poses potential ecological and human health risks (Batang et al., 2016).

The marine or coastal environment serves as a sink for PCBs and other persistent organic pollutants (POPs). These contaminants occur in almost all water bodies, but often at elevated levels in industrialized and densely populated coastal areas (Fowler, 1990). The deterioration of water quality leads to a decreasing amount and diversity of the biota in the aquatic system, as well as to a decline of the quality of aquatic products. It is reported that more than 90% of persistent organic pollutants in human tissue, such as chlorinated hydrocarbons, originates from food (Safe, 1998; Easton et al., 2002), and of these, fish and seafood is one of the major routes of accumulation (Harrison et al., 1998; Alcock, 1998). This will pose health hazards over a period of time (Moore et al., 2002). A growing global concern over such risks has led to the adoption in 2001 and enforcement in 2004 of the Stockholm Convention on POPs, an international treaty that seeks to eliminate or restrict the intentional production, distribution, and use of toxic POPs, including PCBs. The treaty aims to protect human health and the environment from the deleterious effects of POPs based on precautionary principle (UNEP, 2002). According to the Convention, all parties should eliminate the use of equipment containing PCBs by 2025 and make determined efforts to achieve environmentally sound management of wastes containing greater than 50 ppm PCBs by 2028. Upon becoming a party to the Convention, every country is required to submit reports every 5 years to the governing body of the Convention on their progress in eliminating PCBs and other POPs. As a party to the Stockholm Convention, Bangladesh is obligated to abide by the objectives of reducing and ultimately eliminating these pollutants and is encouraged to conduct research on POPs. Hence, an important new stage has started for action to be taken to cease of use of PCB containing equipment, preparation of inventories, identification of polluted sites and eventually, environmentally safe disposal of wastes and remediation of contaminated sites. Identification of sites contaminated with PCBs and their subsequent remediation is a major effort which initially requires a broader approach due to the relatively small amount of information currently present. Hence, many countries, including Bangladesh, need to gather all relevant information leading to the identification of pollution hot spots with the ultimate aim of environmentally sound remediation.

Contrary to the extensive information on PCB use, inventory and disposal over the world, fairly little is known about the status of PCBs in Bangladesh. PCBs have never been manufactured in Bangladesh. PCB mixtures, PCB-containing equipment and other materials (e.g. electrical power transformers, capacitors, lubricating oils etc.,) have been imported to Bangladesh for an indeterminate period, as records were not maintained (DoE, 2007). There are several suspected local emission sources of PCBs in Bangladesh, for example, PCB-containing equipment, PCB stockpiles, landfill sites, ship breaking industries, etc. (ESDO, 2005a,b; 2010; Nøst et al., 2015). PCB compounds are still in use in Bangladesh, mostly in closed systems as dielectrics in transformers and capacitors used in the electrical generating sector (DoE, 2007). PCB content of in-service electrical equipment is estimated at 51.6 MT, of which 48.9 MT is thought to be in distribution transformers and 2.7 MT in other types of equipment. Furthermore, the total electrical sector PCBs requiring destruction is estimated at 55.8 MT (DoE, 2007). There is no formal management and recycling of obsolete PCB-containing equipment and this waste is simply deposited in landfills in many areas. Ship breaking sites are another threat since they release PCBs and other POPs into the environment. The total quantity of PCBs estimated to be contained within the ships dismantled each year in Bangladesh is about 22.5 MT, of which, a significant portion have been disposed in the ground as well as spilled into the environment (DoE, 2007). This has already led to PCB pollution issues in the surrounding area (Wurl et al., 2006; Chakraborty et al., 2013). Other potential sources include release and/or generation of unintentionally produced PCBs by numerous industrial activities including waste incinerators, ferrous and non-ferrous metal production, power generation and heating, production of mineral products, transport sector, other combustion processes such as waste burning and accidental fires, etc. In addition, metal-working lubricants, sealants, plasticizers, paint additives and flame retardants are likely widely used in Bangladesh, which may contain PCBs and other POPs that can also be considered as potential sources (ESDO, 2005a,b; 2010; Nøst et al., 2015).

Bangladesh is an exclusively riverine agricultural country that is undergoing rapid industrialization, urbanization and economic development in recent years, particularly in the coastal regions. The country has a highly irregular deltaic marshy coastline of 580 kilometers, divided by many rivers and streams that enter the Bay of Bengal, which is characterized by a tropical climate. The coastal areas of Bangladesh suffer from environmental degradation due to rapid human settlement, the development of industrial hubs, tourism and transportation, extensive ship breaking and port activities, illegal and mismanaged dumping of e-waste, the operation of an excessive number of mechanized boats, deforestation, and increasing agriculture and aquaculture activity and large discharges of untreated and semi-treated domestic and municipal sewage. There are also effluents containing heavy loads of organic and inorganic pollutants from many large and small local industries. With the population and industrial pressures increasing along the rivers, the estuarine and coastal areas of Bangladesh face threats to their coastal ecosystems from contaminants such as PCBs, which can accumulate in the coastal or marine food chains. Local populations can be exposed to these chemicals via contaminated seafood consumption. About 42 million people (30 % of the total population) live in the coastal area (47,211 km^2; 32 % of the total land area) of Bangladesh, of which about 5 million are engaged directly in commercial fishing (BOBLM, 2011). Seafood is the most important and one of the major dietary components of the coastal populations. Therefore, it is an urgent need to assess the potential health risk that might be posed through PCB-contaminated seafood consumption.

Bangladesh's inland aquatic environments are recognized among the most polluted ecosystems in the world (due to various types of organic and inorganic pollutants; Evans et al., 2012). A very recent study on the pollution status of the coastal area of Bangladesh also revealed severe inorganic contamination in different environmental matrices (water, sediment and biota). Trace metals [e.g., Arsenic (As), Chromium (Cr), Lead (Pb), Zinc (Zn), etc.] were found in the study area, some of which exceeded the guidelines for the protection of the environment and human health (Raknuzzaman et al., 2015; Raknuzzaman et al., 2016). However, to the best of our knowledge, there have been no comprehensive studies of organic pollution in this region, particularly PCBs. Hence, the present study was initiated. This study is the first attempt to determine the levels of all PCB congeners in surface water, sediment and edible seafood (finfish and shellfish) from the coastal area of Bangladesh. The compositional pattern and spatiotemporal distribution of PCBs in the coastal areas of Bangladesh were investigated. A preliminary risk assessment was carried out to elucidate the degree of PCB contamination in the Bay of Bengal coast in Bangladesh.

3.2 Materials and methods

3.2.1 Study area and collection of samples

Four sampling sites (Cox's Bazar, Chittagong, Meghna Estuary and Sundarbans) with fourteen different locations were investigated in the southeast and southwest coastal areas of Bangladesh. A sampling map with sampling sites and locations is shown in **Figure 3-1A**. Sampling sites were chosen in coastal areas to show the influence of the potential pollution sources (cities, industrial areas, rivers). The first site, Cox's Bazaar (Site 1), is a natural sandy sea beach. It was divided into two sub-sites based on ecological features: the hatchery area (CX1–CX2) and the Bakkhali Estuary (CX3–CX4). The second site, Chittagong (Site 2), is located near the Chittagong port (CT1–CT2) and ship breaking area (CT3–CT4). The third site (ME1–ME3), Meghna Estuary (Site 3), is an estuarine area where the main rivers mix into the Bay of Bengal. This site is influenced mainly by the domestic and industrial effluents carried by the inland rivers from Bangladesh and neighboring countries. The fourth site (SN1–SN3), Sundarbans (Site 4), is located near the southwest part of the coastal area, and it is regarded as the largest mangrove ecosystem in Bangladesh. A detailed description of the study area is presented in Appendix A, and the coordinates of sampling sites and location IDs are listed in Table A-1 (Appendix A).

Surface water ($n=28$) and sediment ($n=28$) samples were collected from the coastal area of Bangladesh in January-February and August-September 2015. The sampling times represent two distinct seasons, winter (dry season) and summer (rainy season), respectively. The tides in the study area are semi-diurnal (two nearly equal high and low tides each day), and the samples were collected during low tide. Three composite samples of surface water, approximately 2 L each, were collected at each location in clean polypropylene (PP) bottles. The bottles were rinsed with deionized water, methanol, acetone, and water from the particular sampling location prior to use. Immediately after collection, the samples were filtered through 0.45 µm membranes to remove large particles and biota, and they were transferred to new PP bottles that were pre-washed with methanol, acetone, and deionized water. The surficial sediment samples (top 0–5 cm) were taken using a portable Ekman grab sampler. Three composite samples with masses of approximately 200 g were collected from each sampling location and kept in polyethylene (PE) ziplock bags. Samples were transported in boxes packed with ice stored at −20 °C in a freezer upon arrival at the laboratory of the Department of Fisheries, University of Dhaka. Once there, they were treated within 48 h. Sediment samples were freeze-dried, ground, then kept in PP bottles and stored at −20 °C. All containers used during the process of sample collection, pretreatment, storage and transportation were carefully handled to avoid contamination.

Seafood samples were collected from four main fish landing centers located in the four major coastal areas of Bangladesh (Cox's Bazar, Chittagong, Bhola and Sundarbans; **Figure 3-1B**) which contribute approximately 70–80% of the total catchment of the seafood. The seafood from these fish landing centers are then sold and distributed locally and nationally through a straight marketing channel (e.g. fisherman → fish landing center → fish trader → fish market → consumer). Moreover, the catchment from these fish landing centers also contributes a major portion to the export of Bangladeshi seafood to the overseas countries. A total of 48 seafood samples (5 finfish and 2 shellfish species) were collected in winter and summer of 2015, of which the finfish species included Ilish (*Tenualosa ilisha*), Rupchanda (*Pampus argentius*), Loitta (*Harpadon nehereus*), Sole (*Cynoglossus lingua*), and Poa (*Otolithoides pama*), whereas the shellfish species included shrimp (*Penaeus indicus*) and crab (*Scylla serrata*). The varieties were the most commonly consumed seafood in the coastal area of Bangladesh. Rupchanda and Sole were not collected from Bhola and Sundarbans area due to species unavailability in the catchments. To ensure the representativeness of samples, 10–20 individuals of each species collected from each fish landing center was composited into a single sample as pooled.

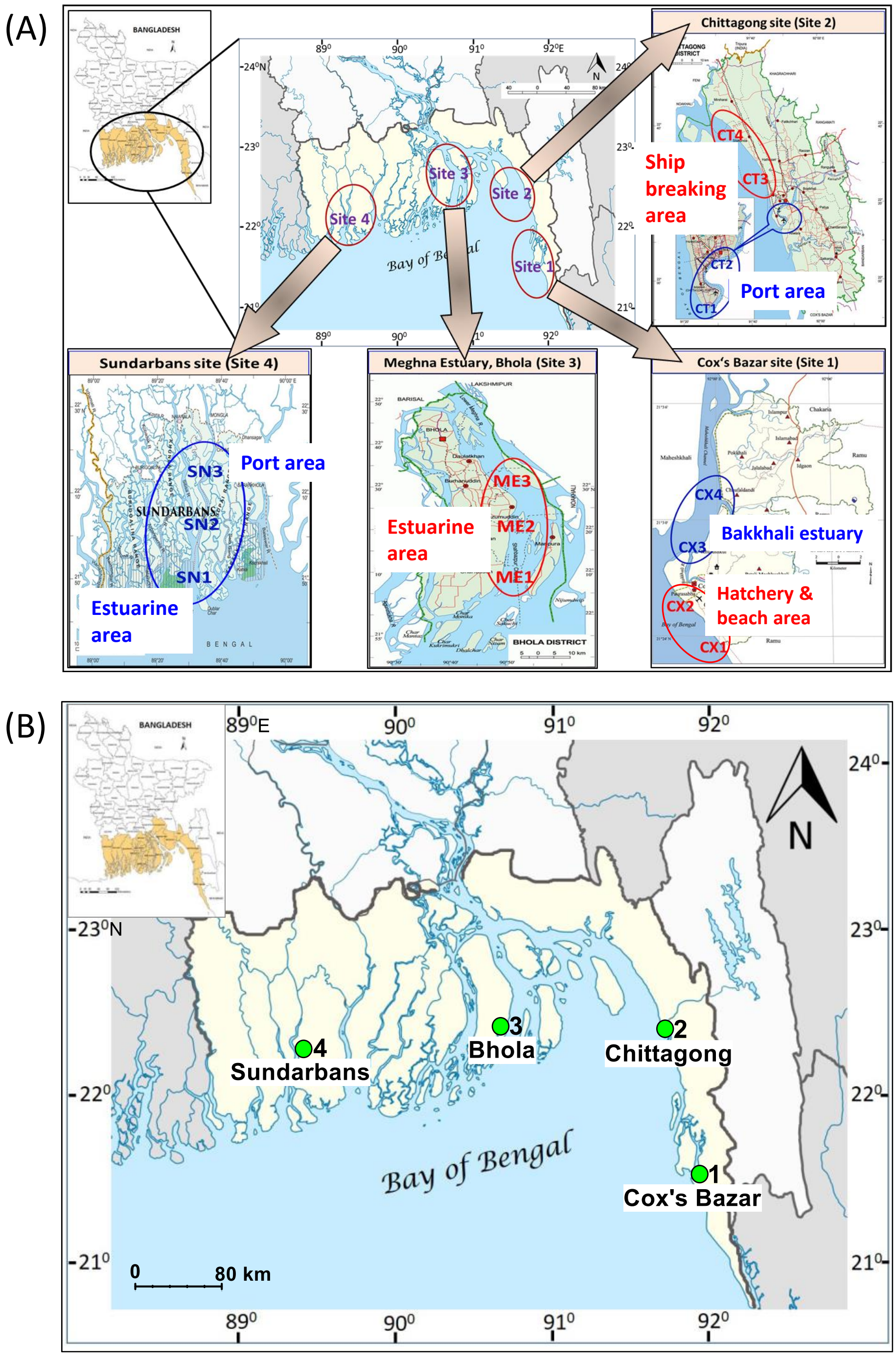

Figure 3-1. Maps showing (A). four sampling sites with 14 sampling locations in the coastal area of Bangladesh from where surface water and sediment samples were collected; (B). four main fish landing centers in the coastal areas of Bangladesh from where seafood samples were collected- (1): Fish Landing and Wholesale Fish Market, Cox's Bazar; (2): Chittagong fishing harbor, Chittagong; (3): Fish Landing and Wholesale Fish Market, Bhola; (4): Fish Processing and Marketing Centre, Sundarbans area in Mongla. Yellow colored area in the inset map represents the coastal area of Bangladesh.

Immediately after collection, seafood samples were wrapped in aluminium foil and kept in airtight insulating box filled with ice and transported to the laboratory of Department of Fisheries, University of Dhaka, Bangladesh. After transportation, finfish and shellfish samples were rinsed in deionized water to remove surface adherents. The length–weight data was recorded for each individual of every single species. Non-edible parts were removed with the help of a steam cleaned stainless steel knife. The edible portion of the collected seafood samples were homogenized, weighed, freeze-dried for about 48 h until the constant weight was attained. The species-specific information with their biometric data and pretreatment are shown in Table A-2 (Appendix A). All of the processed samples were brought to Yokohama National University, Japan, for chemical analysis with the permission of the Yokohama Plant Protection Station.

3.2.2 Chemicals and reagents

Native calibration PCBs standards (BP-MXP Native PCB solution/mixture of PCB 3, 8, 28, 52, 101, 118, 138, 153, 180, 194, 206, and 209) and isotopically labeled internal standards (MBP-MXP Mass-labelled PCB solution/mixture of ^{13}C-PCB 3, 8, 28, 52, 101, 118, 138, 153, 180, 194, 206, and 209) containing at least one congener for each homologue group of PCBs were obtained from Wellington Laboratories Inc. (Ontario, Canada). A complete set of all 209 PCB congeners (C-CSQ-SET Congener Calibration Set containing 209 native PCB congeners) were also 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 PCB and pesticide analysis grade and purchased from Wako Pure Chemical Ind. (Osaka, Japan). Milli-Q (>18.2 MΩ) water was used throughout the experiment and was generated by using an ultrapure water purification system (Millipore, Billerica, MA, USA). Filter membranes (0.45 µm, 47 mm i.d.) were obtained from ADVANTEC® (Tokyo, Japan).

3.2.3 Sample pretreatment

Surface 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 aliquots of dichloromethane, then twice by 10 mL aliquots of methanol and then 10 mL distilled water. One liter of filtered water samples was mixed well and trapped through the SPE tubes with a flow rate at 10 mL/min under vacuum. After the extraction, the cartridges were dried under vacuum for 10 min. The cartridges were then transported in dark, air-tight containers to the Laboratories of Yokohama National University in Japan for elution and analysis of PCBs. The cartridges were eluted with 100 mL dichloromethane:n-hexane (1:1) followed by spiking with 100 µL of 50 ng/mL of a mixed internal standards (IS), which included a mixture of ^{13}C-PCB containing at least one congener for each homologue group of PCBs (MBP-MXP Mass-labelled mono- to deca-PCB solution/mixture, Wellington Laboratories Inc., Canada). 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.

Freeze-dried sediment samples were homogenized with a silica mortar and pestle, then sieved through a 2-mm mesh sieve to remove debris and remove the coarse fraction (> 2 mm), which has low or negligible binding capacity for many contaminants (IAEA, 2003). After that, they were extracted by a Quick, Easy, Cheap, Effective, Rugged and Safe (QuEChERS) method validated by Salem et al. (2016) following some modifications. Briefly, an aliquot of 5 g of sediment was weighed into a polypropylene tube (50 mL capacity). Then, 5 mL of ultrapure water was added, the tube was manually shaken and spiked with 100 µL of 50 ng/mL of a mixture of ^{13}C-PCB containing at least one congener for each homologue group of PCBs (MBP-MXP Mass-labelled mono- to deca-PCB solution/mixture, Wellington Laboratories Inc., Canada) as an internal standard (IS) for quantification. Fifteen (15) mL of extraction solvent (hexane:acetone:dichloromethane ≈ 1:1:1, v/v/v) and two ceramic bars (Agilent p/n 5982–9313) were added to aid in sample extraction and the tube was shaken vigorously by hand for 5 min. Afterwards, the QuEChERS salts ((4 g of magnesium sulfate ($MgSO_4$), 1 g of sodium chloride (NaCl), 1 g of trisodium citrate dihydrate ($Na_3C_6H_5O_7$) and 0.5 g of disodium hydrogen citrate sesquihydrate ($C_6H_8Na_2O_8$); Agilent p/n 5982–6650) was then added and the tube was immediately shaken for 1 min to avoid agglomeration of salts followed by ultrasonic agitation for 20 min. Finally, the tube was centrifuged for 3 min at 3500 rpm and 10 mL of the supernatant were transferred to a QuEChERS dispersive solid phase extraction (d-SPE) clean-up tube (15 mL) containing 0.9 g of anhydrous magnesium sulfate, 0.15 g of primary secondary amine (PSA) and 0.15 g of C18EC (Agilent p/n 5982–5156). One ceramic bar (Agilent p/n 5982–9312) was added and the tube was shaken vigorously by hand for 1 min followed by centrifugation for 3 min at 3500 rpm. The extracts were then treated with activated copper for desulphurization and transferred into a glass test tube and then evaporated to near dryness under a gentle stream of high-purity nitrogen. The residue was finally re-dissolved in 1 mL n-hexane and stored at −20 °C until GC-MS/MS analysis.

Sample preparation for the extraction of PCBs from seafood involves a modified QuEChERS method previously validated by Ahmed et al. (2016) followed by use of EMR-Lipid (‘enhanced matrix removal of lipids’) and an additional ‘salting out/polishing’ step for cleanup validated by Han et al. (2016). Briefly, an aliquot of 5 g of homogenized sample was weighed into a polypropylene tube (50 mL capacity). Then, 5 mL of ultrapure water was added, the tube was manually shaken and spiked with 100 µL of 50 ng/mL of a mixture of ^{13}C-PCB containing at least one congener for each homologue group of PCBs (MBP-MXP Mass-labelled mono- to deca-PCB solution/mixture, Wellington Laboratories Inc., Canada) as an internal standard (IS) for quantification. Ten (10) mL of extraction solvent (hexane:acetone ≈ 1:1, v/v) and two ceramic bars (Agilent p/n 5982–9313) were added to aid in sample extraction and the tube was shaken vigorously by hand for 5 min. Afterwards, the QuEChERS salts (6 g of $MgSO_4$ and 1.5 g of sodium acetate; Agilent p/n 5982–5755) was added and the tube was immediately shaken for 1 min to avoid agglomeration of salts followed by ultrasonic agitation for 20 min. The tube was centrifuged for 5 min at 3500 rpm and 10 mL of the supernatant were transferred to a 15 mL centrifuge tube containing 1 g EMR–Lipid sorbent (Agilent p/n 5982–1010). One ceramic bar (Agilent p/n 5982–9312) was added and the tube was shaken vigorously by hand for 1 min followed by centrifugation for 3 min at 4000 rpm. The entire supernatant was decanted into a second 15 mL polishing tube containing 2 g mixture of 4:1 (w/w) anhydrous $MgSO_4$:NaCl (Agilent p/n 5982–0101), and vortexed immediately to disperse, followed by centrifugation at 4000 rpm for 3 min. The extracts were then transferred into a glass test tube and evaporated to near dryness under a gentle stream of high-purity nitrogen. The residue was finally re-dissolved in 1 mL n-hexane and kept at −20 °C until GC-MS/MS analysis.

3.2.4 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 and a computer with MassHunter software (version B.05.00412) for data acquisition and processing (Agilent Technologies, Palo Alto, CA). Chromatographic separation was achieved on an HT8-PCB column (60 m × 0.25 mm ID, 0.25 µm film thickness, Kanto Chemical Co., Inc., Japan) using Helium as a carrier gas at a flow rate of 1.0 mL/min. The GC oven temperature was initiated at 120 °C for 1 min, increased to 180 °C for 0 min at 20 °C min^{-1}, raised to 210 °C for 0 min at 2 °C min^{-1} and finally held at 310 °C for 3 min at 5 °C min^{-1}. 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 300 °C, 280 °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 'wide' resolution mode. Prior to analysis, MS/MS was auto-tuned with perfluorotributylamine. All of 209 PCB congeners were separated into 167 peaks, herein termed domains, denoting 135 individual and 32 coeluting congeners. The order of chromatograms and peak assignments were set according to Matsumura et al. (2002). GC–MS/MS conditions and/or parameters for the analysis of PCBs are shown in Table A-3 (Appendix A). 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 209 PCB congeners. The quantification of the PCBs 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 or volume (depending on media type). The detailed calculating equation has also been given in the report of Yang et al. (2011). Concentrations of single PCB congeners and total PCB (∑PCBs) are given in nano gram per liter (ng/L) for water, nano gram per gram dry weight (ng/g dw) for sediment and nano gram per gram wet weight (ng/g ww) for seafood samples. In addition, while calculating the concentration of PCB homologs and ∑PCBs the values of <LODs were assigned to zero.

3.2.5 Quality assurance and quality control (QA/QC)

The experiments were operated under strict quality control procedures. During the pretreatment procedure, all containers and equipment were pre-cleaned with methanol followed by acetone. The limit of detection (LOD) for each analyte was defined as the smallest mass of compound resulting in an S/N ratio that was equal to or greater than 3. The LODs were in the range of 0.021 to 0.132 ng/L in water, 0.002 to 0.014 ng/g dw in sediment, and 0.006 to 0.06 ng/g ww in seafood. Instrumental blanks (solvent without internal standard) were analyzed every five to seven samples to monitor instrumental background. Instrumental blanks gave signal-to-noise (S/N) values of less than three. The procedural blanks (method blanks) prepared with Milli-Q water were spiked internal standards. Procedural blanks were analyzed with every batch of samples. Instrumental blanks and procedural blanks were below the LODs. For each matrix, analyte recovery was determined by using spiked samples to validate the accuracy of the methods. Matrix spike recovery (n = 3) was determined by spiking the target compounds into the water (10 ng/L), sediment (10 ng/g dw) and seafood (10 ng/g ww) samples, followed by extraction and analysis as described in the previous section. The mean recoveries of PCBs spiked into the water, sediment and seafood samples were 71%−118%, 62%−116% and 57%−113%, respectively. The detailed QA/QC data are given in Table A-4 (Appendix A).

3.2.6 Data analysis

Statistical analyses were performed with IBM SPSS software (Version 23.0, IBM Corp., NY, USA). The significance level was set at p = 0.05. Before analyzing, concentration values lower than the LODs were set to LOD/2 (Succop et al., 2004). A statistical distribution test called *P–P* plots was carried out to test for normality. Descriptive statistics (range: minimum-maximum, mean, and median) was calculated by using the Microsoft® Excel® 2016 MSO Windows program. A one-way ANOVA was performed to determine the significant differences between the concentrations of PCBs detected in the coastal area of Bangladesh and to examine seasonal variations. The spatial distributions of PCBs were analyzed using MapViewer™ software (Version 8, Golden Software Inc., CO, USA). PCB nomenclature follows the shorthand numbering notation (PCB1–209) of Ballschmiter and Zell (1980). Total PCB TEQ (Toxic Equivalents) was derived using the WHO 2005 TEFs (Toxic Equivalency Factor) (Van den Berg et al., 2006).

3.3 Results and discussion

3.3.1 PCBs in surface water

3.3.1.1 Concentrations of PCBs in surface water and global comparison

Water samples from all 14 sites contained detectable concentrations of PCBs, indicating that PCBs are widespread in the Bangladeshi coastal area. The data of PCBs in water samples are shown in **Table 3-1** and **Figure 3-2**, while the concentrations of individual congeners are presented in Table A-5 and A-6 (Appendix A). A total of 136 domains comprising 109 single congeners, 19 double coeluting isomers, 6 triple coeluting isomers, and 2 quadruple coeluting isomers were resolved in water samples regardless sites and seasons. However, the detected PCBs were only the congeners with 2 to 8 Cl atoms. Congeners with 1 Cl might evaporate in the pretreatment procedure, and the congeners with 9 and 10 Cl atoms were under their detection limits. The total concentrations of the PCBs (∑PCBs) in the water phase ranged from 32.17 to 160.7 ng/L in winter, and from 46.45 to 199.4 ng/L in summer. The distribution of samples by class of ∑PCBs levels were: 36% (>100 ng/L), 43% (50–100 ng/L), 21% (<50 ng/L). The ∑PCBs were positively correlated with the number of detected domains per sample (Pearson correlation, $r = 0.82$, $p < 0.05$). However, the ∑PCBs variation might be dependent on some dominant congeners. The sum of 12 dioxin-like PCB (∑DL-PCBs) and 6 ICES (International Council for the Exploration of the Sea) indicator/marker PCB congeners (∑iPCBs) ranged from 2.79 to 15.13 and 4.22 to 16.74 ng/L in winter, and from 5.27 to 25.79 ng/L and 5.96 to 30.84 ng/L in summer, respectively. The ∑DL-PCBs and ∑iPCBs contributed together only 20–28% to the total PCB concentrations, elucidating that monitoring only of these congeners would be an underestimation to the assessment of PCB contamination in the Bangladeshi coastal waters. However, correlations between ∑PCB and ∑DL-PCBs, and ∑iPCBs were strong ($r = 0.91$ and 0.94, respectively) and significant ($p < 0.05$). It means that the study area might be impacted by many other congeners along with these frequently monitored PCBs.

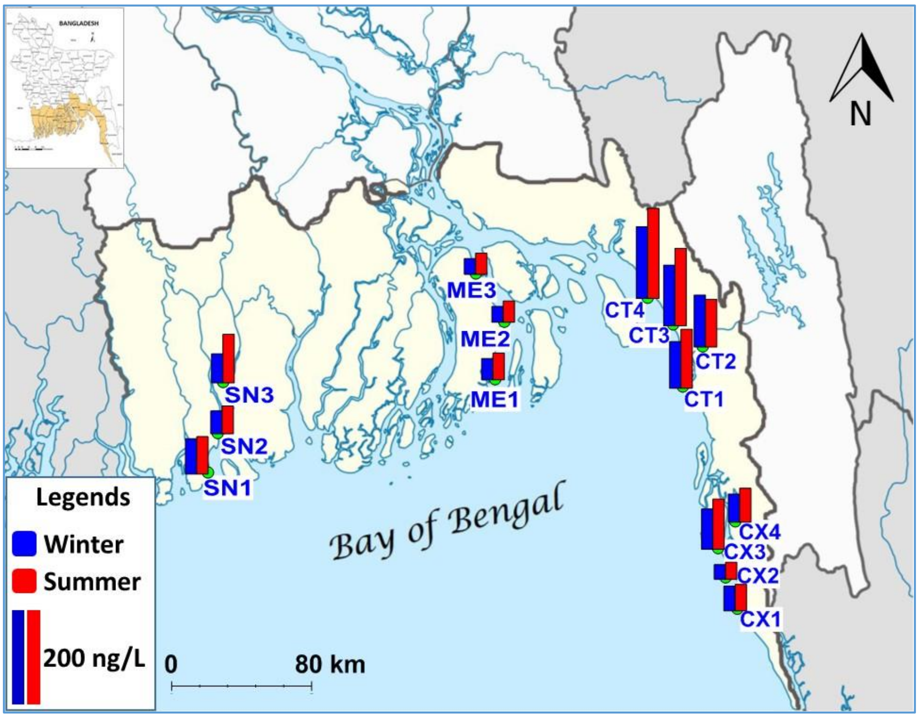

Figure 3-2. Distribution of total PCBs in surface water of the coastal area of Bangladesh. Samples were collected in winter and summer of 2015. Colored area in the inset map represents the coastal area of Bangladesh.

To understand the magnitude of PCB contamination, the results obtained in the present study were compared with those of previous studies conducted in other estuarine and coastal areas around the world in **Table 3-2**. In fact, the scientific literature of PCB levels considering a complete set of 209 congeners in the surface water is still scarce. In general, the levels of water phase PCBs in this study were comparable to or higher than those in Liaodong Bay (China), Okinawa Island (Japan), Tokyo Bay (Japan), Houston Ship Channel (USA), North Western Mediterranean, Tiber River and Estuary (Italy), and Belgian coastal harbors, evidently lower than in Daya Bay and Pearl River Estuary, China (**Table 3-2**). Interestingly, concentration of total PCBs in this study (32.17–199.4 ng/L) was 15–45 times higher than the concentration found in the surface water (1.93–4.46 ng/L) from the Bay of Bengal of Indian coast (Rajendan et al., 2005). This huge difference in PCB concentrations among the neighboring areas implies that some important and/or significant potential sources of PCBs exist in the Bangladeshi coast of the Bay of Bengal.

Table 3-1. Concentration (ng/L) of PCB homologs and total PCBs in the surface water samples collected from the coastal area of Bangladesh in winter and summer, 2015.

Season	Site ID	Concentration (ng/L)												
		Mono-CB	Di-CB	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB	Octa-CB	Nona-CB	Deca-CB	∑DL-PCBs[a]	∑iPCBs[b]	∑PCBs[c]
Winter	CX1	<LOD	3.34	11.36	15.63	13.51	9.32	7.17	2.30	<LOD	<LOD	5.07	6.76	62.62
	CX2	<LOD	2.71	6.85	10.41	8.87	6.28	5.18	1.25	<LOD	<LOD	6.19	5.58	41.55
	CX3	<LOD	4.64	16.89	26.59	21.53	14.29	8.73	2.48	<LOD	<LOD	7.73	11.72	95.16
	CX4	<LOD	3.18	14.37	22.30	15.48	6.98	5.84	1.73	<LOD	<LOD	6.40	7.38	69.87
	CT1	<LOD	7.61	19.88	29.93	21.44	19.48	8.33	2.11	<LOD	<LOD	6.89	15.81	108.8
	CT2	<LOD	5.00	20.41	31.33	26.89	21.08	11.24	3.45	<LOD	<LOD	9.59	14.73	119.4
	CT3	<LOD	5.59	18.14	41.76	34.60	21.73	13.24	2.60	<LOD	<LOD	11.24	16.74	137.7
	CT4	<LOD	10.38	20.14	41.92	49.15	23.64	11.86	3.65	<LOD	<LOD	15.13	14.15	160.7
	ME1	<LOD	4.20	11.22	12.91	14.64	5.27	5.46	1.67	<LOD	<LOD	4.77	5.38	55.37
	ME2	<LOD	2.08	6.81	10.61	11.75	6.12	5.63	0.92	<LOD	<LOD	5.60	5.89	43.93
	ME3	<LOD	2.86	8.58	7.31	5.60	4.17	2.75	0.90	<LOD	<LOD	2.79	4.22	32.17
	SN1	<LOD	5.66	16.59	27.62	14.99	9.65	8.46	1.60	<LOD	<LOD	4.88	12.08	84.57
	SN2	<LOD	4.75	12.82	16.74	10.67	6.22	5.73	1.70	<LOD	<LOD	5.57	7.39	58.63
	SN3	<LOD	5.74	16.50	21.57	12.13	8.24	5.67	1.87	<LOD	<LOD	4.42	9.16	71.72
Summer	CX1	<LOD	5.14	12.16	18.63	13.49	8.74	6.96	1.13	<LOD	<LOD	7.32	8.06	66.26
	CX2	<LOD	3.30	7.41	13.20	11.26	5.78	4.58	0.91	<LOD	<LOD	5.60	5.96	46.45
	CX3	<LOD	8.55	19.33	30.18	32.93	12.04	10.61	2.34	<LOD	<LOD	10.04	14.95	116.0
	CX4	<LOD	4.55	15.48	21.80	19.35	11.60	7.99	1.04	<LOD	<LOD	9.68	12.73	81.80
	CT1	<LOD	12.43	28.46	35.34	29.98	14.16	11.61	2.20	<LOD	<LOD	12.93	18.52	134.2
	CT2	<LOD	8.07	20.94	35.44	20.60	13.06	11.35	0.92	<LOD	<LOD	10.24	12.57	110.4
	CT3	<LOD	8.54	31.69	39.18	45.82	23.99	19.41	4.11	<LOD	<LOD	21.52	24.99	172.8
	CT4	<LOD	11.00	27.65	48.50	54.75	29.06	21.51	6.89	<LOD	<LOD	25.79	30.84	199.4
	ME1	<LOD	4.15	13.28	19.68	11.69	9.49	6.81	1.78	<LOD	<LOD	6.51	8.61	66.88
	ME2	<LOD	3.19	14.43	14.23	9.11	7.06	6.31	0.91	<LOD	<LOD	5.50	8.87	55.24
	ME3	<LOD	2.17	11.76	15.48	8.97	6.14	4.95	0.30	<LOD	<LOD	5.27	7.76	49.77
	SN1	<LOD	5.69	17.71	19.27	19.18	13.26	11.83	2.15	<LOD	<LOD	8.70	14.92	89.10
	SN2	<LOD	4.12	16.05	19.32	12.82	8.99	6.91	0.92	<LOD	<LOD	9.56	10.76	69.12
	SN3	<LOD	9.53	21.22	28.57	22.42	14.02	13.82	2.85	<LOD	<LOD	10.48	13.15	112.4

[a] Sum of 12 dioxin-like PCBs (4 non-ortho substituted PCBs: PCB77, 81, 126, 169; 8 mono-ortho substituted PCBs: PCB105, 114, 118, 123, 156, 157, 167, 189);

[b] Sum of 6 ICES indicator or marker PCBs (ICES6-PCBs: PCB28, 52, 101, 138, 153, and 180) (European Commission, 2011);

[c] Sum of 209 PCBs;

Table 3-2. Comparison of total PCBs (ng/L) in surface water between this study and other studies worldwide.

Location	Sampling year	N^a	∑PCBs	References
Daya Bay, China	1999	12	91.1–1360	Zhou et al., 2001
Pearl River Estuary, China	2000	21	33.4–1060	Zhang et al., 2002
Liaodong Bay, China	2007	41	5.51–40.28	Men et al., 2014
Bay of Bengal, India	1998	–[b]	1.93–4.46	Rajendran et al., 2005
Okinawa Island, Japan	2002	–[c]	1.59–2.48	Sheikh et al., 2007
Tokyo Bay, Japan	2006	209	0.04–0.64	Kobayashi et al., 2010
Houston Ship Channel, USA	2003	209	0.49–12.5	Howell et al., 2008
North Western Mediterranean	2001	41	2.42–70.5	Garcia-Flor et al., 2005
Tiber River and Estuary, Italy	2014–15	32	0.54–74.75	Montuori et al., 2016
Belgian coastal harbors	2007–10	14	0.03–3.1	Monteyne et al., 2013
Coastal area of Bangladesh	2015	209	32.17−160.7 (W)[d] 46.45−199.4 (S)[d]	This study

[a] Number of PCB congeners;
[b] Sum of di- to deca-CB homologs;
[c] Sum of di- to nona-CB homologs;
[d] W and S represent winter and summer, respectively;

3.3.1.2 Seasonal and spatial distribution of water-phase PCBs

The seasonal variations and spatial distributions of PCBs in the surface water are presented in **Figure 3-2**. Small variations were observed in the levels of water phase PCB between the two seasons (winter vs. summer), although the difference was not statistically significant ($p > 0.05$). Thirteen out of the 14 water samples investigated showed relatively higher concentration of ∑PCBs in summer season than in winter. Precipitation and pollution sources are considered to be the main factors causing fluctuations in water quality. The variations of PCBs in the two seasons might be mainly caused by the difference of precipitation in these two periods. In summer (wet season), PCBs previously buried in the surface soil of heavily contaminated sites and accumulated in dry weather were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. While in winter (dry season), with less precipitation and surface runoff, the contaminants might selectively be accumulated in surface soil instead of being washed away. PCB levels in water phase might also be attributed to the water surface temperature, air/gas exchange rate and desorbing rate of PCBs from sediment to water which also varied seasonally. PCB concentrations in water could be affected by many other factors, such as water flow, water quality properties, human activities like fishing and boating, and PCB usage and discharge in aquatic systems from seasonally operated industries. However, a different trend was observed at CT2 where the concentration of ∑PCBs was higher in winter (119.4 ng/L) than in summer (110.4 ng/L). Although the reason for the ∑PCBs concentration difference between the two seasons in this location (CT2) is not clear, it may be an effect of intense dredging particularly in winter to facilitate shipping activities. A portion of sedimentary PCBs might be released into the water phase during the dredging operation due to substantial mixing of sediment and water. In general, the mean concentration of total PCBs in summer (97.83 ng/L) was slightly higher than in winter (81.58 ng/L).

Figure 3-2 shows the spatial distribution of PCBs in surface water samples. Levels of PCBs in surface water differed significantly between the four coastal regions ($p < 0.05$), indicating the pollution of PCBs mainly influenced by the local/regional source inputs in the study areas. Concentrations of ∑PCBs were higher in the coastal waters at sampling locations CT1–CT4 (average of 131.6 and 154.2 ng/L in winter and summer, respectively), SN1–SN3 (average of 71.64 and 90.21 ng/L in winter and summer, respectively) and CX1–CX4 (average of 67.3 and 77.62 ng/L in winter and summer, respectively) in comparison to sampling locations ME1–ME3 (average of 43.82 and 57.3 ng/L in winter and summer, respectively), which indicates that the industrialized regions (Chittagong, Cox's Bazar and Sundarbans) in this area are potential sources of PCBs and that economic development seems to be associated with the amount of PCB emissions. Hong et al. (2005) and Wurl and Obbard (2005) found higher PCB concentrations near industrialized areas, and thus it is not at all surprising to find that the more highly industrialized regions in the Bangladeshi coastal area contain higher PCB concentrations. Additionally, the lower concentrations of PCBs found in water taken from the Meghna Estuary (ME1−ME3) could result from the mixing or dilution effects of huge volumes of inland freshwater (2.73–4.17 trillion m^3 per annum) with seawater which was assumed to be less contaminated by PCBs. In particular, water samples at location CT4 showed the highest levels of PCBs for both seasons (160.7 and 199.4 ng/L in winter and summer, respectively) followed by CT3 (137.7 and 172.8 ng/L in winter and summer, respectively). Other two sites from Chittagong area, CT1 and CT2 also exhibited elevated PCB levels compared to other areas, indicating the existence of point source(s) in the area of Chittagong. CT3 and CT4 are located very close to Chittagong ship breaking yard. Ship breaking activities along the coast of Chittagong may contribute to the PCB pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PCBs (Neşer et al., 2012; Siddiquee et al., 2012). Large amounts of PCB were used (especially in the electrical cable sheathing, pains and hydraulic fluids) in the ships built before banning of PCB. It was estimated that each old ship contained 250 kg of PCBs, inclusive of transformer oil (DoE, 2007). It is thus, due to lack of proper management and monitoring, a huge amount of PCB might be released into the environment during the dismantling or demolition processes of old ships. There are approximately 100 e-waste recycling shops located very close to site CT4, generating approximately 2.7 million metric tons of toxic e-waste each year, the majority of which comes from the ship breaking sector. However, the waste generated from the e-waste recycling shops is discharged directly into the adjacent coastal water, which might be one of the most significant PCB pollution sources in the study area. Nøst et al. (2015) found an elevated PCB levels in air at sites near the ship breaking activities in Chittagong which might be re-deposited from air to water through wet deposition, air/water exchange as well. Moreover, CT1 and CT2 are within the Chittagong port area which is the largest seaport in Bangladesh. These sites are predominantly influenced by the port activities. It was previously reported that the use of paints and grease repellents for ship and dock protection could contribute to the relatively high levels of PCBs in port seawaters. Shipping and boat maintenance in the ports may also be responsible for the release of PCBs into the surrounding waters (Rajendan et al., 2005). Moreover, there are numerous multipurpose industrial establishments along the coast of Chittagong producing paper and pulp (e.g., Karnaphuli paper mills), cement clinkers, fertilizers (e.g., Karnaphuli fertilizer), steel products, rubber and plastic, petroleum products (e.g., Super petrochemical), beverages, sugar, pharmaceuticals, tobacco, jute, textiles, printing and dyeing, fish products, tannery products, paint, rechargeable batteries, jewelry, plating, automobile engines, and electronics. The discharge of untreated or semi-treated effluents from these industries may pose a significant contribution to the PCB contamination into the nearby environment (Yang et al., 2011).

Within Cox's Bazar, water samples from CX3 (Bakkhali Estuary) showed elevated concentration of PCB (95.16 and 116.0 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 PCB contamination in the environment (Rajendan et al., 2005; Binelli et al., 2009). In addition, the main municipal garbage dump (mostly consumer products including obsolete electronics) of Cox's Bazar city is also located near CX3. Therefore, a significant amount of PCBs might be emitted from this dumping site into the adjacent water body.

The levels of PCB in water from Sundarbans sites exhibited an interesting trend: concentration of PCB first decreased from upper estuary (SN3) to mid-estuary (SN2) and then increased further towards lower estuary (SN1) or mouth of the estuary to the Bay of Bengal. The highest concentration was found at SN3 (112.4 ng/L) which is located vary close to Mongla port. There is high density of shipping activities in and around the port area and hence high PCB levels in water are related to potential discharges from the ships. Higher PCB levels might be attributed to the huge discharge of untreated or partially treated effluents from numerous multipurpose industries such as cement, paint, paper, printing and dyeing, plastics, leather, etc. Zhou et al. (2001) reported that the wastewater discharged from factories (e.g. leather, paper, electronics, plastics, etc.) was the major sources of PCB inputs into the Daya Bay, China. In addition, intense dredging operations in this area along with agricultural runoffs, dumping of industrial wastes, surface runoff and atmospheric depositions further aggravate the PCB pollution. The decrease in PCB concentration in water from mid-estuary (SN2) might be attributed to the dilution effects from mixing of inland fresh water with the salty seawater (An et al., 2009; Men et al., 2014). Moreover, the coastal area of Bangladesh is influenced by daily tidal action (two high and two low tides at the same day). Therefore, PCBs might be carried downward by water during low tides and then retain in the mouth of the estuary, finally can be carried offshore into the coastal area and beyond. In addition, many ships and fishing boats travel in this area, which might be associated with the increase of PCBs as well. The levels of PCB in water taken from the Meghna estuary also showed a similar trend, where concentrations increased downward 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 PCB in water from this site suggest that there are non-point sources of PCBs (e.g., surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc.) in the coastal area of Bangladesh. Furthermore, the Meghna Estuary sites receive major volumes of water from the Ganges River of India through the Yamuna River flowing down to the Meghna Estuary via the Mehgna River. Previous studies reported that the surface water of these rivers was contaminated by PCBs (Chakraborty et al., 2014; Kumar et al., 2013). The authors also noted that PCB concentrations gradually declined due to dilution as the water mass increased downstream of the rivers. Thus, the coastal areas of Bangladesh to some extent might receive

PCBs through the water flow of these upstream rivers, which flow into the Meghna Estuary sites. Generally speaking, the concentrations of ∑PCBs in surface water at the Chittagong sites (CT1–CT4), Cox's Bazar (CX1–CX4) and Sundarbans sites (SN1–SN3) were higher than those in the Meghna Estuary (ME1–ME3) and because of greater development, including industrialization and urbanization activities.

3.3.1.3 Homolog composition

The PCB congener and homolog profiles in environmental matrices can often provide valuable information on the environmental source, transport and fate processes of PCBs. PCBs are grouped into homologs, herein termed mono–deca-CB, each comprising congeners with equal numbers but different positions of Cl atoms (isomers). **Figure 3-3** shows the concentration percentages of PCB homologs in the surface water samples, which generally exhibited the following order: medium (4–6 Cl, 53–71%) > light (1–3 Cl, 17–36%) > heavy (7–10 Cl, 10–16%). Specifically, the homolog distribution was dominated by tetra-CB (23–33% and 22–32% of total PCBs in winter and summer, respectively), followed by penta-CB (17–31% and 16–28% of total PCBs in winter and summer, respectively) and tri-CB (13–27% and 14–26% of total PCBs in winter and summer, respectively). However, the penta- and tri-CB homologs contributed almost equally to the total PCB distribution in the surface water of the Bangladeshi coastal area. In seawater from the Bay of Bengal in the Indian coast, proportion of lighter homologs (di-, tri- and tetra-CB) were found as 74.4%–85.6% higher than the heavier homologs (penta-, hexa- and hepta-CB) with 25.6%–14.4%, respectively (Rajendan et al., 2005), and the reason was explained as the low chlorinated biphenyls are, in general, more water-soluble than the high chlorinated biphenyls. On the contrary, Zhou et al. (2001) reported predominance of high chlorinated congeners like penta-, hexa- and octachlorobiphenyls, accounting for 94% of total PCB in water from the Daya Bay, China. The PCB distribution profiles in Daya Bay indicated a number of sources contributing to total contaminant burden in the bay, including surface runoffs, wastewater discharges, sewage outfalls, and shipping activities (Zhou et al., 2001). Men et al. (2004) reported that paint from ships and fishing vessels is responsible for the dominance of penta-CB residues in the estuarine water of Liaodong Bay, Bohai Sea (China). Our results were consistent with the PCB distribution in water from Tiber River and Estuary, where tetra- and penta-CBs were found as abundant homologs, accounting for 39% and 29% of ∑PCBs, respectively, followed by the less chlorinated CBs (di- + tri-CBs) accounted for 18% of total PCBs (Montuori et al., 2016). The authors (Montuori et al., 2016) assumed that the inputs into the estuary by atmospheric deposition, surface and inland river runoff might be the reasons for the less contribution of lighter PCBs. In fact, the lighter congeners exhibit higher mobility in the atmosphere and aquatic environment (Gao et al., 2013; Men et al., 2014). Also, if there were no nearby sources of PCBs, the congeners would be evenly distributed over all levels of chlorination (Bremle and Larsson, 1997), where the distribution may predominantly be influenced by the atmospheric deposition and oceanic transportation. Therefore, in the present study, the PCB homologs distribution may suggest a low impact from atmospheric inputs to the water phase PCB burden in the Bangladeshi coastal area, and may also suggest the presence of potential point sources of PCBs.

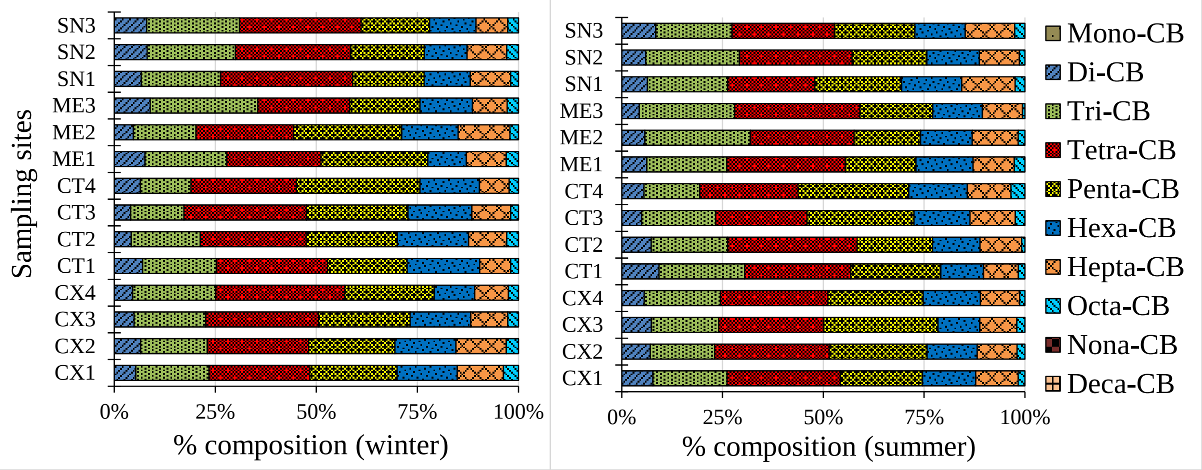

Figure 3-3. Relative contribution of PCB homologs (% composition) to the total PCB in the surface water of the Bangladeshi coastal area in winter and summer, 2015.

3.3.1.4 Congener profiles and source characterization

We identified the dominant PCBs in water phase both by occurrence and abundance by combining the detection rate and mean relative concentration of each congener/co-elute across samples and ranked them averagely in decreasing order. The most abundant PCB congeners were PCB43+49, 44, 70, 120+110, 52, 20+33, 149+139, 138, 153, 28, 101, 18, 31, 118, 40, 66, 17, 105, 180, and 170, comprising up to 47–76% of ∑PCBs by sites, and highly correlated with ∑PCB ($r = 0.976$–0.979; $p < 0.05$), well representing the ∑PCB in the surface water of the Bangladeshi coastal area.

Apparently, the dominant congeners in the present water samples were also key constituents of commercial PCB mixtures, e.g. PCBs 18, 28, 31, and 20+33 in Aroclors 1242;

52, 44, 43+49 (49), 44, 40, 66, and 70 in Aroclor 1248; 101, 120+110 (110), 118, 105, and 138 in Aroclor 1254; and 149+139, 153, 180, and 170 in Aroclor 1260 (Jones, 1988; Ishikawa et al., 2007). It indicates that the environmental PCB burdens in the Bangladeshi coastal waters might be originated from more than one PCB Aroclors and the most likely mixtures were Aroclors 1248, 1254 and 1260. PCBs were never produced, but only imported for use in Bangladesh. A general assessment of PCB use together with information on the available import records in Bangladesh is given elsewhere (DoE, 2007; ESDO, 2005a,b; 2010). Presently, no specific information exists regarding the identity of PCB mixtures used in Bangladesh historically. This lack of information results in difficulties during the selection of PCB mixtures for consideration in sources analyses. According to the available reports (DoE, 2007; ESDO, 2005a,b; 2010), a significant amount of transformer oils with different brand names (e.g. askarel, sovtol-10, and many more) was imported in Bangladesh. Transformer askarels contained either 70% Aroclor 1254 or 60% Aroclor 1260 (Erickson, 1997) and sovtol-10 is a mixture of PCB congeners of 4 and 5 Cl atoms (Zorrilla et al., 2012). Moreover, hundreds of PCB-containing equipment (e.g. transformers, capacitors, etc.) were also imported from different PCB producing countries such as Japan, Germany, France, England, USA, China (DoE, 2007) where the commercial PCB mixtures were known by different trade names. In this report, we used ‘Aroclor’ to refer PCB mixtures/formulations as a general practice in PCB analyses. Since the 1980s, national environmental regulations have existed in Bangladesh, such as the ban of PCBs (ESDO, 2005a,b; 2010; Nøst et al., 2015). However, PCB compounds are still in use in Bangladesh, mostly in closed systems as dielectrics in transformers and capacitors being operated in the electrical generating sector (DoE, 2007). Moreover, the outdated PCB-containing equipment, (old transformers, capacitors, etc.) along with non-recycled e-waste mostly originated from ship breaking industries is simply deposited in landfills particularly in coastal areas. Therefore, leakage from the PCB-containing in-service equipment and leachates from landfill sites might be considered as potential sources of significant PCB inputs into the environment.

Interestingly, PCB11, a non-aroclor congener, was observed frequently in water samples from almost all sites (86% in winter and 100% in summer samples), although contributing only 0.4–2% of ∑PCBs. PCB 11 is a non-legacy signature of PCB contamination, being absent in commercial PCB formulations, but largely originates as a byproduct of paint and pigment production and a residue in printed products (Hu and Hornbuckle, 2010; Hu et al., 2014; Vorkamp, 2016). Therefore, the effluents from industries that are using and producing pigments and dyes, printing of consumer goods (e.g. magazines, books, maps, posters, brochures, napkins, garments, etc.) should be considered as PCB source in the study area.

Among the 12 DL-PCBs studied, PCB77, 81, 105, 114, 118, 123, 156, 157, 167, and 189 were observed in all of the samples for both seasons, whereas PCB126 and 169 were found in 36–50% and 21–64% of samples, respectively. Thus, emission from industrial applications might be the major sources of DL-PCBs in the study area, whereas combustion processes apparently contributed to some extent what confirms the presence of PCB77 and 126 (marker PCBs for combustion processes) representing a contribution of 0.29–3.03% of ∑PCBs. According to Chi et al. (2007), non-ortho-PCBs that are characteristic of coal combustion and industrial waste incineration, do not originate solely from commercial PCB formulations.

Moreover, the observed finding of congener-specific distribution towards moderately to highly chlorinated compounds is in well agreement with previously published data indicating the preferential retention of these less volatile and more lipophilic compounds in the estuarine or marine environment (de Voogt et al., 1990). Differences in congener composition in the aquatic systems may also be attributed to a decline in the proportion of less chlorinated PCBs that are more susceptible to losses through volatilization, sedimentation, and possibly microbial degradation (Brown et al., 1987). Moderately and highly chlorinated PCBs may therefore remain persistent in the aquatic environment because they are less volatile and more soluble in lipids, adsorb more readily to sediments, and are more resistant to microbial degradation (Tyler and Millward, 1996).

3.3.2 PCBs in surface sediment

3.3.2.1 Concentrations of PCBs and their relationship with sediment properties

The estuary or coastal waterbody is a sink for variety of chemicals (Loizeau et al., 2001; Ma et al., 2001), especially for some POPs such as PCBs with high hydrophobic and lipophilic characteristics. Sediment medium acts as a final reservoir for these chemicals (Wang et al., 2008; Sahu et al., 2009, Gao et al., 2013). Therefore, the analysis of sediment samples constitutes a very important part for assessing the environment quality of a region and anthropogenic activities on aquatic system. All sediment samples analyzed in the present study contained detectable levels of PCBs, demonstrating ubiquitous contamination by these compounds in the aquatic environment of the Bangladeshi coastal area. The data of PCBs in sediment samples are shown in **Figure 3-4** and **Table 3-3**, while the concentrations of individual congeners are presented in Table A-7 and A-8 (Appendix A). A total of 135 domains comprising 110 single congeners, 17 double coeluting isomers, 6 triple coeluting isomers, and 2 quadruple coeluting isomers were resolved in sediment samples regardless sites and seasons. However, the detected PCBs were only the congeners with 2 to 8 Cl atoms. Congeners with 1 Cl might evaporate in the pretreatment procedure, and the congeners with 9 and 10 Cl atoms were under their detection limits. The concentrations of total PCBs (∑PCBs) in the sediment samples varied from 5.27 to 92.21 ng/g dw in winter, and from 4.61 to 105.3 ng/g dw in summer. The distribution of samples by class of ∑PCBs levels were: 71% (<25 ng/g dw), 14% (25–50 ng/g dw), and 14% (>50 ng/g dw). The ∑PCBs were positively correlated with the number of detected domains per sample ($r = 0.73$, $p < 0.05$).

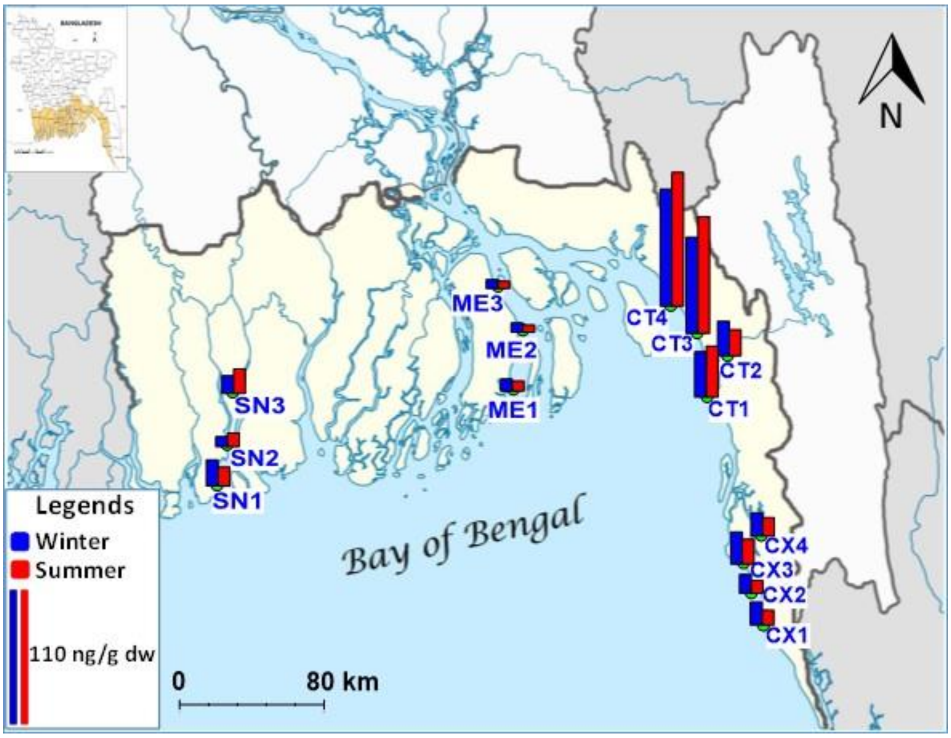

Figure 3-4. Distribution of total PCBs in surface sediment of the coastal area of Bangladesh collected in winter and summer, 2015. Colored area in the inset map represents the coastal area of Bangladesh.

The sum of 12 dioxin-like PCB (∑DL-PCBs) and 6 ICES indicator/marker PCB congeners (∑iPCBs) were in the range of 0.62–10.65 and 0.61–13.01 ng/g dw in winter, and

0.79–14.83 and 0.92–20.67 ng/g dw in summer, respectively. The ∑DL-PCBs and ∑iPCBs only account for 20–45% of the ∑PCB concentrations. Therefore, the DL-PCBs and iPCBs could not comprehensively reflect the contamination level of PCB in sediment, elucidating that the degree of PCB contamination in the Bangladeshi coastal area might be impacted by some other congeners, assumed to be dominant in the present case. However, correlations between ∑PCB and ∑DL-PCBs, and ∑iPCBs were strong (r = 0.98 and 0.97, respectively) and significant ($p < 0.05$).

The physicochemical properties of sediment (e.g. grain size/texture, organic matter) have been recognized as a major influential factor in determining the distribution and transport of hydrophobic organic compounds (OC) like PCBs in the aquatic environments (Binelliet al., 2009; Hong et al., 2012; Gao et al., 2013; Men et al., 2014; Ali et al., 2014). Among them total organic carbon (TOC) is crucial for their sorption, a key process in trapping and transporting PCBs in sediments (Hung et al., 2010; Hong et al., 2012; Ali et al., 2014). Within total organic moieties, black carbon (BC) with the highest sorption capacity has been shown to large extent affect the distribution of OCs in sediments (Accardi-Dey and Gschwend, 2002). PCBs may be extensively adsorbed onto condensed carbonaceous geo-sorbents such as BC owing to its condense and aromatic structure (Semple et al., 2013). It has been reported that adsorption of various POPs including PCBs by BC can be approximately two orders of magnitude higher than by sedimentary TOC (Hung et al., 2010; Ali et al., 2014 and references therein). Here, we performed a Pearson correlation analysis to investigate the influence of sediment properties on the distribution of PCBs in the study area.

The concentrations of sedimentary TOC (%) and BC (%) were recorded as 0.06–1.28 and 0.06–0.53 in winter and 0.15–0.69 and 0.06–0.28 in summer, respectively (Table A-9, Appendix A). No significant variation was observed in sediment properties between the two seasons (t-test, $p > 0.05$). However, the correlation results revealed a significant positive association between the ∑PCB and BC (winter: $r = 0.68$, $p < 0.001$; summer: $r = 0.71$, $p < 0.001$), whereas the correlations between ∑PCB and TOC were not significant in either seasons (winter: $r = 0.39$, $p > 0.05$; summer: $r = 0.41$, $p > 0.05$). Interestingly, irrespective to seasons, all homolog groups of PCBs (di- to octa-CBs) showed significant positive correlations with BC (r = 0.55–0.67; $p < 0.001$), while in case of TOC, the correlations were positive but not significant (r = 0.27–0.39; $p > 0.05$), suggesting BC plays a major role in the distribution of PCBs. Our findings were in agreement with others reported the similar observations elsewhere (Hung et al., 2006; Ali et al., 2014).

Table 3-3. Concentration (ng/g dw) of PCB homologs and total PCBs in the surface sediment samples collected from the coastal area of Bangladesh in winter and summer, 2015.

Season	Site ID	Concentration (ng/g dw)												
		Mono-CB	Di-CB	Tri-CB	Tetra-CB	Penta-CB	Hexa-CB	Hepta-CB	Octa-CB	Nona-CB	Deca-CB	∑DL-PCBs[a]	∑iPCBs[b]	∑PCBs[c]
Winter	CX1	<LOD	0.31	2.39	3.52	5.37	3.86	2.60	0.42	<LOD	<LOD	2.07	2.83	18.47
	CX2	<LOD	0.25	2.45	3.06	3.58	2.75	2.48	0.50	<LOD	<LOD	1.40	2.61	15.06
	CX3	<LOD	1.66	4.42	4.78	6.44	4.53	2.86	0.63	<LOD	<LOD	2.73	3.98	25.33
	CX4	<LOD	1.56	3.70	4.40	3.90	2.29	2.55	0.32	<LOD	<LOD	1.57	3.06	18.71
	CT1	<LOD	0.55	3.20	9.90	9.59	6.96	4.22	1.38	<LOD	<LOD	3.09	3.97	35.79
	CT2	<LOD	0.74	3.83	3.56	6.41	7.26	3.74	1.83	<LOD	<LOD	2.36	4.35	27.36
	CT3	<LOD	2.85	10.92	18.99	19.73	13.32	7.67	2.72	<LOD	<LOD	10.42	11.28	76.20
	CT4	<LOD	3.82	12.63	20.90	25.96	13.52	12.14	3.23	<LOD	<LOD	10.65	13.01	92.21
	ME1	<LOD	0.57	1.57	2.42	2.04	1.70	1.13	0.10	<LOD	<LOD	1.07	1.19	9.52
	ME2	<LOD	0.18	1.17	1.55	1.80	1.26	1.15	0.37	<LOD	<LOD	0.62	0.95	7.48
	ME3	<LOD	0.22	1.19	1.22	0.83	1.20	0.43	0.17	<LOD	<LOD	0.62	0.61	5.27
	SN1	<LOD	0.53	3.72	4.82	4.77	3.19	3.38	0.44	<LOD	<LOD	2.52	2.90	20.85
	SN2	<LOD	0.24	1.21	1.68	2.00	1.31	1.37	0.34	<LOD	<LOD	0.83	1.05	8.15
	SN3	<LOD	0.69	2.53	3.08	4.07	1.92	1.45	0.54	<LOD	<LOD	1.13	1.71	14.27
Summer	CX1	<LOD	0.38	1.02	1.49	3.99	3.24	1.64	0.33	<LOD	<LOD	1.89	2.52	12.10
	CX2	<LOD	0.18	0.93	1.65	2.78	2.33	1.63	0.73	<LOD	<LOD	1.27	1.70	10.23
	CX3	<LOD	0.74	1.69	2.41	5.13	5.10	4.18	0.52	<LOD	<LOD	3.02	3.59	19.76
	CX4	<LOD	0.33	1.45	3.78	5.10	2.25	1.82	0.12	<LOD	<LOD	2.60	2.63	14.86
	CT1	<LOD	1.49	3.54	5.25	13.43	10.05	4.79	1.78	<LOD	<LOD	6.07	5.47	40.32
	CT2	<LOD	0.50	2.10	2.89	6.12	4.77	4.30	0.10	<LOD	<LOD	2.33	4.15	20.79
	CT3	<LOD	3.06	9.26	18.70	35.48	14.90	10.01	0.85	<LOD	<LOD	12.08	17.62	92.26
	CT4	<LOD	3.02	9.73	23.52	29.53	24.20	12.80	2.44	<LOD	<LOD	14.83	20.67	105.3
	ME1	<LOD	0.25	0.95	1.40	2.21	1.64	1.47	0.12	<LOD	<LOD	1.10	1.42	8.05
	ME2	<LOD	0.13	0.67	0.72	2.28	1.25	0.97	0.11	<LOD	<LOD	1.44	1.30	6.12
	ME3	<LOD	0.23	0.39	0.58	1.17	1.13	0.98	0.13	<LOD	<LOD	0.79	0.92	4.61
	SN1	<LOD	0.47	1.36	3.52	4.81	2.32	2.37	0.23	<LOD	<LOD	1.57	2.37	15.08
	SN2	<LOD	0.29	1.17	1.92	2.86	2.25	2.27	0.29	<LOD	<LOD	1.65	2.43	11.06
	SN3	<LOD	0.73	2.30	3.79	5.13	4.09	3.34	0.25	<LOD	<LOD	3.23	3.10	19.62

[a] Sum of 12 dioxin-like PCBs (4 non-ortho substituted PCBs: PCB77, 81, 126, 169; 8 mono-ortho substituted PCBs: PCB105, 114, 118, 123, 156, 157, 167, 189);

[b] Sum of 6 ICES (International Council for the Exploration of the Sea) indicator or marker PCBs (ICES6-PCBs) (PCB28, 52, 101, 138, 153, 180) (European Commission, 2011);

[c] Sum of 209 PCBs;

In addition, lack of significant association between PCB and TOC also indicates the PCB in the Bangladeshi coastal area mainly originated from local emissions or point sources, because Cui et al. (2016) reported that if the sources of sedimentary PCBs are controlled by atmospheric emission or long-range transport (non-point/diffuse sources), then the amounts of PCBs should be well correlated with the sedimentary TOC. Furthermore, sediment texture is considered to be one of the main factors affecting the accumulation profile of PCBs in sediment. In the present study, sediment quality characteristics ranged from silt loam or clay to sandy loam (Table A-9, Appendix A), but the texture varied seasonally. However, significant correlation between ∑PCBs and clay particles (winter: $r = 0.54$, $p < 0.001$; summer: $r = 0.64$, $p < 0.001$) implies the role of the fine-grained particles in the transportation and redistribution of PCBs in the coastal area of Bangladesh.

3.3.2.2 Seasonal and spatial distribution of sedimentary PCBs

The seasonal variations and spatial distributions of PCBs were investigated in surface sediment samples collected from the coastal area of Bangladesh, and the findings are presented in **Figure 3-4**. A two-tailed t-test analysis on both the winter and summer seasons' results were in favor of the fact that there was no significant difference between the levels of PCBs ($p > 0.05$). Furthermore, analysis of variance (ANOVA) results also revealed that more that 95% of the detected congeners did not differ significantly in their distribution patterns between the two seasons. This implied that the distribution pattern of PCB congeners in sediments along the coastlines studied were basically the same for both the seasons and that the PCBs were probably coming from the same sources (Rudel et al., 2008; Dodoo et al., 2012). However, seasonal variations to an extent were noticeable depending on the locations. For example, CT1, CT3, CT4, SN2 and SN3 showed slightly higher concentrations in summer, whereas the rest of the locations exhibited the reverse trends. The elevated levels of PCBs in summer samples from these locations might be due to the surface runoff from heavily contaminated sites resulted from heavy rain and floods, and intense shipping and fishing activities during summer. We suspected these reasons because CT3 and CT4 are located very close to the shipbreaking industries, whereas CT1, SN2 and SN3 are near shipping ports or fishing harbors. On the contrary, elevated levels of PCBs in winter samples, in the cases of CX1–4, CT2, ME1–3 and SN1, were probably due to either co-evaporation of PCB congeners with water (Larsson and Soedergren, 1987; Dodoo et al., 2012) or intense sedimentation from less mixing effects due to comparatively weaker tidal movement or wave action and lower inflow from upstream rivers in the dry season as well. This variation might also be influenced by the physico-chemical properties of the deposited sediments, absorption-desorption mechanism, dechlorination or degradation of PCB congeners along with other properties of themselves. However, the exact reasons are yet to be investigated in the study area. In general, regardless of sampling sites the mean concentration of ∑PCBs in summer (27.15 ng/g dw) was slightly higher than in winter (26.76 ng/g dw).

Spatial distributions of concentrations of ∑PCBs in the surface sediments from the Bangladeshi coastal area (**Figure 3-4**) revealed that the PCB concentrations varied widely depending on their locations. However, as for the spatial distribution of ∑PCBs in winter and summer seasons, it can be seen in **Figure 3-4** that the patterns of ∑PCBs' spatial distribution were almost similar. Furthermore, a high value for the correlation coefficient ($r = 0.99$, $p < 0.05$) was obtained, indicating that the PCBs exhibited very similar spatial distribution in the two periods. Concentrations of ∑PCBs were higher in the surface sediments at sampling locations CT1–CT4 (average of 57.89 and 64.65 ng/g dw in winter and summer, respectively), CX1–CX4 (average of 19.39 and 14.24 ng/g dw in winter and summer, respectively) and SN1–SN3 (average of 14.43 and 15.25 ng/g dw in winter and summer, respectively) in comparison to sampling locations ME1–ME3 (average of 7.42 and 6.26 ng/L ng/g dw in winter and summer, respectively), which indicates that the industrialized regions (Chittagong, Cox's Bazar and Sundarbans) in this area are potential sources of PCBs and that economic development seems to be associated with the amount of PCB emissions. Many previous studies reported the elevated PCB levels in sediments near industrialized areas around the world (Hong et al., 2005; Gedik et al., 2010; Barakat et al., 2013; Duan et al., 2013; Nouira et al., 2013), and thus it is not at all surprising to find that the more highly industrialized regions in the Bangladeshi coastal area contain higher PCB concentrations.

In particular, sediment samples at location CT4 showed the highest levels of PCBs for both seasons (92.21 and 105.3 ng/g dw in winter and summer, respectively) followed by CT3 (76.20 and 92.26 ng/g dw in winter and summer, respectively). Other two sites from Chittagong area, CT1 and CT2 also exhibited elevated PCB levels compared to other areas, indicating the existence of point source(s) in the area of Chittagong. CT3 and CT4 are located very close to the Chittagong ship breaking yard. Ship breaking activities along the coast of Chittagong may contribute to the PCB pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PCBs (Neşer et al., 2012; Siddiquee et al., 2012; Nøst et al., 2015). Large amounts of PCB were used (especially in the electrical cable sheathing, pains and hydraulic fluids) in the ships built before banning of PCB. It was estimated that each old ship contained 250 kg of PCBs, inclusive of transformer oil (DoE, 2007). It is thus, due to lack of proper management and monitoring, a huge amount of PCB might be released into the environment during the dismantling or demolition processes of old ships. There are approximately 100 e-waste recycling shops located very close to site CT3 and CT4, generating approximately 2.7 million metric tons of toxic e-waste each year, the majority of which comes from the ship breaking sector. However, the waste generated from the e-waste recycling shops is discharged directly into the adjacent coastal water, which might be one of the most significant PCB pollution sources in the study area. Nøst et al. (2015) confirmed PCB contamination in this area by measuring elevated levels of air born PCB which mainly originated from shipbreaking activities. Moreover, CT1 and CT2 are within the Chittagong port area which is the largest seaport in Bangladesh. These sites are predominantly influenced by the port activities. It was previously reported that the use of paints and grease repellents for ship and dock protection could contribute to the relatively high levels of PCBs in port seawaters. Shipping and boat maintenance in the ports may also be responsible for the release of PCBs into the surrounding waters (Rajendan et al., 2005, Wurl and Obbard, 2005). Moreover, there are numerous multipurpose industrial establishments along the coast of Chittagong producing paper and pulp (e.g., Karnaphuli paper mills), cement clinkers, fertilizers (e.g., Karnaphuli fertilizer), steel products, rubber and plastic, petroleum products (e.g., Super petrochemical), beverages, sugar, pharmaceuticals, tobacco, jute, textiles, printing and dyeing, fish products, tannery products, paint, rechargeable batteries, jewelry, plating, automobile engines, and electronics. The discharge of untreated or semi-treated effluents from these industries may pose a significant contribution to the PCB contamination into the nearby environment (Yang et al., 2011).

Within Cox's Bazar sites (CX1–4), sediment sample from CX3 (Bakkhali Estuary) showed elevated concentration of PCB (25.33 and 19.76 ng/g dw in winter and summer, respectively). It was expected because this site is adjacent to Cox's bazar city which has known a rapid industrialization and socio-economic development during the past 20 years where tourism is the main influential factor. The pollution in this area might be affected by Cox's bazar fishing harbor, marked by an intensive activity of fishery, fish processing works, boat manufacturing and painting and numerous multipurpose industrial facilities including e-waste recycling activities. All of these were identified to a considerable extent as potential contributing factors to the PCB contamination in the environment (Rajendan et al., 2005; Binelli et al., 2009). It should also be taken into account that the untreated or semi-treated municipal and industrial wastewaters from the Cox's bazar city are discharged into the Bakkhali estuary through the Bakkhali channel. In addition, the main municipal garbage dump (mostly consumer products including obsolete electronics) of Cox's Bazar city is also located very close to CX3. Therefore, a significant amount of PCB might be emitted from this dumping site into the adjacent environment. However, sediments from other 3 locations (CX1, CX2 and CX4) had almost similar levels of PCBs, suggesting a similar impact and that of mainly from industrial and urban discharges along with runoff from contaminated sites.

In case of Sundarbans sites (SN1–3), the levels of PCB in sediment showed an interesting trend: concentration of PCB first decreased from upper estuary (SN3) to mid-estuary (SN2) and then increased further towards lower estuary (SN1) or mouth of the estuary to the Bay of Bengal. The site SN3 is located vary close to Mongla port. There is high density of shipping activities in and around the port area and hence high PCB levels are related to potential discharges from the ships. Higher PCB levels might be attributed to the huge discharge of untreated or partially treated effluents from numerous multipurpose industries such as cement, paint, paper, printing and dyeing, plastics, leather, etc. Sediments from locations close proximity to these industries have been reported to be heavily contaminated with POPs, including PCBs (Hong et al., 2005; Nie et al., 2005; Zhou et al., 2012; Duan et al., 2013; Nouira et al., 2013). In addition, intense dredging operations in this area along with agricultural runoffs, dumping of industrial wastes, surface runoff and atmospheric depositions further aggravate the PCB pollution. The decrease in PCB concentration in sediment from mid-estuary (SN2) might be attributed to the dilution effects from mixing of inland fresh water with the salty seawater (An et al., 2009; Men et al., 2014). However, a further increase in PCB concentrations near the mouth of the estuary (SN1) might be attributed to the river-sea boundary zone sedimentation or marginal filter effects (Wang et al., 2016). A marginal filter functions as a barrier that contributes to sorption and sedimentation (Yang et al., 2012) resulting in high contamination. Besides, fine particles carried by the inland rivers to the zone where the rivers discharge their loads might be a major carrier of PCBs from the upstream source areas to the mouth of the estuary. In addition, many ships and fishing boats travel in this area, which might be associated with the increase of PCBs as well.

The levels of PCB in sediments taken from the Meghna estuary also showed a downward increasing trend. It is to be noted that the Meghna estuary is an exclusively unindustrialized area. Therefore, lower in concentration but detection of PCB in this area suggest that there are non-point sources of PCBs (e.g., surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc.) in the coastal area of Bangladesh. Furthermore, the Meghna Estuary receives major volumes of water from the Ganges River of India through the Yamuna River flowing down to the Meghna Estuary via the Mehgna River. Previous studies reported that these rivers were contaminated by PCBs (Chakraborty et al., 2014; Kumar et al., 2013). The authors also noted that PCB concentrations gradually declined due to dilution as the water mass increased downstream of the rivers. Thus, the coastal areas of Bangladesh might receive PCBs to an extent through the water flow of these upstream rivers, which flow into the Meghna Estuary sites. And PCBs transported by rivers together with suspended particles are likely deposited near the river mouth by avalanche-type sedimentation. This rapid sedimentation in the river mouth within the river-seawater mixing zone might be responsible for higher accumulation of PCBs in sediment (Nemirovskaya, 2009; Yang et al., 2012). Generally speaking, the concentrations of ∑PCBs in surface sediments at the Chittagong (CT1–CT4), Cox’s Bazar (CX1–CX4) and Sundarbans sites (SN1–SN3) were higher than those in the Meghna Estuary (ME1–ME3) and because of greater development, thus associating these compounds to urbanization and industrialization.

3.3.2.3 Homolog composition

Figure 3-5 shows the PCB homologs composition (%) in the surface sediment samples for both seasons (winter and summer). Regardless of sites and seasons, the proportions of the homologs with different degree of chlorination were in the decreasing order of: medium (tetra- to hexa-CBs) > heavier (hepta- to deca-CBs) > lighter (mono- to tri-CBs), contributing 57–75%, 12–24%, and 10–15% to the ∑PCBs, respectively. In general, penta-CB was the most prevalent homolog in the sediment samples, accounting for 16–29% and 25–38% of ∑PCBs in winter and summer, respectively, followed by hexa-CB (12–21% and 12–24% of ∑PCBs in winter and summer, respectively) and tetra-CB (10–28% and 11–15% of ∑PCBs in winter and summer, respectively). Although the observed trends in PCB homologs composition were almost similar in the majority of the samples (**Figure 3-5**), the distribution patterns are to some extent different for some locations suggesting different or multiple input sources of PCBs in the study area (Barakat et al., 2002). Overall, this pattern indicates the existence of local input sources and possibly ongoing release into the study area as well. In the Bay of Bengal of India, Rajendran et al. (2005) reported a 65–78% dominance of tetra-, penta- and hexa-CBs at some sites while other sites, di-, tri- and tetra-PCB homologs were more prevalent. Men et al. (2014) reported the main homolog groups were penta- to octa-CBs in sediment of the Daliao River estuary. Sediments of Alexandria Harbor, Egypt were predominantly composed of tetra- to hepta-CBs (Barakat et al., 2002). Montuori et al. (2014) also reported that penta- and hexa-CBs were abundant in sediments from Sarno River and Estuary, Southern Italy, accounting, for 37 and 25 % of ∑PCBs, respectively. The abundance of heavier homologs in these researches was explained as the dominance of recent releases from the local inputs of PCBs in the study areas. However, the dominance of tri- and tetra-CBs due to the impact of historical PCB usage were reported in the surface sediments from Hyeongsan River of Korea (Koh et al. 2004) and Sea Lots, Port-of-Spain, Trinidad and Tobago (Mohammed et al., 2011).

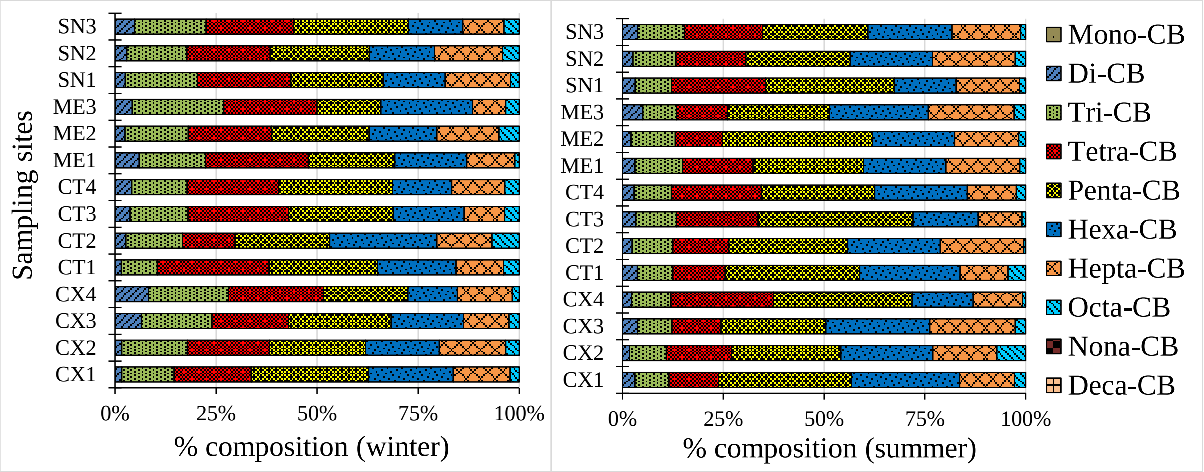

Figure 3-5. Relative contribution of PCB homologs (% composition) to the total PCB in the surface sediment of the Bangladeshi coastal area in winter and summer, 2015.

Although the homolog composition did not differ significantly between the two seasons ($p > 0.05$), collectively a shift to a smaller extent towards high chlorinated homologs were noticeable in the case of summer samples (**Figure 3-5**). This could be attributed to an assumption that the deposited sediments during summer might be originated from the surface soils that already enriched with high chlorinated PCB congeners which were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. Ilyas et al. (2011b) reported the dominance of hexa-, hepta- and penta-CBs in the surface soils from the coastal area of Surabaya, Indonesia. Ren et al. (2007) also reported that the hexa-CB (31%) was the most abundant homolog for Chinese urban surface soils.

3.3.2.4 Congener profiles and source characterization

We identified the dominant PCBs in the sediment samples both by occurrence and abundance by combining the detection rate and mean relative concentration of each congener/co-elute across samples and ranked them averagely in decreasing order. The most abundant twenty congeners were PCB120+110, 118, 138, 153, 101, 180, 102+93+98+95, 170, 149+139, 43+49, 70, 105, 128, 66, 18, 44, 28, 20+33, 52, and 74, comprising up to 46–75% of ∑PCBs by sites, and sum of them was highly correlated with ∑PCB (r = 0.993, winter; 0.996, summer; $p < 0.05$), well representing the ∑PCB in the surface sediment of the Bangladeshi coastal area. Kobayashi et al. (2010) also reported a similar assemblage of dominant congeners (e.g. PCB28, 74, 70, 66, 120+110, 118, 153, and 138) in the sediment samples from the Tokyo Bay, Japan. However, the dominant congeners identified in the present study corresponds to diverse homolog groups (tri- to hepta-CBs), suggesting multiple sources of PCBs in this area. Sahu et al. (2009) reported that the presence of different congeners belonging to different homologs was an indication of various sources of PCBs in marine sediments.

Apparently, the dominant congeners in the sediment samples were also key constituents of several commercial PCB mixtures, e.g. Aroclors 1242, 1248, 1254, 1260 (Jones, 1988; Ishikawa et al., 2007). It indicates that the environmental PCB burdens in the Bangladeshi coastal sediments might be originated from more than one PCB Aroclors and the most likely mixtures were Aroclors 1248, 1254 and 1260. PCBs were never produced, but only imported for use in Bangladesh. A general assessment of PCB use together with information on the available import records, therein termed transformer oil, in Bangladesh is given elsewhere (DoE, 2007; ESDO, 2005a,b; 2010), and we discussed briefly on this in the earlier sections of this chapter. Presently, no specific information exists regarding the identity and quantitative proof of PCB mixtures used in Bangladesh historically. This lack of information results in difficulties during the selection of PCB mixtures for consideration in sources analyses. Since the 1980s, national environmental regulations have existed in Bangladesh, such as the ban of PCBs (ESDO, 2005a,b; 2010; Nøst et al., 2015). However, PCB compounds are still in use in Bangladesh, mostly in closed systems as dielectrics in transformers and capacitors being operated in the electrical generating sector (DoE, 2007).

Moreover, the outdated PCB-containing equipment, (old transformers, capacitors, etc.) along with non-recycled e-waste mostly originated from ship breaking industries is simply deposited in landfills particularly in coastal areas. Aroclor 1242, 1254 and 1260 were extensively used in closed systems like transformers and capacitors. Aroclor 1248 and 1254 were also widely used as plasticizer in multipurpose industries (Sahu et al., 2009). Therefore, leakage from the PCB-containing in-service equipment, effluents from PCB using industries and leachates from landfill sites might be considered as potential sources of significant PCB inputs into the environment (Fu et al., 2003; Nie et al., 2005). Furthermore, together with the identified most abundant congeners, there were hundreds of other congeners found in the sediment samples investigated which might be originated from different sources including point and non-point/diffuse sources, suggesting a possibility of multidimensional impact in the study area. For example, PCB11 was determined in 50% and 64% of the sediment samples, in winter and summer, respectively, although contributing only 0.20–2.42% of ∑PCBs. PCB 11 is a non-legacy signature of PCB contamination, being absent in commercial PCB formulations, but largely originates as a byproduct of paint and pigment production and a residue in printed products and painted surfaces (Hu and Hornbuckle, 2010; Hu et al., 2014; Vorkamp, 2016). Therefore, the effluents from industries that are using and/or producing pigments and dyes, printing of consumer goods (e.g. magazines, books, maps, posters, brochures, napkins, garments, etc.) along with surface paintings of ships and fishing vessels should be considered as PCB source in the study area. Moreover, Baek et al. (2010) suggested that the source of PCB11 is probably the building materials or the decoration and furnishings of the buildings, particularly in the urban residential area. However, PCB11 may be also produced by the dechlorination from dioxin-like PCBs such as PCB77 (Kobayashi et al., 2010). In fact, the specific source of PCB11 in sediments is still not clearly defined.

Among the 12 DL-PCBs studied, PCB105, 114, 118, 123, 156, 157, 167, and 189 were observed in all of the samples for both seasons, whereas PCB77, 81, 126 and 169 were found in 86, 93, 29, 71% and 100, 100, 71, 93% of winter and summer samples, respectively. Thus, emission from industrial applications might be the major sources of DL-PCBs in the study area (Azari et al. 2007), whereas combustion processes apparently contributed to some extent what confirms the presence of PCB77 and 126 (marker PCBs for combustion processes) with a contribution of up to 1.6% of ∑PCBs. According to Chi et al. (2007), non-ortho-PCBs that are characteristic of coal combustion and industrial waste incineration, do not originate solely from commercial PCB formulations.

3.3.2.5 Principal component analysis (PCA)

To further understand the source origins of PCB emissions in the Bangladeshi coastal area, the PCB congener compositions in the sediment samples were subjected to principal component analysis (PCA) in a varimax rotation mode (**Figure 3-6**). When the congeners of a specific organic contaminant are loaded onto a PCA statistical platform, the data points can be rotated such that maximum variability is visible, and points affected by same factor aggregate into a cluster. The presence of congener markers within a cluster may provide a clue regarding probable source of origin/emission. In this study, only congeners with values greater than limit of detection in more than 40% of samples were used for PCA analysis. In addition, we used BC-normalized concentrations of PCBs because the concentrations of PCBs showed strong and positive association with sedimentary BC, which suggested that the BC was the key influential factor in explaining the distribution of PCBs in the Bangladeshi coastal sediments.

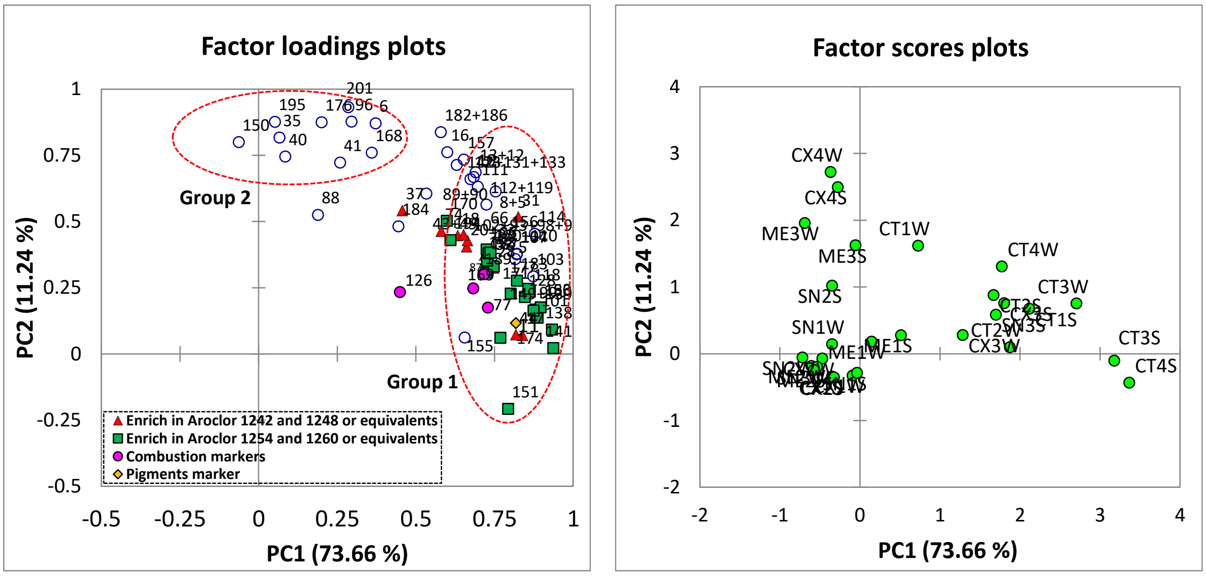

Figure 3-6. Factor loadings and corresponding factor scores plots for PCB congeners in sediment samples. Numbers on factor loading plots indicate PCB congeners. On the factor scores plots, site IDs with W and S indicate winter and summer season, respectively. Marker congeners were obtained from the work of Anezaki et al. (2016).

The first PC (PC1) accounted for 73.66 % of the total variance, whereas the second (PC2) reached 11.24 %, for a cumulative explained variance of 84.90 % of the total variance. The PC1 correlated highly with the Group 1 cluster of PCB congeners, which are mostly moderate to highly chlorinated (**Figure 3-6**). This cluster to an extent might denote local impacts, given that heavier congeners travel relatively poorly and tend to accumulate/adsorb more readily to sediments near sources (Tolosa et al., 1995; Borja et al., 2005). Surprisingly, many characteristic congeners of different PCB formulations (e.g. Aroclor 1242, 1248, 1254 and 1260 or equivalents), combustion (e.g. PCB77, 169, 189) and pigments and dyes (e.g. PCB11, a non-aroclor marker for paints and pigments) (Anezaki et al., 2016) were clustered in Group 1, suggesting that either of these sources might be important in local emissions of PCBs in the study area. Interestingly, all of the twenty identified most abundant congeners in sediment samples belong to this cluster, and majority of them are enrich in Aroclor 1254 and 1260 or equivalents. However, the prevailing abundance of these Aroclor congeners in recently deposited sediments is of interest because its production and/or application was already banned decades ago, suggesting recent inputs and/or ongoing usage of PCB compounds in Bangladesh. In addition, The Group 2 set of congeners, which correlated highly with PC2, contains PCB6, 35, 40, 41, 96, 150, 168, 176, 195, and 201. According to Anezaki et al. (2016), among these congeners PCB35, 41 and 195 might be originated from combustion and carried into the coastal environment by atmospheric wet and dry deposition. However, the exact sources of the congeners clustered in Group 2 still remain unidentified.

Based on the PCB homolog compositions, the pattern of individual congeners in the sediment along with the extracted PCA results, we can say that point and/or land-based sources was possibly the major input of the PCBs to the Bangladeshi coastal area, but there still existed the inputs from atmospheric transport and deposition. However, due to lack of previous data, suspected multi-dimensional sources of PCB inputs, both point and diffusive, along with complex nature of the environment, it is difficult to pinpoint precisely a clear source occurrence relationship in the Bangladeshi coastal area. Further study will be focused on sedimentary core analysis or depth profiles of PCBs to explore an in-depth scenario of PCB contamination in a complex ecosystem with diverse ecological features and resources.

3.3.2.6 Comparison with previous studies worldwide

The levels of PCBs in the surface sediment form the Bangladeshi coastal area were compared with that of from other coastal areas around the world and presented in **Table 3-4**. To some degree, the results of comparison could at least reflect the pollution levels of PCBs in sediment, although the numbers of monitored PCB congeners, the sampling time and methods, and analytical processes among these studies are different. In general, elevated concentrations have generally been observed in those countries previously involved in PCB manufacture, particularly the USA, Spain, and Japan. In the present study, the concentration of PCBs in sediments was close to the levels in sediment from Tokyo Bay (Japan) and Italian coastal areas. The levels were remarkably higher compared to concentrations reported for coastal areas in Korea, China, and Taiwan, whereas evidently lower than those from Spain, Egypt, Trinidad and Tobago, USA and Indonesia (**Table 3-4**).

Table 3-4. Comparison of total PCBs (ng/g dw) in sediments of coastal areas worldwide.

Location	Sampling year	*N*[a]	∑PCBs	References
Masan Bay, Korea	1997	22	1.24–41.4	Hong et al., 2003
Daya Bay, China	2003	136	8.7−34.3	Wang et al., 2008
Bay of Bengal, India	1998	–[b]	0.02–6.57	Rajendran et al., 2005
Italian coastal areas	2001	36	0.3–84	Pozo et al., 2009
Southwest coast of Taiwan	2005	113	0.88–7.13	Jiang et al., 2011
Tokyo Bay, Japan	2006	209	2.7–110	Kobayashi et al., 2010
Commercial ports, Spain	2001–03	7	<LOD–468.2	Casado-Martínez et al., 2006
Alexandria Harbor, Egypt	1998	96	0.9–1211	Barakat et al., 2002
Sea Lots, Trinidad and Tobago	2009	136	62–601	Mohammed et al., 2011
Salton Sea, California, USA	2000	55	116–304	Sapozhnikova et al., 2004
Coastal area of Surabaya, Indonesia	2008	62	<LOD–420	Ilyas et al., 2011a
Coastal area of Bangladesh	2015	209	5.3−92.21 (W)[c] 4.6−105.3 (S)[c]	This study

[a] Number of PCB congeners;
[b] Sum of di- to deca-CB homologs;
[c] W and S represent winter and summer, respectively;

Remarkably, our results (4.6–105.3 ng/g dw) were far higher than the PCB levels found in sediments from the Bay of Bengal of Indian coast (0.02–6.57 ng/g dw) although the sampling was done about two decades ago (Rajendran et al., 2005). This huge difference in PCB concentrations among the neighboring areas implies that some important and/or significant potential sources of PCBs exist in the Bangladeshi coast of the Bay of Bengal. Additionally, as no data is available for past levels of PCBs in the studied area, no conclusion can be made concerning temporal changes in PCB levels in the Bangladeshi coastal area.

Since the physico-chemical properties of PCBs are dependent on their degree of chlorination (Fiedler, 1997), the analysis of PCB congeners and homologue profiles provide useful insights on the behavior and the possible sources of PCBs to the marine or coastal environments (Mai et al., 2005; Li et al., 2009; Barakat et al., 2013). The PCBs homologues profiles in sediments vary depending on PCBs sources, age of the contamination, and environmental conditions as well as properties of PCB itself (Li et al., 2012). Low-chlorinated PCBs are more volatile, highly water soluble and less persistent in the marine environment, being usually related to long-range transport (e.g. atmospheric wet and dry deposition and ocean currents/tides driven migration), and/or dechlorination processes, while the abundance of higher chlorinated compounds are an indication of low influence or absence of dechlorination/degradation or the presence of local input sources or recent releases into the environment, and influenced by contaminated areas as well (Tolosa et al., 1995; Hong et al., 2005; Borja et al., 2005). Moreover, lower concentration of total PCBs with higher proportions of high chlorinated congeners is associated with the places further away from direct pollutant sources, where precipitation was usually the main pathway and source of the PCBs (Goerke and Weber, 1998).

3.3.3 Ecotoxicological concern

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

Considering all the sites, irrespective to seasons, the concentrations of ∑PCBs (32.17–199.35 ng/L) in the surface water were higher than the criterion continuous concentration (CCC) for water quality (14 and 30 ng/L for inland and coastal water, respectively) recommended by the USEPA for the protection of aquatic and human health (USEPA, 2010). The levels also exceeded the Chinese national environmental quality standards for surface water (GB 3838-2002) where the concentration of ∑PCBs was restricted to 20 ng/L. Therefore, the quality of surface water of the Bangladeshi coastal area is heavily polluted with PCBs, suggesting that the toxicity of the PCBs in the water ecosystem could adversely affect the ecological environment and human health through biomagnification.

Many approaches have been developed to evaluate the ecotoxicological aspect of sediment-bound PCB contamination. One such evaluation method is the sediment quality guidelines (SQG). In this study, we followed two most common SQGs: (a) sediment quality guidelines developed by Long et al. (1995) and (b) Canadian Environmental Quality

Guidelines (CCME, 1999). None of the stations in either seasons exceeded the “effect range median” (ERM, 180 ng/g) or “probable effect level” (PEL, 189 ng/g). However, ∑PCBs concentrations were higher than the “effect range low” (ERL, 22.7 ng/g), the “interim sediment quality guideline” (ISQG, 21.5 ng/g) in about 35% and 22% of stations in winter and summer, respectively, indicating the potential adverse effects on benthic and bentho-pelagic organisms may occasionally occur. The concentrations of ∑PCBs higher than ERL/ISQG were observed at sites located in the vicinity of Chittagong ship breaking (CT3 and CT4) and port area (CT1 and CT2), and Bakkhali estuary near a fishing harbor in Cox’s Bazar (CX3).

To assess the potential dioxin-like toxicity of the sediment samples, we estimated the toxic equivalency (TEQ) concentrations of the sediment-bound PCBs by using the 2005 WHO toxic equivalent factors (TEFs) (Van den Berg et al., 2006). The WHO_{2005}-TEQs for the 12 DL-PCBs in sediments of the present study varied from 0.06 to 3.59 pg TEQ/g dw in winter and 0.8 to 4.16 pg TEQ/g dw in summer, respectively. The WHO_{2005}-TEQs in 85% of the samples, irrespective to seasons, were higher the interim sediment quality guideline of 0.85 pg TEQ/g dw recommended by the Canadian Council of Ministers of the Environment (CCME, 2002). Thus the sediment-bound DL-PCB concentrations are likely to pose threat to benthic organisms in the coastal area of Bangladesh.

3.3.4 PCBs in seafood

3.3.4.1 Concentration of PCBs in seafood and global comparison

PCBs were detected in all of the 48 seafood samples including 5 finfish and 2 shellfish species, suggesting the ubiquitous occurrence and continuous accumulation of these compounds in the Bangladeshi seafood. The data of PCBs in seafood samples are presented in **Table 3-5** and **Figure 3-7**, whereas the detailed results are presented in Table A-10 and A-11 (Appendix A). A total of 141 domains comprising 114 single congeners, 20 double coeluting isomers, 5 triple coeluting isomers, and 2 quadruple coeluting isomers were resolved in 48 tissue samples regardless of species and seasons. However, the detected PCBs were only the congeners with 2 to 8 Cl atoms. Congeners with 1 Cl atom might evaporate in the pretreatment procedure, and congeners with 9 and 10 Cl atoms were under their LODs.

Table 3-5. Concentrations (ng/g ww) of PCBs and fat content (%) in seafood (finfish and shellfish) collected from the coastal area of Bangladesh in the two seasons.

PCB homolog	Finfish																Shellfish							
	Ilish				Rupchanda		Loitta				Sole		Poa				Shrimp				Crab			
	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	BH	SN	CX	CT	BH	SN
Winter																								
Fat (%)	19.3	21.1	14.5	13.2	2.8	2.1	2.6	2.9	1.9	2.0	3.1	2.2	3.0	3.7	3.0	2.3	2.0	1.9	1.2	3.0	4.1	4.5	3.6	3.9
Mono-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Di-CB	1.32	1.05	1.24	1.33	0.38	0.31	0.54	0.41	0.16	0.42	1.04	0.56	0.50	0.42	0.38	0.20	0.26	0.17	0.03	0.57	1.43	0.94	0.46	1.34
Tri-CB	4.25	3.83	3.62	4.35	1.18	1.22	1.81	1.51	0.68	0.80	1.94	2.12	1.63	1.28	0.94	0.85	1.03	0.63	0.55	1.83	4.26	4.08	2.32	2.90
Tetra-CB	7.75	13.60	7.62	7.37	1.87	1.43	2.53	2.64	1.03	1.34	2.62	4.22	2.56	3.11	1.61	1.37	1.90	1.34	1.26	2.12	5.96	5.72	2.26	3.34
Penta-CB	22.84	22.45	17.40	15.70	2.71	2.59	4.80	6.62	2.03	2.14	7.30	9.52	4.65	5.26	3.22	2.21	2.71	2.13	1.14	2.80	7.76	10.81	3.73	7.14
Hexa-CB	18.36	19.43	13.49	13.69	2.93	2.15	4.07	4.96	1.37	1.51	7.05	7.53	5.27	6.86	4.68	1.97	1.86	1.38	1.02	2.89	7.27	7.64	7.00	6.17
Hepta-CB	17.54	18.22	11.54	11.48	2.22	1.53	2.53	3.81	1.03	1.57	4.67	5.11	4.25	4.15	1.86	1.12	1.33	1.60	1.11	2.14	6.48	7.26	4.87	6.03
Octa-CB	0.57	1.03	0.88	0.60	0.08	0.14	0.46	0.31	0.09	0.13	0.39	0.47	0.46	0.29	0.15	0.12	0.12	0.05	0.05	0.28	1.12	1.26	0.63	0.86
Nona-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Deca-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑DL-PCBs[a]	8.76	7.85	5.62	5.79	1.04	1.16	2.09	2.51	0.78	0.87	2.73	4.11	2.16	2.47	1.68	1.12	1.24	1.14	0.64	1.66	5.00	4.64	3.15	3.72
∑iPCBs[b]	17.18	17.27	15.37	14.93	3.07	2.26	4.14	5.58	1.58	2.07	6.87	7.08	4.71	5.67	4.09	1.77	1.88	1.77	1.06	2.56	6.38	7.32	4.24	5.08
∑PCBs[c]	72.63	79.62	55.80	54.52	11.37	9.37	16.74	20.26	6.39	7.92	25.01	29.53	19.32	21.35	12.84	7.84	9.20	7.30	5.16	12.64	34.29	37.70	21.27	27.78
Summer																								
Fat (%)	20.1	22.3	16.2	14.1	3.5	3.0	2.4	3.5	1.9	2.1	3.4	3.8	3.1	3.9	2.9	2.4	2.3	2.0	1.5	2.6	4.0	4.2	3.1	3.4
Mono-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Di-CB	2.93	3.12	1.83	2.22	0.48	0.42	0.55	0.44	0.24	0.30	1.30	0.52	0.29	0.47	0.47	0.21	0.07	0.12	0.02	0.19	1.26	1.02	0.40	0.86
Tri-CB	7.22	4.66	5.83	5.03	1.19	1.66	2.59	2.95	1.12	0.79	5.07	5.80	3.44	1.91	1.04	0.78	0.92	0.68	0.49	1.10	3.43	4.52	1.61	1.94
Tetra-CB	13.54	16.46	14.33	12.51	2.84	1.94	4.61	4.51	1.15	1.85	5.41	7.18	3.59	6.96	4.08	1.82	1.77	1.67	1.13	1.76	4.55	5.30	1.94	2.38
Penta-CB	22.89	24.57	18.57	15.29	5.03	2.49	5.05	8.16	1.81	2.90	9.79	11.33	5.30	6.90	5.25	2.81	2.27	1.45	0.87	1.86	5.32	7.04	2.73	3.86
Hexa-CB	17.14	22.26	15.11	15.88	5.23	2.43	3.74	5.10	1.85	2.50	7.91	8.14	6.60	5.66	4.71	2.08	1.52	0.90	0.68	2.24	5.21	6.21	4.17	5.36
Hepta-CB	12.09	13.77	12.25	9.91	2.56	1.31	2.28	3.50	1.22	2.48	5.32	5.95	3.61	4.14	2.11	1.39	0.87	1.00	0.62	1.27	4.54	5.36	2.88	6.04
Octa-CB	2.36	1.35	1.17	0.84	0.13	0.24	0.56	0.32	0.16	0.28	1.06	0.72	0.72	0.78	0.44	0.14	0.09	0.02	0.02	0.20	0.67	0.62	0.47	0.43
Nona-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Deca-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑DL-PCBs	10.86	13.20	9.52	8.45	1.55	1.43	2.53	3.66	0.68	1.03	3.55	4.18	2.56	2.69	2.04	1.32	1.01	0.93	0.42	1.21	3.24	3.12	1.87	1.73
∑iPCBs	14.70	17.15	14.67	11.15	4.41	2.52	3.63	5.24	2.11	3.08	7.00	8.54	4.84	6.41	3.63	1.99	1.31	1.30	0.87	1.74	4.59	6.86	3.61	5.09
∑PCBs	78.16	86.18	69.10	61.67	17.47	10.50	19.39	24.98	7.55	11.11	35.85	39.65	23.54	26.81	18.11	9.23	7.51	5.85	3.82	8.63	24.97	30.06	14.20	20.87

[a] Sum of 12 dioxin-like PCBs (4 non-ortho substituted PCBs: PCB77, 81, 126, 169; 8 mono-ortho substituted PCBs: PCB105, 114, 118, 123, 156, 157, 167, 189); [b] Sum of 6 ICES indicator or marker PCBs (ICES6-PCBs: PCB28, 52, 101, 138, 153, and 180) (European Commission, 2011); [c] The concentrations of congeners with <LOD were assumed to be zero while calculating the total PCBs; CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans;

The total concentrations of the PCBs (∑PCBs, sum of detected domains) in the seafood samples ranged from 5.16 to 79.62 ng/g ww in winter, and from 3.82 to 86.18 ng/g ww in summer. The distribution of samples by class of ∑PCBs levels were: 17% (>50 ng/g ww), 17% (25–50 ng/g ww), 66% (<25 ng/g ww). The ∑PCBs were positively correlated with the number of detected domains per sample ($r = 0.86$, $p < 0.05$). However, the ∑PCBs variation might be dependent on some dominant congeners. The sum of 12 dioxin-like PCB (∑DL-PCBs) and 6 ICES (International Council for the Exploration of the Sea) indicator/marker PCB congeners (∑iPCBs) ranged from 0.64 to 8.76 ng/g ww (mean: 3 ng/g ww) and 1.06 to 17.27 ng/g ww (mean: 6 ng/g ww) in winter, and from 0.42 to 13.2 ng/g ww (mean: 3.45 ng/g ww) and 0.87 to 17.15 ng/g ww (mean: 5.69 ng/g ww) in summer, respectively. The ∑DL-PCBs and ∑iPCBs together account for 26–47% of the ∑PCBs concentrations. Therefore, the DL-PCBs and iPCBs could not comprehensively reflect the contamination level of PCB in seafood, elucidating that the degree of PCB contamination in the Bangladeshi coastal area might be impacted by some other congeners along with these frequently monitored PCBs. However, correlations between ∑PCBs and ∑DL-PCBs, and ∑iPCBs were strong ($r = 0.98$ and 0.99, respectively) and significant ($p < 0.05$).

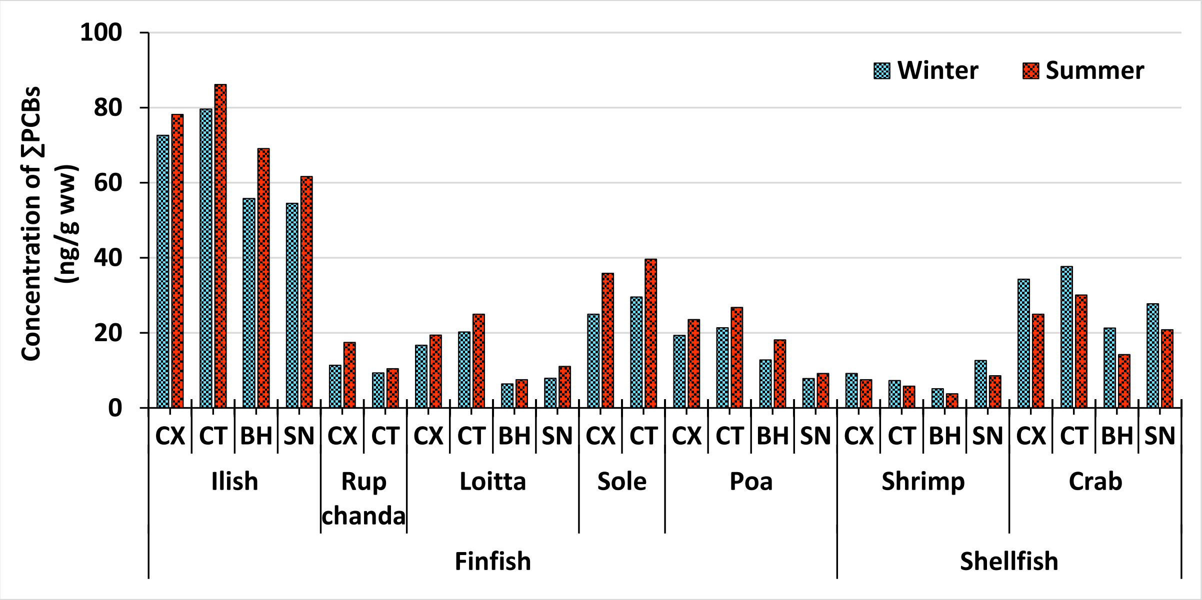

Figure 3-7. Concentrations of total PCB (∑PCBs) in the seafood samples (finfish and shellfish) from the coastal area of Bangladesh in winter and summer. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans.

The levels of PCBs in the Bangladeshi seafood were compared with that of from other areas around the world and presented in **Table 3-6**. To some degree, the results of comparison could at least reflect the status of PCB contamination in the Bangladeshi seafood, although the numbers of monitored PCB congeners, the sampling time and methods, and the investigated species among these studies might be different from each other. The comparison results revealed that the ∑PCBs concentrations for both seasons in this study were comparable or higher than those observed in finfish and shellfish from Spain (Perello et al., 2012), South Korea (Moon et al., 2009), Dalian, Tianjin and Shanghai in China (Yang et al., 2006), Indonesia (Sudaryanto et al., 2007), Eastern Coast of Thailand (Jaikanlaya et al., 2009), Southern Iran (Mohebbi-Nozar et al., 2014) and Hong Kong (Chan et al., 1999), but lower than that from Scheldt estuary (Netherlands–Belgium) (Van Ael et al., 2012), Houston Ship Channel, USA (Howell et al., 2008), and Hyderabad, India (Ahmed et al. 2016). Besides, the ∑PCBs in half of the seafood samples (50–58%) were greater than the USEPA screening value of 20 ng/g ww (USEPA, 2000) and were comparable to or at the middle range of the worldwide PCBs data in edible marine species (Domingo and Bocio, 2007). Therefore, the present level of PCBs measured in the Bangladeshi seafood is obviously a matter of concern.

Table 3-6. Global comparison of total PCBs (ng/g ww) in seafood.

Location	Sampling year	Species	*N*[a]	∑PCBs	References
Twelve cities of Catalonia, Spain	2008	Finfish and shellfish	18	0.35–48.8	Perello et al., 2012
South Korea	2005–07	Finfish and shellfish	22	0.2–41	Moon et al., 2009
Dalian, Tianjin and Shanghai in China	2002	Finfish and shellfish	–[b]	0.83–11.4	Yang et al., 2006
Scheldt estuary (Netherlands–Belgium)	2010	Finfish and shellfish	33	3.27–285	Van Ael et al., 2012
Indonesia	2003	Finfish	NA	23	Sudaryanto et al., 2007
Eastern Coast of Thailand	2006–07	Shellfish	49	0.05–7.53	Jaikanlaya et al., 2009
Houston Ship Channel, USA	2003	Finfish and shellfish	209	3.44–1596	Howell et al., 2008
Southern Iran	2010–11	Finfish and shellfish	7	1.91–7.05	Mohebbi-Nozar et al., 2014
Hyderabad, India	2013–14	Finfish	45	0.22–118.7	Ahmed et al. 2016
Hong Kong	1997	Finfish	51	<0.01–94	Chan et al., 1999
Coastal area of Bangladesh	2015	Finfish and shellfish	209	5.16−79.48 (W)[c] 3.82−86.04 (S)[c]	This study

[a] Number of PCB congeners;

[b] Sum of di- to deca-CB homologues;

[c] W and S represent winter and summer, respectively;

3.3.4.2 Influence of seasons and species on PCB accumulation in seafood

The levels of ∑PCBs in finfish were slightly higher in summer (7.55–86.18 ng/g ww) than in winter (6.39–79.62 ng/g ww), whereas in shellfish it was higher to an extent in winter (5.16–37.70 ng/g ww) compared to summer (3.82–30.06 ng/g ww), although these differences were not statistically significant in either cases ($p > 0.05$). These variations might be attributed to the seasonal discrepancies in their physiological activities, feeding behavior as well as the degree of contamination of their habitats and these will be explained in details in the following discussions. As a whole, the loads of ∑PCBs in the Bangladeshi seafood after combining finfish and shellfish data together did not differ significantly between winter and summer ($p > 0.05$), elucidating that the present study area might be influenced by almost similar sources in the two seasons and that are mainly from local inputs rather than atmospheric and/or long range transportation.

Significant interspecies differences were found with regard to ∑PCBs concentrations in the four coastal areas ($p < 0.05$). In particular, the highest levels of ∑PCBs were found in Ilish (54.52 –79.62 and 61.67–86.18 ng/g ww in winter and summer, respectively) among the finfish species and Crab (21.27–37.70 and 14.2–30.06 ng/g ww in winter and summer, respectively) within the shellfish species, probably due to the elevated lipid content in these species (**Table 3-5**). PCBs accumulate in adipose tissue and thus tend to increase with lipid content (Batang et al., 2016). Our analysis revealed a positive and strong correlation between the levels of PCB and the lipid content in the studied seafood samples ($r = 0.95$; $p < 0.01$). In finfish, irrespective to seasons, the concentrations of ∑PCBs showed the following trend: Ilish > Sole > Poa > Loitta > Rupchanda. This pattern correlates well with lipid yields obtained from the individual seafood species. Besides the species-specific lipid content, also biological (trophic level, feeding behavior, reproductive status and metabolism including uptake and elimination) and environmental parameters (depth and habitat location) might influence the pollutant body burden. Comparatively higher PCBs levels along with larger number of detected domains in Ilish could be explained by the following phenomena: [1] It is a high trophic carnivorous species that tends to concentrate contaminants to a higher degree than other species (Das and Das, 2004; Miao et al., 2000); [2] It is an anadromous species which migrates from the sea to the rivers for spawning. Consequently, this species is habitated to different ecosystems (marine, estuarine, brackish and freshwater) and exposed to various degree of contamination; [3] Ilish has relatively a larger body surface area with a very thin layer of skin and a larger gill surface area that facilitate the process of accumulation of the contaminants like PCBs from water column into the fish body. McKim and Heath (2010) reported that PCBs can undergo bioconcentration via gills in fish although the main uptake is through their diet. In addition, there were elevated concentrations of PCBs in the Sole tissue, which was perhaps relative to their living habits. PCBs were prone to sink in the sediments where the benthic fish such as Sole often predate benthic organisms or eat some organic particles, and so tend to accumulate these organic compounds like PCBs.

Within shellfish species, ∑PCBs levels in Shrimp were consistently low and of fewer congeners and the low extractable lipids of Shrimp (2.1 ± 0.4%) were probably related to these observations. This can also be partially explained by their pelagic nature and feeding pattern (Voorspoels et al., 2004). Shrimp live slightly above the seabed, resulting in less intense contact with the sediment compared to the other exclusively benthic species, Crab. Shrimp primarily feed on mysids and amphipods (Oh et al., 2001), that occupy a low trophic level. In addition, it was reported that shrimp could be able to metabolize and eliminate certain PCBs faster than some other seafood species (Goerke and Weber, 2001; Voorspoels et al., 2004). The other shellfish species (Crab) contained relatively higher PCB levels and that can be attributed to their relatively high lipid content and their living and feeding habit as well (Voorspoels et al., 2004). In particular, Crab is a typical benthic organism, also known as a scavenger that tends to feed partially on detritus or decaying organic material (such as carcasses of dead fish and other organisms), which can bear relatively high pollutant loads (Everaarts et al., 1998; Ip et al., 2005). Organisms living in close relationship with sediment such as crabs, are exposed to sedimentary PCBs via several routes, such as direct contact with sediment, respiration of interstitial water and incidental ingestion of sediment, consequently they show a contaminant load comparatively higher than in other organisms (Storelli et al., 2003). Moreover, crabs also possess gills with a relatively larger surface area which could facilitate the accumulation of PCBs into their bodies since gills can continuously transfer the pollutants from both water and suspended particles onto its surface that are subsequently distributed throughout the whole body via blood (Yang et al., 2007).

Another important factor that can play a key role in determining the contaminant burdens between the species is the depth at which organisms live. In general, higher concentrations of POPs including PCBs have been detected in seafood from deeper waters (de Brito et al., 2002; Storelli et al., 2009). This agrees well with our results, both Ilish, Sole and Crab being organisms inhabiting deeper marine or coastal areas. In general, the concentration of ∑PCBs in finfish (6.39–79.62 and 7.55–86.18 ng/g ww in winter and summer, respectively) were significantly higher ($p < 0.05$) than those in shellfish samples (5.16–37.70 and 3.82–30.06 ng/g ww in winter and summer, respectively), elucidating a higher potential of PCB bioaccumulation in finfish compared to the shellfish species. Shen et al. (2009) also reported the elevated levels of certain POPs including PCBs in fatty fish (finfish) compared to that of shellfish from six coastal provinces in China. It was noted that, on average, the ∑PCBs concentrations in catfish tissue (finfish) exceeded those found in crab tissue (shellfish) by a factor of approximately six (Howell et al., 2008).

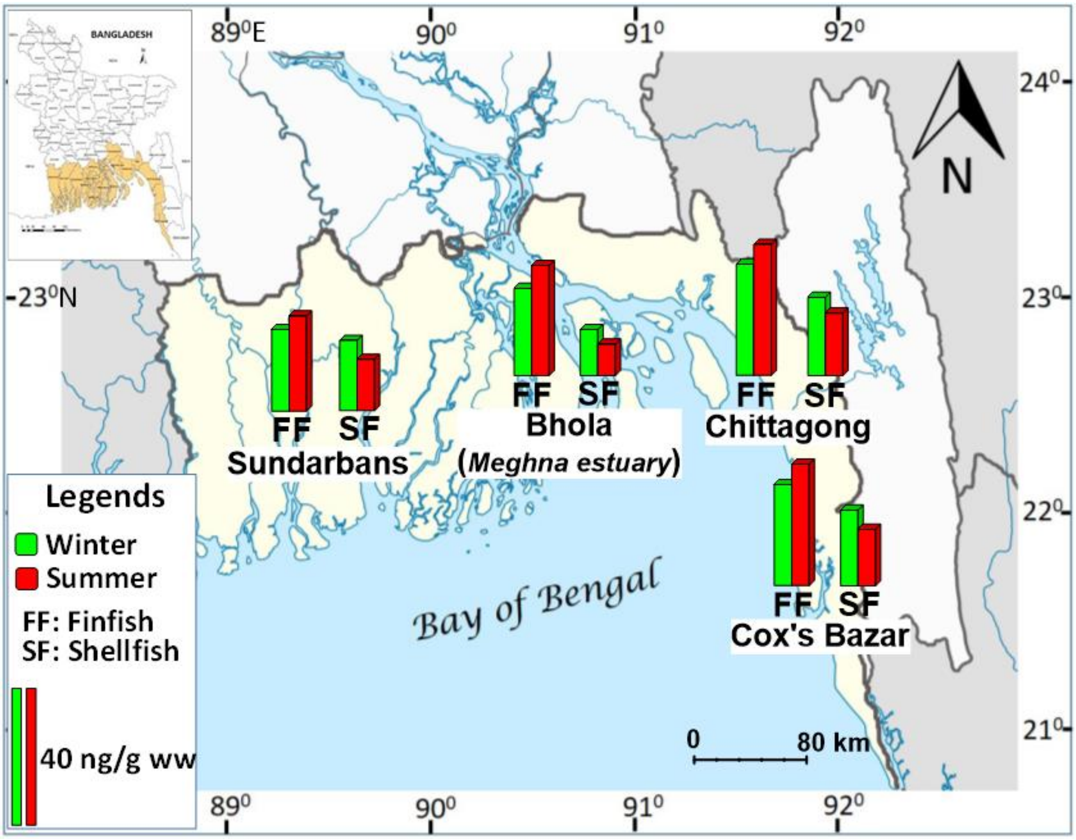

Figure 3-8. Spatial distribution of average ∑PCBs in seafood (finfish and shellfish) from the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh.

3.3.4.3 Spatial distribution

The spatial distributions of PCBs were investigated in seafood collected from the coastal area of Bangladesh and the variations in the average of ∑PCBs are presented in **Figure 3-8**. Geographically, no significant differences in ∑PCBs concentrations among the four coastal areas were observed ($p > 0.05$). However, regarding finfish, the highest mean concentration of ∑PCBs were found in samples from Chittagong in both seasons (32.03 ng/g ww in winter, 37.62 ng/g ww in summer). On the other hand, shellfish samples from Chittagong also exhibited the maximum mean levels of ∑PCBs (22.50 ng/g ww in winter, 17.95 ng/g ww in summer). In general, the mean concentration of ∑PCBs seemed to have the following trends: Chittagong > Cox's Bazar > Bhola > Sundarbans, and Chittagong > Cox's Bazar > Sundarbans > Bhola in finfish and shellfish, respectively (**Figure 3-8**). The contamination level of PCBs in seafood is significantly affected by environmental factors such as water and sediment, since the aquatic organisms are primarily exposed to the contaminants like PCBs from water and sediment. Interestingly, the spatial distribution pattern of PCBs in seafood coincided to a greater extent with that of water and sediment (data presented and interpreted in sections 3.3.1 and 3.3.2). Furthermore, relationship between levels of ∑PCBs in water-seafood and sediment-seafood was investigated and the results revealed significant correlations for both cases ($r = 0.72$, $r = 0.74$; $p < 0.05$). However, our PCB data for water and sediment samples elucidated that the PCBs in the aquatic environment of the Bangladeshi coastal area were mainly originated from point sources such as ship breaking industries, port activities, e-waste landfills and dumping, untreated or semi-treated industrial and municipal effluents, etc. along with non-negligible but minor contribution from non-point/defuse sources such as atmospheric wet/dry deposition and long range transport via ocean currents/tides. In general, the present study shown that the seafood from Chittagong, Cox's Bazar, and Sundarbans area was more contaminated with PCBs than that from Bhola (Meghna Estuary), and because of greater development, thus associating these compounds to urbanization and industrialization.

3.3.4.4 Homolog composition

The PCB congener and homolog profiles in environmental matrices can often provide valuable information on the environmental source, transport and fate processes of PCBs. **Figure 3-9** shows the PCB homologs composition (%) in the seafood samples for both seasons (winter and summer). Analysis of variance (ANOVA) revealed that, regardless of species, there was no significant difference in homolog compositions between the two seasons ($p > 0.05$). Also, the homolog composition in seafood species, regardless of source, did not differ significantly ($p > 0.05$), although the compositions were not fully identical. In general, regardless of source (sites and species) and season, the proportions of the homologs with different degree of chlorination were in the decreasing order of: medium (tetra- to hexa-CBs) > heavier (hepta- to deca-CBs) > lighter (mono- to tri-CBs), contributing 56–78%, 13–31%, and 6–20% to the ∑PCBs, respectively.

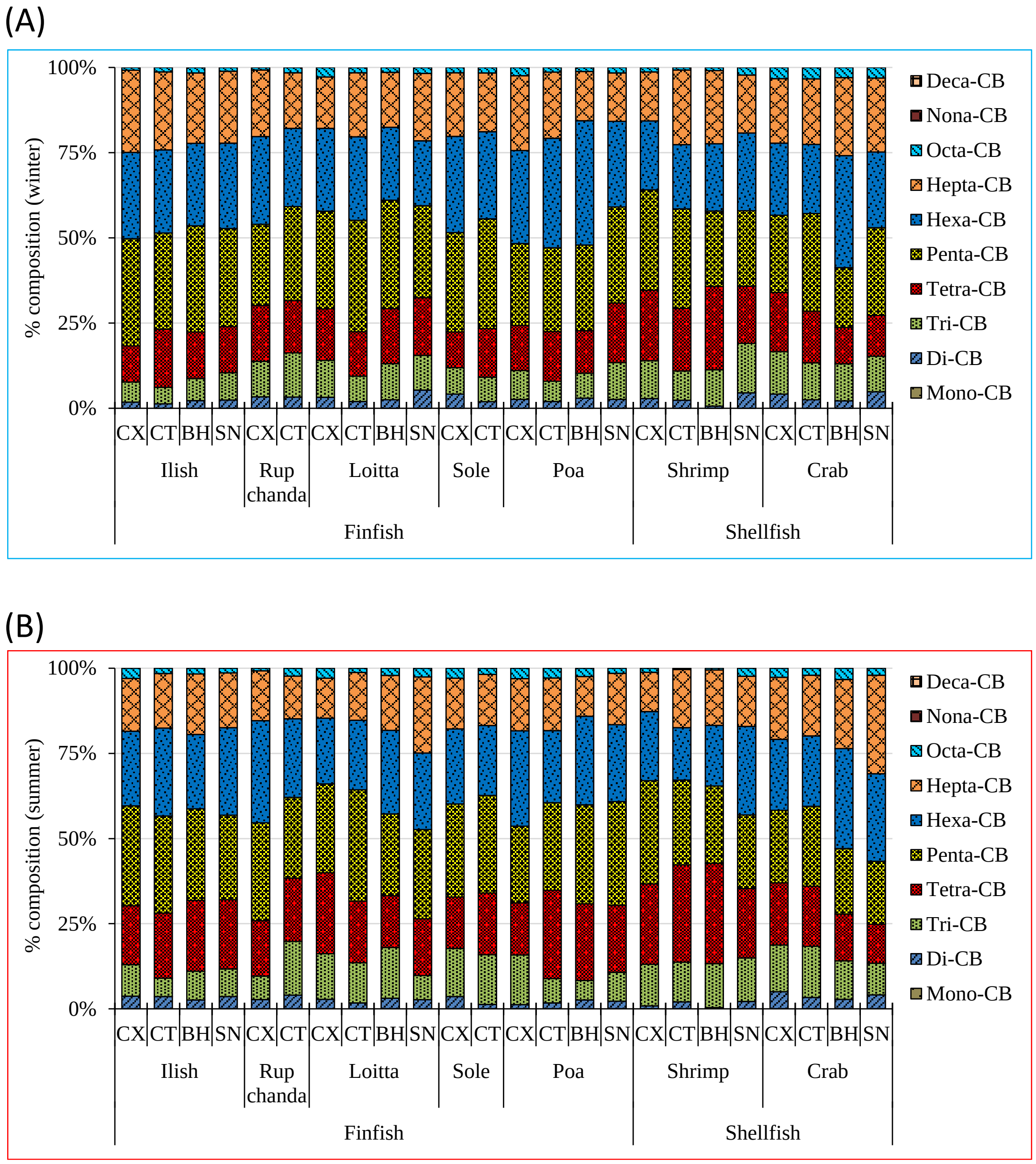

Figure 3-9. Relative contribution of PCB homologs (% composition) to the total PCB in the Bangladeshi seafood (finfish and shellfish) in winter (A) and summer (B), 2015. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans.

In general, penta- and hexa-CB was the most prevalent homologs in the seafood samples, accounting for about half of ∑PCBs (42–62% and 40–50% of ∑PCBs in winter and summer, respectively) detected in the present study, followed by hepta-CB (14–24% and 12–

29% of ∑PCBs in winter and summer, respectively) and tetra-CB (10–24% and 11–29% of ∑PCBs in winter and summer, respectively). Predominance of penta- and hexa-CB homologs were also reported for seafood from the Eastern Coast of Thailand (Jaikanlaya et al., 2009), Houston Ship Channel, Texas (Howell et al., 2008), South Korea (Moon et al., 2009), Black Sea, Bulgaria (Stancheva et al., 2017), the Mediterranean Sea (Storelli et al., 2009), Gulf of Naples, Southern Italy (Naso et al., 2005) and coastal fisheries in China (Pan et al., 2016). In general, higher chlorinated congeners were accumulated more than lower chlorinated congeners in most of the samples. This can be explained by the fact that the accumulative properties of PCB congeners increase with the number of chlorine atoms substituted to the hydrogen atoms in biphenyl rings and the resulting increase in their lipophilicity (Bernes, 1998). Furthermore, it can also be explained by that more or less fully chlorinated molecules are so large that they have greater difficulty passing through the body's cell membranes, resulting in less efficient accumulation in fish samples (Bernes, 1998).

3.3.4.5 Congener profiles and source characterization

We identified the dominant PCBs in the seafood samples both by occurrence and abundance by combining the detection rate and mean relative concentration of each congener/co-elute across samples and ranked them averagely in decreasing order. The top 20 domains were assessed for their relative contributions to ∑PCBs and these were PCB153, 180, 138, 120+110, 102+93+98+95, 187, 170, 118, 101, 52, 43+49, 99, 128, 66, 105, 151, 177, 28, 18, and 8+5, comprising up to 59–82% of ∑PCBs by species, and sum of them was highly correlated with ∑PCBs ($r = 0.98$; $p < 0.01$), well representing the ∑PCBs in the Bangladeshi seafood. Eventually, the top PCBs in this study are the most frequently reported congeners in the environment and biota (Jones, 1988; Hansen, 1998). PCB153, 180 and 138 are generally dominant in finfish and/or shellfish tissue (Miao et al., 2000; Storelli et al., 2009; Xia et al., 2012; Mohebbi-Nozar et al., 2014; Batang et al., 2016). These congeners, being refractory to metabolic attack by monooxygenases, tend to be more slowly eliminated because of their high degree of chlorination and the lack of adjacent unsubstituted H-atoms in ortho–meta and/or meta–para position on the aromatic ring (Storelli et al., 2009). Moreover, the most abundant congeners identified in the present study corresponds to diverse homolog groups (tri- to hepta-CBs), suggesting multiple sources of PCBs in this area. Sahu et al. (2009) reported that the presence of different congeners belonging to different homologs was an indication of various sources of PCBs in marine/coastal environments. Apparently, the dominant congeners in the seafood samples were also key constituents of several commercial PCB mixtures, e.g. Aroclors 1242, 1248, 1254, 1260 or equivalents (Jones, 1988; Ishikawa et al., 2007). It indicates that the environmental PCB burdens in the Bangladeshi seafood might be originated from more than one PCB Aroclors or equivalents and the most likely mixtures were Aroclors 1248, 1254 and 1260 or equivalents. Interestingly, almost similar assemblage of top PCBs were also observed the sediment samples of the present study area and we discussed in details about the possible source-occurrence relationship in the section 3.3.2 of this chapter.

Among the 12 DL-PCBs studied, PCB81, 105, 114, 118, 123, 156, 167, and 189 were detected in all of the samples for both seasons, whereas PCB77, 126, 157 and 169 were found in 67, 33, 100, 33% and 71, 33, 96, 25% of winter and summer samples, respectively. Regardless of source and season, PCB118 contributed 16–57% to ∑DL-PCB by species, followed by PCB105 (4–43%) and 156 (2–17%). Apparently, the predominance of PCB105, 118, and 156 among DL-PCBs in the present samples is a typical pattern in fish and shellfish (Bright et al., 1995; Bhavsar et al., 2007; Mezzetta et al., 2011). However, based on the abundant DL-PCB congeners, we can say that emission from industrial applications might be the major sources of DL-PCBs in the study area (Azari et al. 2007), whereas combustion processes apparently contributed to some extent what confirms the presence of PCB77 and 126 (marker PCBs for combustion processes) with a contribution of up to 1.7% of ∑PCBs.

3.3.5 Exposure assessment of dietary PCBs from seafood consumption

Seafood has been proven to be one of the major routes of human exposure to organic contaminants including PCBs (Smith and Gangolli, 2002). Storelli et al. (2003) suggested seafood as a primary vector of these pollutants/toxicants for human. Since seafood contributes a major portion in the diet for the Bangladeshi coastal population, the consumption of contaminated seafood can be a potential risk for the consumers. To comprehensively evaluate risk exposure, the estimated daily intakes (EDI) for PCBs for both the adults and children were calculated and presented in **Figure 3-10**. In this study, we evaluated two approaches: [1] EDI of ∑PCBs considering all 209 congeners where the toxic effects of non-dioxin like PCBs (NDL-PCBs) were also taken into account, and [2] EDI of ∑DL-PCBs aiming to assess health risk from dioxin like toxicities. Lower-bound approach (non-detected and congeners with <LOD were assigned to zero) was followed in both cases.

EDI of ∑PCBs and ∑DL-PCBs were calculated according to the equation (3.1) and (3.2), respectively.

$$\mathrm{EDI}_{\Sigma\mathrm{PCBs}} = \frac{(\mathrm{C}_{\mathrm{finfish}} \times \mathrm{Intake}_{\mathrm{finfish}}) + (\mathrm{C}_{\mathrm{shellfish}} \times \mathrm{Intake}_{\mathrm{shellfish}})}{\mathrm{BW}} \tag{3.1}$$

$$\mathrm{EDI}_{\Sigma\mathrm{DL\text{-}PCBs}} = \frac{(\mathrm{TEQ}_{\mathrm{finfish}} \times \mathrm{Intake}_{\mathrm{finfish}}) + (\mathrm{TEQ}_{\mathrm{shellfish}} \times \mathrm{Intake}_{\mathrm{shellfish}})}{\mathrm{BW}} \tag{3.2}$$

where $\mathrm{EDI}_{\sum\mathrm{PCBs}}$ is the estimated daily intake of ∑PCBs (ng/kg bw/day), C is the concentration of ∑PCBs in seafood (ng/g ww), $\mathrm{EDI}_{\sum\mathrm{DL\text{-}PCBs}}$ is the estimated daily intake of ∑DL-PCBs (pg TEQ/kg bw/day), TEQ is the Toxic Equivalent concentrations (pg TEQ/g ww) which were calculated by summing the concentration of each DL-PCB congener weighted by the 2005 WHO toxic equivalent factors (TEFs) (Van den Berg et al., 2006), Intake is the daily consumption of seafood (g/day), BW is the average body weight (60 kg for adults and 25 kg for children). The daily consumption data of seafood for the adults (≥18 years) and children (6–17 years) in the coastal area of Bangladesh were obtained from a questionnaire based dietary survey during our sampling campaign. The seafood consumption data and the estimated daily intake (EDI) of ∑PCBs and ∑DL-PCBs by the adults and children in the coastal area of Bangladesh are given in Table A-12 (Appendix A).

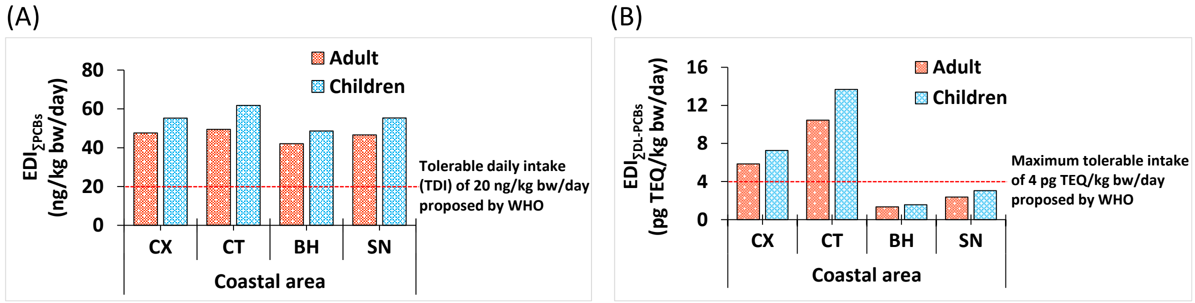

Figure 3-10. Estimated daily intake (EDI) of total PCBs (A) and DL-PCBs (B) for adults and children along with WHO guideline values (horizontal red lines).

The estimated exposure to ∑PCBs ($\mathrm{EDI}_{\sum\mathrm{PCBs}}$), regardless of sites, ranged from 42.06 to 49.46 ng/kg bw/day for adults and 48.62 to 61.82 ng/kg bw/day for children. These values were 2–3 times higher than the tolerable daily intake (TDI) of 20 ng/kg bw/day proposed by WHO as a health based guidance value for total PCB (WHO, 2003). Hence, the risk from dietary intake of ∑PCBs through seafood consumption appears to be high for both the adults and children. Likewise, the estimated daily intake of DL-PCBs ($\mathrm{EDI}_{\sum\mathrm{DL\text{-}PCBs}}$) from seafood consumption were from 1.34 to 10.45 pg TEQ/kg bw/day and 1.56 to 13.68 pg TEQ/kg bw/day for adults and children, respectively. As seen in **Figure 3-10**, EDI$_{\sum \text{DL-PCBs}}$ for both the adults and children from Chittagong and Cox's Bazar were higher than the maximum tolerable intake of 4 pg TEQ/kg bw/day recommended by WHO (1998). However, the estimated exposure to DL-PCBs for both population subgroups, frequently exceeded the most restrictive value of the tolerable daily intake (TDI; < 1 pg TEQ/kg bw/day) (WHO, 1998). In particular, the children were exposed slightly higher to the dietary PCBs in seafood than that for the adults, due to their low average body weight. Furthermore, the highest EDIs of ∑PCBs and ∑DL-PCBs were found in Chittagong area for both the adults and children (**Figure 3-10** and Table A-12, Appendix A), elucidating that the coastal residents in the Chittagong area were more prone to adverse health effects associated with dietary exposure to PCBs from seafood consumption.

In addition, it should be pointed out that since the WHO proposed TDI of 1–4 pg TEQ/kg bw/day includes not only the DL-PCBs but also the PCDDs and PCDFs, it is highly likely that the daily intake we calculated might be underestimated. However, several previous studies have proven that DL-PCBs are the most important chemicals contributing up to 70–90% to the total TEQ intake from seafood consumption (Moon and Choi, 2009; Focant et al., 2002; Shaw et al., 2006; Lee et al., 2007). Moreover, the main food type that contributed to TEQ might be fish and other seafood. For example, the contribution of finfish and shellfish was especially remarkable in two Finnish studies (> 80%) (Kiviranta et al., 2001, 2004), a Spanish study (78%) (Bascompta et al., 2002), a Japanese study (50%) (Tsutsumi et al., 2001), and a Hong Kong study (62%) (Wong et al., 2013). Fish and fish products were also identified as the main contributors in children and adults in Italy, Belgium, France and Norway (Fattore et al., 2008; Kvalem et al., 2009; Sirot et al., 2012; Cimenci et al., 2013). Taken these into consideration, we can say that our exposure assessment is relevant to examine the potential health hazard of dietary DL-PCBs from seafood consumption for the Bangladeshi coastal residents. Also, dietary exposure to other contaminants such as polybrominated diphenyl ethers (PBDEs), polycyclic aromatic hydrocarbons (PAHs), polychlorinated naphthalenes (PCNs) which could be present in these seafood samples will further elevate the daily intake value of TEQ. Thus, results of this study suggested severe human health risks via seafood consumption from both dioxin-like and non-dioxin like toxicological point of view.

3.4 Conclusion

For the first time, this study determined the full PCB profiles in surface water, sediment and commonly consumed seafood from the coastal area of Bangladesh. Although their use is presently severely restricted, the results from this study show that PCBs contamination is widespread in the coastal environment of Bangladesh and is presumably due to continuing and/or historical usage. ∑PCBs levels were at the middle of reported global range. No significant seasonal variation was reported in the levels of ∑PCBs in either medium. Spatial distribution revealed that the Chittagong, Cox's Bazar and Sundarbans areas were more contaminated with PCBs than the Meghna Estuary and because of greater development, thus associating these compounds to urbanization and industrialization. Moderately chlorinated congeners (4–6 Cl) dominated the PCB profiles. The distribution of PCB in the Bangladeshi coastal environments elucidated that the PCB contamination are mainly associated with the anthropogenic activities such as urban developments, commercial and industrial establishments. However, the profile data indicated that the prominent sources of PCBs in the Bangladeshi coastal areas were derived as related to PCB technical mixtures, pigments/dyes, and combustion. In addition, our results identified a few dominant congeners that could serve as markers of PCB contamination in Bangladesh. While the monitoring of all PCBs is vital and thus recommended, such a set of indicator congeners may be useful for selective monitoring in case of reasonable constraints on full congener approach.

The concentrations in water, sediment and tissue exceeded the guidance levels in a high percentage of samples and thus, could adversely affect the ecological environment and human health through biomagnification. Besides, the dietary intakes of total PCBs along with dioxin like PCBs through the analyzed seafood species for the adults and children of the Bangladeshi coastal area exceeded the WHO proposed health based guidelines, suggesting a significant health risk in terms of toxicological effects by both the dioxin like and non-dioxin like PCBs.

The results from this study indicated that Bangladesh still has highly unacceptable levels of PCBs in her environment and it is therefore, in doubt, whether Bangladesh will be able to totally eliminate from the environment or reduce PCBs emission to an acceptable level by 2028 in accordance with the Stockholm Convention. This study is very important to Bangladesh because it gives first-hand information about the levels and distribution of PCBs along the coastal region which serve as centers for a wide range of commercial activities, both locally and internationally. It will also contribute to the global inventory and pave the way for adequate risk assessment where necessary. This will go a long way to augment the efforts of Bangladesh's National Implementation Plan (NIP) towards the elimination of PCBs in line with the Convention.

This study also serves as a snapshot for current seafood safety and environmental health assessment and for baseline data for subsequent studies. It is also concluded that the levels of POPs including PCBs should be monitored regularly and rigorously by the appropriate government agency and that there should be mandatory public reporting. At the same time, it is imperative that resource managers, producers and consumers be educated about the hazards of these compounds and how to manage fisheries by giving emphasize on seafood safety to minimize contamination.

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Chapter 4

OCCURRENCE AND DISTRIBUTION OF PERFLUOROALKYL ACIDS (PFAAs) IN SURFACE WATER AND SEDIMENT OF A TROPICAL COASTAL AREA (BAY OF BENGAL COAST, BANGLADESH)

Publication based on this chapter:

Md. Habibullah-Al-Mamun, Md. Kawser Ahmed, Mohammad Raknuzzaman, Md. Saiful Islam, Junya Negishi, Shihori Nakamichi, Makoto Sekine, Masahiro Tokumura, Shigeki Masunaga. Occurrence and distribution of perfluoroalkyl acids (PFAAs) in surface water and sediment of a tropical coastal area (Bay of Bengal coast, Bangladesh). *Science of the Total Environment*, 2016, 571, 1089–1104.

Occurrence and distribution of perfluoroalkyl acids (PFAAs) in surface water and sediment of a tropical coastal area (Bay of Bengal coast, Bangladesh)

Abstract

This study reports the first evidence of perfluoroalkyl acids (PFAAs) in surface waters and sediments collected from the coastal area of Bangladesh. Fifteen target PFAAs, including $C_{4–14}$-PFCAs (perfluoroalkyl carboxylates) and C_4, C_6, C_8, and C_{10}-PFSAs (perfluoroalkyl sulfonates), were quantified by HPLC-MS/MS. The ∑PFAAs in surface water and sediment samples were in the range of 10.6 to 46.8 ng/L and 1.07 to 8.15 ng/g dw, respectively. PFOA in water (3.17–27.8 ng/L) and PFOS in sediment samples (0.60–1.14 ng/g dw) were found to be the most abundant PFAAs, and these concentrations were comparable to or less than most other reported values, particularly those recorded from the coastal areas of China, Japan, Korea and Spain. The majority of the monitored PFAAs did not show clear seasonal variation. The southeastern part (Cox's Bazar and Chittagong) of the Bangladeshi coastal area was more contaminated with PFAAs than the southern (Meghna Estuary) and southwestern parts (Sundarbans). Industrial and municipal wastewater effluents, ship breaking and port activities were identified as potential sources of the PFAA contamination in this region. Field-based sediment water distribution coefficients (K_D) were calculated and corrected for organic carbon content (K_{OC}), which reduced the variability between samples. The values of log K_D (1.63–2.88) and log K_{OC} (4.02–5.16) were higher than previously reported values, which may indicate that the partitioning of PFAAs in a tropical coastal ecosystem is different from other ecosystems, such as temperate and sub-tropical regions. Although a preliminary environmental hazard assessment indicated that PFOA or PFOS levels do not currently exceed the acute safety thresholds, we should keep in mind that they are bioavailable and can accumulate in the food chain. Therefore, the ubiquity of PFAAs in the coastal area of Bangladesh warrants further studies characterizing their specific sources and the potential long-term risks they present to both humans and wildlife.

4.1 Introduction

Perfluoroalkyl acids (PFAAs) are an emerging class of organic pollutants that have hydrophobic properties in addition to being lipophobic/oleophobic (Giesy and Kannan, 2002). PFAAs are highly stable, bio-accumulative and resistant to degradation in the environment. PFAAs have been used extensively for several decades in industrial and consumer applications. For example, they are used as surfactants and surface protectors in carpets, leather, paper, cooking pans and utensils, food containers, textiles, and upholstery, and they are used as performance chemicals in products such as fire-fighting foams, polishes, shampoos, pesticide formulations, grease, lubricating oils, mist suppressants, photolithography chemicals, and chemicals in electronics (Kissa, 2001, Giesy and Kannan, 2002). Their wide range of applications coupled with their unique physicochemical characteristics—high surface activity, thermal and acid resistance, weak intermolecular interactions, and water and oil repellency—make them ubiquitously distributed in various types of environmental matrices in riverine, estuarine and coastal ecosystems including water (Yamashita et al., 2008; Zushi and Masunaga, 2009; Naile et al., 2013; Lam et al., 2014; Ahrens et al., 2015), sediments (Zushi et al., 2010; Naile et al., 2013; Zhu et al., 2014; Lam et al., 2014; Ahrens et al., 2015), wildlife biota (Naile et al., 2013; Lam et al., 2014; Ahrens et al., 2015), and the human body (Zhang et al., 2010; Cho et al., 2015). They are even found in remote areas such as the Arctic (Martin et al., 2003) and the Tibetan Plateau (Shi et al., 2010). Perfluorinated carboxylic acids (PFCAs) and perfluorinated sulfonic acids (PFSAs) are the two most prevalent groups of PFAAs in the environment (Pan et al., 2014). In particular, long chain (more than seven fully fluorinated carbon atoms, e.g., perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS)) compounds are known to be bioaccumulative (Martin et al., 2003), and they have received great attention from the public and scientific community due to their potential adverse impacts on ecosystems and human health. In 2000, 3M announced a phase-out of PFOS and PFOA and their longer chain homologs (Lindstrom et al., 2011). Production of PFOS and similar perfluorooctyl products was phased out in the USA and Europe in 2000–2002 (OECD, 2002), but production continues elsewhere (Wang et al., 2009). Rapidly growing economies in developing countries place great demand on high-performance materials (e.g., PFAAs) despite the existing regulations (Zushi et al., 2012). The European Union started to ban the use of PFOS in consumer products, and PFOS, along with its precursor, perfluorooctanesulfonyl fluoride (POSF), was listed as a persistent organic pollutant (POP) in Annex B of the Stockholm Convention in 2009. That convention called for restricted production and usage worldwide (UNEP, 2009). Recently, a proposal to list perfluorooctanoic acid (PFOA), PFOA salts and PFOA-related compounds in Annexes A, B and/or C to the Stockholm Convention has been submitted by the European Union at the Eleventh Meeting in October 2015. Additionally, the United States Environmental Protection Agency (USEPA) has received and reviewed over 300 alternative chemicals for PFOA and related compounds through September 2015 (USEPA, 2015).

The main sources of PFAAs in the aqueous environment are the direct discharge of industrial or municipal wastewater and effluents, runoff from contaminated soil due to precipitation, accidental spills, the release of untreated discharges, discharge from landfills, and the use of aqueous film forming foams (AFFFs) (Paul et al., 2009; Corsolini et al., 2012; Zhu et al., 2014; Campo et al., 2015; Ahrens et al., 2015). PFAAs can be further transported directly or via rivers into the coastal environments, which have become vulnerable areas because of their intermediate position between open seas and human activities. Coastal environments can also be influenced by PFAAs carried indirectly by long-range atmospheric transport or PFAAs resulting from abiotic and/or biotic degradation of precursor chemicals (Yamashita et al., 2008).

Information on the partitioning of PFAAs between water, sediments, and/or suspended solids is crucial for understanding the transport and fate of PFAAs in aquatic environments (Zhao et al., 2012). Understanding of PFAA transport is needed to design strategies for pollution control (Zhang et al., 2012). PFAAs can accumulate in aquatic systems and are readily transported by typical hydrological or adsorption processes (Eschauzier et al., 2010). There have been few laboratory studies on the sorption processes of PFAAs that were conducted under controlled laboratory conditions (in an equilibrium state) (Higgins et al., 2005; Higgins and Luthy, 2006). The results of studies conducted under laboratory conditions and those based on field observations can be different (Hong et al., 2013) because sediment-water equilibrium is rarely reached in natural environments. The distribution and transport of PFAAs in the sub-tropical and temperate regions have well been investigated in recent decades (Ahrens et al., 2010; Zhu et al., 2014; Campo et al., 2015), but the findings have been inconsistent and show that the partitioning of PFAAs is a complex process that depends not only on the physicochemical characteristics of the compounds but also on environmental parameters (e.g., the organic carbon fraction of the sediment, water salinity and pH) (Ahrens et al., 2010; Zhao et al., 2012) that vary regionally. However, the partitioning behavior of PFAAs in tropical regions, particularly coastal environments, remains unexplored. Hence, to gain a better understanding of the fate of PFAAs in the environment, more field studies are needed.

Bangladesh is an exclusively riverine agricultural country that is undergoing rapid industrialization, urbanization and economic development. The country has a highly irregular deltaic marshy coastline of 580 kilometers, divided by many rivers and streams that enter the Bay of Bengal, which is characterized by a tropical climate. Within the coastal areas of Bangladesh, Sundarbans (UNESCO World Heritage Site) is one of the most sensitive but complex ecosystems in the world, and it suffers from environmental degradation due to rapid human settlement, the development of industrial hubs, tourism, port activities, the operation of an excessive number of mechanized boats, deforestation, and increasing agriculture and aquaculture activity. Two major rivers, Padma and Yamuna, are the key recipients of the industrial and municipal effluents from major cities in the middle and upper parts of the country (e.g., Dhaka, the most populous city and the capital) and from neighboring countries, such as India and Nepal. The rivers merge in Chandpur, forming the Meghna River, which carries waste materials through the Meghna Estuary to the Bay of Bengal (Hoq et al., 2011). The estuarine and coastal environments of the southeastern part of the Bangladeshi coast (Cox's Bazar and Chittagong area) have been severely degraded by extensive ship breaking and port activities, tourism, expanding aquaculture, and large discharges of untreated and semi-treated domestic and municipal sewage. There are also effluents containing heavy loads of organic and inorganic pollutants from many large and small local industries. Additionally, this coastal region is a major commercial transportation route, and it is a region of significant urbanization and industrialization in Bangladesh. With the population and industrial pressures increasing along the rivers, the estuarine and coastal areas of Bangladesh face threats to their coastal ecosystems from contaminants such as PFAAs, which can accumulate in the coastal or marine food chains. Local populations can be exposed to these chemicals via contaminated fish and seafood consumption.

Bangladesh's inland aquatic environments are recognized among the most polluted ecosystems in the world (due to various types of organic and inorganic pollutants; Evans et al., 2012). A very recent study on the pollution status of the coastal area of Bangladesh also revealed severe inorganic contamination in different environmental matrices (water, sediment and biota). Trace metals [e.g., Arsenic (As), Chromium (Cr), Lead (Pb), Zinc (Zn), etc.] were found in the study area (Raknuzzaman et al., 2015). However, to the best of our knowledge, there have been no studies of organic pollution in this region, particularly PFAAs. Hence, the present study was initiated. For the first time, we measured PFAAs in the coastal areas of Bangladesh. The compositional pattern and spatiotemporal distribution of PFAAs in coastal surface waters and sediments were investigated. Partition coefficients were also calculated to explore the partitioning behavior and environmental fate of these class of compounds, particularly in a tropical coastal ecosystem. A preliminary risk assessment was carried out to determine the PFAA hazards to benthic, benthopelagic and surface water-dwelling organisms of the Bay of Bengal coast in Bangladesh.

4.2 Materials and Methods

4.2.1 Study area and collection of samples

Surface water (*n=14*) and sediment (*n=14*) samples were collected from four sampling sites (Cox's Bazar, Chittagong, Meghna Estuary and Sundarbans) with fourteen different locations were investigated in the southeast and southwest coastal areas of Bangladesh (**Figure 4-1**).

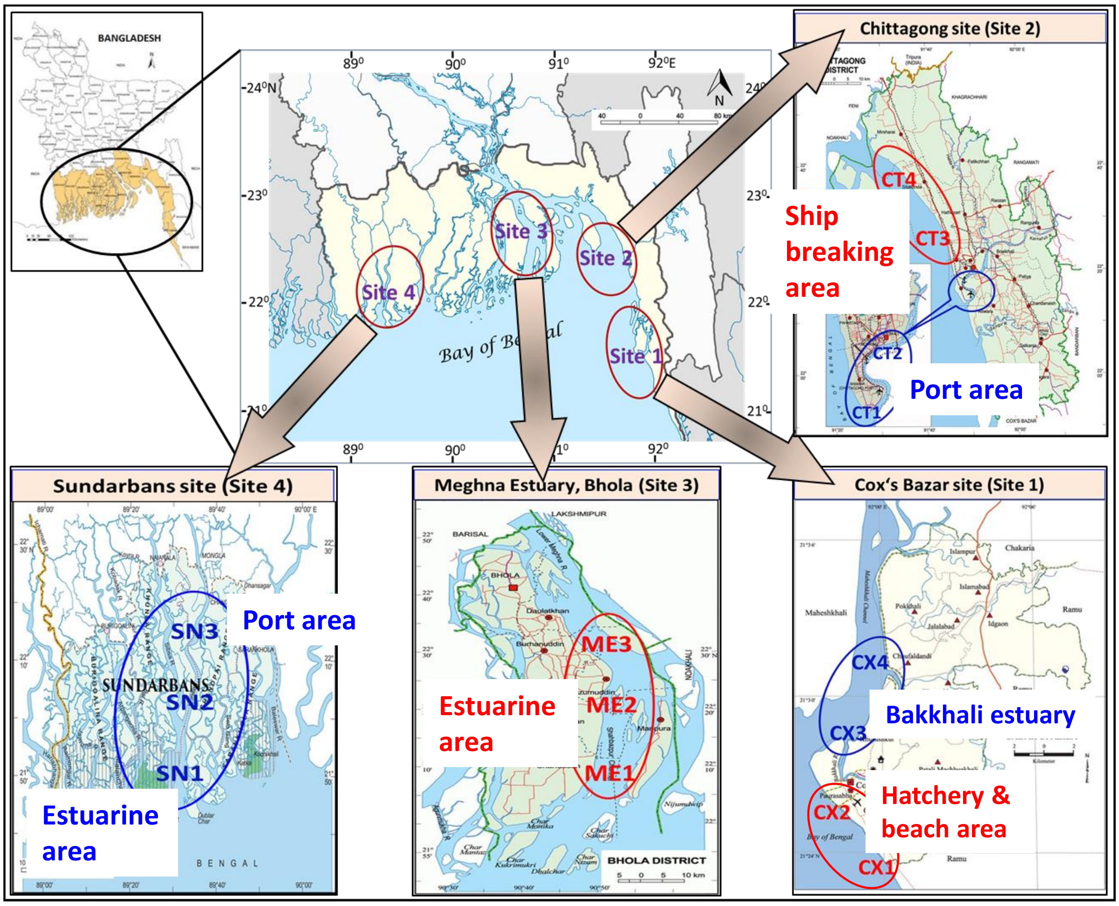

Figure 4-1. Maps showing four sampling sites with 14 sampling locations in the coastal area of Bangladesh from where surface water and sediment samples were collected. Yellow colored area in the inset map represents the coastal area of Bangladesh.

Samples were collected in January-February and August-September 2015. The sampling times represent two distinct seasons, winter (dry season) and summer (rainy season), respectively. The tides in the study area are semi-diurnal (two nearly equal high and low tides each day), and the samples were collected during low tide. For detailed description of the study area, please see section 3.2.1 (Chapter 3) and Appendix A. Three composite samples of surface water, approximately 100 mL each, were collected at each location in clean polypropylene (PP) bottles. The bottles were rinsed with deionized water, methanol, and water from the particular sampling location prior to use. Immediately after collection, the samples were filtered through 0.45 μm membranes to remove large particles and biota, and they were transferred to new PP bottles that were pre-washed with methanol and deionized water. The surficial sediment samples (top 0–5 cm) were taken using a portable Ekman grab sampler. Three composite samples with masses of approximately 200 g were collected from each sampling location and kept in polyethylene (PE) ziplock bags. Samples were transported in boxes packed with ice stored at −20 °C in a freezer upon arrival at the laboratory of the Department of Fisheries, University of Dhaka. Once there, they were treated within 48 h. Water samples were stored at −20 °C. Sediment samples were freeze-dried, ground, then kept in PP bottles and stored at −20 °C. All containers used during the process of sample collection, pretreatment, storage and transportation were carefully handled to avoid contamination. All of the processed soil samples were brought to Yokohama National University, Japan, for chemical analysis with the permission of the Yokohama Plant Protection Station.

4.2.2 Chemicals and reagents

Samples were analyzed for 15 PFAAs: perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS) and perfluorodecane sulfonate (PFDS). Native calibration standards and isotopically labeled internal standards were obtained from Wellington Laboratories (Ontario, Canada) and Kanto Chemical (Tokyo, Japan). A list of abbreviations for the standards used is presented in Table B-1 (Appendix B). All stock standards and solutions were prepared in methanol and stored in polypropylene (PP) tubes or vials at 4 °C. Oasis® weak anion exchange (WAX; 6 cc, 150 mg, 30 μm) solid phase extraction (SPE) cartridges were purchased from Waters (Milford, MA, USA). Supelclean™ ENVI-Carb™ tubes (1 mL, 100 mg) were obtained from SUPELCO® (PA, USA). Methanol and acetonitrile (>99%, HPLC grade), ammonium acetate (>98%, HPLC grade), ammonium solution and acetic acid (>99%, HPLC grade) were purchased from Wako Pure Chemical Industries (Osaka, Japan). Milli-Q (>18.2 MΩ) water was used throughout the experiment and was generated by using an ultrapure water purification system (Millipore, Billerica, MA, USA). Filter membranes (0.45 µm, 47 mm i.d.) were obtained from ADVANTEC® (Tokyo, Japan).

4.2.3 Sample pretreatment

Surface water samples were pretreated as previously described by Taniyasu et al. (2005) and Pico et al. (2012) with some modifications. Briefly, 100 mL of prefiltered sample was spiked with 800 µL of 5 ng/mL of $^{13}C_4$-PFBA, $^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_2$-PFDoDA and $^{13}C_4$-PFOS as internal standards. The samples were subsequently loaded onto an Oasis® WAX SPE cartridge that had been preconditioned with 4 mL of 0.1% ammonium hydroxide (in methanol), 4 mL of methanol, and 4 mL of Milli-Q water at a rate of one drop per second. The eluent was discarded. The cartridge was then washed with 4 mL of 50% methanol in Milli-Q water, and the eluent was again discarded. Cartridges were then centrifuged for 10 min at 3000 rpm to remove the residual water. Target compounds were eluted with 4 mL of methanol followed by 4 mL of 0.1% ammonium hydroxide (in methanol) at a rate of 1 drop per second. They were then applied to an ENVI-Carb™ tube (1 mL, 100 mg) for additional clean-up and were collected in a 15 mL PP centrifuge tube. Finally, the eluate was concentrated to 400 µL under a gentle stream of high-purity nitrogen gas at 40 °C, and it was filtered by using a disposable NORM-JECT® PP syringe (Henke-Sass Wolf GmbH, Deutschland, Germany) fitted with a disposable 0.2 µm nylon membrane filter (Merck Millipore, Cork, Ireland). Samples were stored and analyzed in PP auto-sampler vials fitted with PP septa (Agilent Tec., Santa Clara, CA, USA), as it has been shown that glass vials and PTFE septa may cause loss of the target compounds and increased contamination, respectively (Yamashita et al., 2004).

Freeze-dried sediment samples were homogenized with a silica mortar and pestle, then sieved through a 2-mm mesh sieve to remove debris and remove the coarse fraction (> 2 mm), which has low or negligible binding capacity for many dissolved contaminants (IAEA, 2003). After that, they were extracted using a methanol-based extraction method (Higgins et al., 2005; Zhao et al., 2015) with minor modifications. Briefly, 1 g of sieved sediment samples were transferred to 50 mL PP centrifuge tubes and spiked with 800 µL of 5 ng/mL of $^{13}C_4$-PFBA, $^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_2$-PFDoDA and $^{13}C_4$-PFOS as internal standards, to which 10 mL of a 1% acetic acid solution was added. Each vial was then vortexed and placed in a heated (50 °C) sonication bath for 25 min. After sonication, the tubes were centrifuged at

3000 rpm for 5 min, and the acetic acid solution was decanted into a new clean 50 mL PP tube. A mixture of 2.5 mL of a 90:10 (v/v) methanol and 1% acetic acid was then added to the original vial, and the vial was again vortex mixed and sonicated for 25 min before being centrifuged and decanted into the second tube. This process was repeated once more, and a final 10 mL acetic acid wash was performed. All extracts were combined in the second tube before being passed through the SPE cartridge. An ENVI-Carb™ tube was then used as described above in the water extraction procedure.

4.2.4 Instrumental Analysis

The PFAAs in the extract were analyzed by a high-performance liquid chromatography-tandem mass spectrometry system (HPLC-MS/MS) that consisted of an HP 1100 HPLC system (Agilent Technologies) connected to a Micromass Quattro Ultima Pt mass spectrometer (Waters Corp., Milford, MA) operated in the electrospray negative ionization mode. Separation was achieved on an Agilent Zorbax XDB-C18 column (150 mm × 2.1 mm, aperture size 5 μm) after the injection of a 10 μL aliquot of sample extract. The column was maintained at 40 °C during sample analysis. Multiple reaction monitoring (MRM) analysis was used to identify analytes. Data were processed using MassLynx™ Mass Spectrometry Software (Version 4.1, Waters, Milford, MA) for qualitative and quantitative (internal standard methodology based on peak areas) analysis. Detailed information about the instrumental measurements is listed in Table B-2 (Appendix B).

4.2.5 Quality assurance and quality control

To minimize background contamination, samples and extracts were prevented from making contact with polytetrafluoroethylene (PTFE) or other fluoropolymer materials during sample collection and preparation. During the pretreatment procedure, all containers and equipment were pre-cleaned with methanol. Quantification of all PFAAs was conducted using an internal standard calibration curve (For details, please see Table B-3, Appendix B). The limit of detection (LOD) for each analyte was defined as the smallest mass of compound resulting in a S/N ratio that was equal to or greater than 3, and the limit of quantitation (LOQ) was defined as the concentration in a standard yielding an S/N ratio of 10:1. The LOQs for target PFAAs ranged from 0.016 to 0.4 ng/L in water, and 0.006 to 0.102 ng/g dry weight (dw) in sediment. Instrumental blanks (methanol without internal standard) were analyzed every five to seven samples to monitor instrumental background. Instrumental blanks gave signal-to-noise (S/N) values of less than three. The procedural blanks (method blanks) prepared with

Milli-Q water were spiked with each of the four internal standards. Procedural blanks were analyzed with every batch of samples. Instrumental blanks and procedural blanks were below the limit of quantification (LOQ). For each matrix, analyte recovery was determined by using spiked samples to determine the accuracy of the methods. Matrix spike recovery (n = 3) was determined by spiking the target compounds into the water (10 ng/L) and sediment (10 ng/g dw) samples, followed by extraction and analysis as described in the previous section. The mean recoveries of PFAAs spiked into the water and sediment samples were 74%-114% and 87%-108%, respectively. The recovery, LOD, and LOQ for each compound are given in Table B-3 (Appendix B).

4.2.6 Statistical analysis

Statistical analyses were performed with IBM SPSS software (Version 23.0, IBM Corp., NY, USA). The significance level was set at $p = 0.05$. Before analyzing, concentration values lower than the LODs were set to LOD/2 (Succop et al., 2004). A statistical distribution test called *P–P* plots was carried out to test for normality. Descriptive statistics (range: minimum-maximum, mean, and median) was calculated by using the Microsoft® Excel® 2016 MSO Windows program. Spearman rank correlation analysis was used to examine possible correlations between various PFAAs in the samples. A one-way ANOVA was performed to determine the significant differences between the concentrations of PFAAs detected in the coastal area of Bangladesh and to examine seasonal variations. The spatial distributions of PFAAs were analyzed using MapViewer™ software (Version 8, Golden Software Inc., CO, USA).

4.3 Results and Discussion

4.3.1 Occurrence, concentrations and composition of PFAAs in surface water

All of the fifteen target PFAAs were detectable in surface waters of the coastal areas of Bangladesh. The concentrations of individual PFAAs are shown in **Figure 4-2** and Table B-5 (Appendix B). The total concentrations of the PFAAs (sum of 15 PFAAs, ∑PFAAs) in the water phase ranged from 10.6 to 45.2 ng/L in winter, and from 11.5 to 46.8 ng/L in summer (**Table 4-1**). The mean ∑PFCAs (mean from sum of PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, and PFTeDA) of all the samples was 18.7 ng/L (8.23–44.3 ng/L) and 21.2 ng/L (10.8–39.3 ng/L), whereas the mean ∑PFSAs (mean from sum of PFBS, PFHxS, PFOS, and PFDS) was 2.80 ng/L (0.73–6.75 ng/L) in winter and 2.77 ng/L (0.70–9.99 ng/L) in summer. Among the 15 target PFAAs, PFPeA, PFHxA, PFOA, PFNA and PFDA were the most frequently detected compounds; they were found in 100% of samples in both seasons. PFOS was detected in 100% and 93% of samples in winter and summer, respectively. The detection frequencies for the rest of the PFAAs were in the range of 50% to 93%. In the surface water of the study area, PFOA was the most abundant PFAA compound. It had a relatively high concentration (3.17–27.8 ng/L) and 100% detection frequency (**Table 4-1**). The profiles of the relative concentrations of the 15 individual PFAAs in surface water for winter and summer are displayed in Fig. S1A. In the present study, PFOA contributed 52–54% of the total PFAAs, but lower contributions of PFOS were measured. PFOS accounted for 5–7% of all analytes. Similarly, PFOS, PFPeA, PFDA and PFBS contributed 7–10%, 4–8% and 4–7% of the total target analytes in the study area (Figure B-1A, Appendix B). The other PFAAs, including PFBA, PFHpA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFHxS and PFDS, contributed lesser percentages of the total PFAAs and were found at lower concentrations (**Table 4-1**, **Figure 4-2**). In general, the concentration of PFOA was approximately 8–10 times higher than that of PFOS in water samples, which can be explained by its higher water solubility, lower bioaccumulation potential and lower sorption potential in sediment compared to PFOS (Brooke et al., 2004). However, PFOA concentrations are usually higher than PFOS in open waters due to different amounts of discharge into the aqueous environment. The discharges also come from different sources, undergo different degradation processes, and have different physicochemical characteristics (Paul et al., 2009). Previous studies conducted elsewhere found an opposite trend: PFOS levels in surface waters were higher than PFOA (Labadie and Chevreuil, 2011; Thompson et al., 2011; Naile et al., 2013; Campo et al., 2015; Zhao et al., 2015), which might be attributed to the presence of point sources that contribute more PFOS than PFOA in these study areas.

Although PFOS and PFOA were considered the most industrially utilized PFAAs (Zushi et al., 2011), the low levels of PFOS found in the present study might indicate a decrease in the use and production of PFOS after the introduction of regulations with the Stockholm Convention (UNEP, 2009). However, moderate but significant levels with higher detection frequency of PFNA (0.57–2.08 and 0.46–4.17 ng/L in winter and summer, respectively), PFDA (0.12–5.72 and <LOQ–5.32 ng/L in winter and summer, respectively),

and PFBS (<LOD–3.67 and <LOD–1.5 ng/L in winter and summer, respectively) might be an indication that alternatives to PFOA and PFOS are being used in Bangladesh.

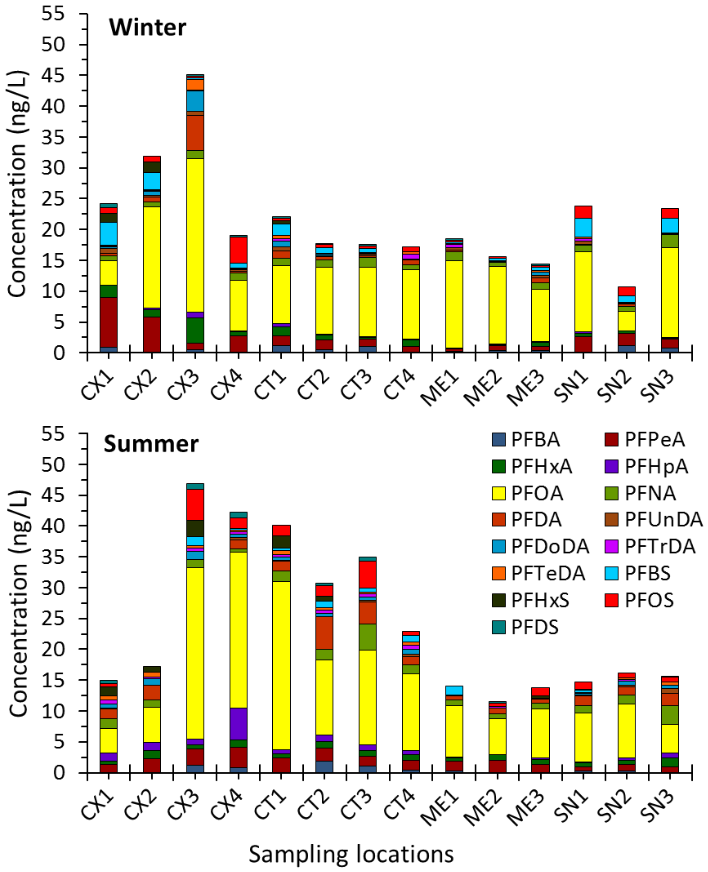

Figure 4-2. Concentrations of PFAAs in the surface water of the Bangladeshi coastal area in two seasons.

Table 4-2 compares the results obtained in the present study with those of previous studies conducted in other estuarine and coastal areas around the world, covering tropical, sub-tropical and temperate regions. In general, concentrations measured in this study were comparable with those found in the surface waters of the Yangtze Estuary of China (Pan et al., 2014) and the southern coast of India (Yeung et al., 2009) and were lower than in coastal water samples from the North Bohai Sea (Chen et al., 2011), Tokyo Bay Basin of Japan (Zushi et al., 2011), the west coast of Korea (Naile et al., 2013) and the Cape Fear River Basin in the USA (Nakayama et al., 2007).

Table 4-1. Range, mean, median concentrations and detection frequencies of PFAAs in water (ng/L) and sediment (ng/g dw) in two seasons (winter and summer) in the coastal area of Bangladesh.

Matric	Season		PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFAAs
Water	Winter	Range	<LOD[a]–1.2	0.47–8.07	0.16–4.07	<LOD–1.03	3.17–24.8	0.57–2.08	0.12–5.72	<LOD –0.74	<LOD –3.3	<LOD –0.69	<LOD –1.66	<LOD –3.67	<LOD –1.81	0.16–4.26	<LOD –0.61	10.6−45.2
		Mean	0.48	2.21	0.98	0.22	11.6	1.14	0.83	0.36	0.45	0.20	0.28	1.35	0.29	1.02	0.18	21.5
		Median	0.39	1.46	0.67	0.11	11.3	1.13	0.35	0.35	0.11	0.10	0.13	0.85	0.03	0.63	0.17	18.7
		D.F.[c]	86	100	100	86	100	100	100	93	79	86	93	93	50	100	64	
	Summer	Range	<LOD –1.82	0.69–3.24	0.49–1.48	<LOQ[b]–5.12	3.98–27.8	0.46–4.17	<LOQ –5.32	<LOD –0.70	<LOD –1.34	<LOD –0.62	<LOD –0.85	<LOD –1.5	<LOD –2.59	<LOD –5.10	<LOD –0.86	11.5−46.8
		Mean	0.45	1.76	0.87	0.99	12.4	1.55	1.74	0.15	0.57	0.33	0.38	0.51	0.60	1.45	0.26	24.0
		Median	0.24	1.62	0.85	0.74	8.50	1.32	1.54	0.11	0.56	0.37	0.36	0.35	0.09	1.22	0.05	16.7
		D.F.	64	100	100	93	100	100	100	71	93	79	86	71	64	93	71	
Sediment	Winter	Range	<LOQ –1.37	0.09–1.01	0.07–0.51	<LOQ –0.38	0.14–1.49	0.04–0.53	0.04–0.99	<LOD–0.55	<LOD –0.49	<LOD –0.12	<LOD –0.23	<LOD –0.87	<LOD –0.96	0.44–3.56	<LOD –0.99	2.48−8.15
		Mean	0.48	0.30	0.17	0.11	0.82	0.20	0.18	0.25	0.13	0.04	0.09	0.53	0.20	1.14	0.25	4.89
		Median	0.38	0.21	0.14	0.07	0.75	0.15	0.12	0.25	0.12	0.02	0.10	0.52	0.06	0.93	0.04	4.41
		D.F.	93	100	100	93	100	100	93	79	93	86	79	93	64	100	50	
	Summer	Range	<LOD –0.55	0.06–0.17	0.08–0.17	<LOD –0.51	0.09–0.69	0.05–0.21	<LOD –0.16	<LOD –0.47	0.01–0.13	<LOD –0.09	<LOD –0.11	<LOD –0.35	<LOD –0.11	0.30–1.64	<LOD –0.29	1.07−3.81
		Mean	0.18	0.12	0.11	0.07	0.30	0.11	0.07	0.07	0.07	0.05	0.03	0.15	0.03	0.60	0.06	2.02
		Median	0.16	0.12	0.11	0.03	0.27	0.11	0.06	0.02	0.07	0.04	0.03	0.17	0.01	0.51	0.51	1.95
		D.F.	71	100	100	86	100	100	93	79	93	86	79	79	43	100	43	

[a] Limit of detection; [b] Limit of quantification; [c] Detection frequency (%); *n=14* for each matrix of each season; While calculating mean and median, values for <LOD and <LOQ were assigned to LOD/√2 and LOQ/2, respectively (Succop et al., 2004). Please refer to Table B-3 (Appendix B) for the LOD and LOQ values of investigated PFAAs.

In particular, PFOA and PFOS are the two most discussed PFAAs that have been measured in aquatic environments around the world. In the present study, the concentrations of PFOA (3.17–27.8 ng/L) were comparable to or less than those detected in the coastal waters of China (Wang et al., 2012), Korea (Naile et al., 2013), the USA (Nakayama et al., 2007), and India (Yeung et al., 2009), and they were much less than those measured in the Tokyo Bay Basin of Japan (Zushi et al., 2011), the Mediterranean area of Spain (Campo et al., 2015), and the major river basins in Vietnam (Lam et al., 2016). The PFOA concentrations were higher than those measured in the coastal area of Hong Kong (Yamashita et al., 2005), the Cantabrian Sea of Spain (Gómez et al., 2011), the German coast (Ahrens et al., 2010), the Orge River basin in France (Labadie and Chevreuil, 2011), and Sydney Harbor in Australia (Thompson et al., 2011). In the present study, the concentration of PFOS (<LOD–5.10 ng/g dw) in Bangladeshi coastal surface water was comparable to or less than the concentrations measured in surface waters from the coastal area of China (Wang et al., 2012), Tokyo Bay of Japan (Zushi et al., 2011), the west coast of Korea (Naile et al., 2013), the major river basins in Vietnam (Lam et al., 2016), the Cantabrian Sea of Spain (Gómez et al., 2011), the Orge River basin of France (Labadie and Chevreuil, 2011) and Sydney Harbor in Australia (Thompson et al., 2011), but it was higher than those reported from the coastal area of Hong Kong (Yamashita et al., 2005), the German coast (Ahrens et al., 2010) and the southern coast of India (Yeung et al., 2009). Moreover, the comparison results (**Table 4-2**) also indicate that there are differences in the contamination level and compositional pattern of PFAAs other than PFOA and PFOS, the two most industrially utilized PFAAs, between countries and/or areas worldwide. For example, PFNA (<LOQ–3703.64 ng/L) was the most prevalent PFAA in the Tokyo Bay basin, Japan (Zushi et al., 2011) which differs from many earlier reports worldwide by several order of magnitudes. The high level of PFNA attributed to its selective industrial production in Japan might be an indication of shifting in PFAAs production and usage because of the introduction of restrictions. Similar results for many other PFAAs regarding the degree of their usage and applications in the industrial and/or consumer products might be obtained from the global PFAAs survey. These findings also revealed that the existence of manufacturing companies for particular PFAAs might be responsible for the environmental emission for a particular PFAA in a particular area. Thus, the different patterns of concentration of PFAAs analyzed in the present study, combined with the comparisons with other previous studies, might suggest a site-specific PFAAs sources in Bangladeshi coastal waters.

Table 4-2. Concentrations of PFAAs in surface water (ng/L) and sediment (ng/g dw) in samples from the present study and around the world.

Location	Matric	Year	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS
Coastal area, China(1)	Water	2008	–	–	–	<LOD–2.6	3.0–12	<LOD–4.9	<LOD–3.8	<LOD	<LOD	–	–	–	–	0.1–10	–
	Sediment		–	–	–	<LOD–0.56	<LOD–1.5	<LOD–1.0	0.96–2.3	<LOD–1.2	0.27–0.81	–	–	–	–	<LOD–4.3	–
North Bohai Sea, China(2)	Water	2008	–	<LOD–2.3	–	<LOD–11	<LOD–82	<LOD–8	<LOD–10	–	–	–	–	–	–	<LOD–9.2	–
	Sediment		–	–	<LOD–0.4	–	<LOD–0.5	–	<LOD–1	<LOD–0.4	–	–	–	–	–	<LOD–1.97	–
Yangtze river estuary, China(3)	Water	2013	0.35−8.38	<LOD−2.59	0.11−22.7	<LOD−2.61	0.52−18.0	<LOD−0.86	<LOD–0.33	<LOD	–	–	–	<LOD−41.9	<LOD−4.50	<LOD−3.39	<LOD–0.21
	Sediment		<LOD	<LOD–0.04	<LOD–0.32	<LOD	0.03–0.72	<LOD–0.06	<LOD–0.06	<LOD–0.10	–	–	–	<LOD–0.20	<LOD	<LOD–0.59	<LOD
Coastal area, Hong Kong(4)	Water	2005	–	–	–	–	0.67–5.45	0.022–0.21	–	–	–	–	–	–	<0.005–0.31	0.07–2.6	–
Pearl River Delta, Hong Kong(5)	Sediment	2010	0.09–0.4	–	<LOD	–	<LOD–2.25	<LOD–0.05	<LOD–0.07	<LOD–0.06	<LOD–0.07	–	–	–	<LOD	0.08–0.36	–
Tokyo Bay basin, Japan(6)	Water	2009	–	–	<LOQ–423.3	<LOQ–460.81	<LOQ–3703.64	<LOQ–1121.56	<LOQ–122.39	<LOQ–29.45	<LOQ–19.12	<LOQ–4.31	<LOQ–1.6	–	<LOQ–22.14	<LOQ–99.39	<LOQ
Tokyo Bay, Japan(7)	Sediment	2004	–	–	<LOQ–0.09	<LOQ–0.08	0.12–0.45	0.15–0.56	<LOQ–0.19	<LOQ–0.55	<LOQ–0.32	<LOQ–1.22	–	–	<LOQ	0.39−1.79	–
West coast, Korea(8)	Water	2009	–	–	–	<1.0–110	0.54–31	<0.20–5.9	<0.20–9.3	0.22–1.3	–	–	–	<0.2–16	<0.2–8.7	0.35–47	–
	Sediment		–	–	–	–	<0.2–2.4	–	–	–	–	–	–	–	–	<0.2–5.8	–
Mediterranean area, Spain(9)	Water	2010	0.07–111	0.08–2.50	0.63–25.2	0.63–30.9	0.07–146	0.77–52.4	0.07–4.25	0.09–0.09	–	0.03–9.75	–	0.41–4.10	14.2–33.2	0.01–2710	–
	Sediment		0.61–12.9	0.26–1.06	–	0.38–0.38	0.36–1.52	3.87–3.87	0.09–0.55	0.12–0.41	0.11–0.22	0.19–0.19	–	0.91–3.53	0.03–0.29	0.15–11.4	–
Cantabrian Sea, Spain(10)	Water	2009	–	–	–	–	0.05–0.31	<LOD–0.20	–	–	–	–	–	<LOD–1.25	<LOD–0.19	<LOD–6.57	–
	Sediment		–	–	–	–	<LOD–0.03	<LOD–0.08	–	–	–	–	–	<LOD–0.01	<LOD	<LOD–0.02	–
German coast, Germany(11)	Water	2007	<LOD–4.73	<LOD–0.38	<LOD–1.18	<LOD–0.58	0.08–3.02	0.05–0.37	<LOD–0.17	<LOD				0.01–6.51	<LOD–0.28	<LOD–2.26	
German Bight, Germany(12)	Sediment	2011	–	0.003–0.007	0.002–0.019	0.001–0.031	0.007–0.43	0.003–0.43	0.009–0.61	0.007–0.21	0.003–0.04	0.008–0.024	0.003–0.009	0.006–0.17	0.002–0.30	0.023–5.4	0.014–0.014
Orge River basin, France(13)	Water	2010	–	8.9	13.3	4.5	9.4	1.3	1.1	–	–	–	–	4.4	–	17.4	<0.02
	Sediment		–	<LOD	0.06	0.03	<0.07	0.05	0.3	–	–	–	–	<0.05	–	4.3	0.12
Sydney Harbour, Australia(14)	Water	2009	–	–	2.8–3.2	1.4–2.0	4.2–6.4	0.60–2.0	0.80–1.6	0.20–0.30	0.20–0.30	<LOD	<LOD	1.2–1.5	2.7–4.3	7.5–21	<LOD
	Sediment		–	–	<LOD	<LOQ	0–0.16	<0.10–0.11	0–0.81	0.10–0.61	0–1.1	<0.10–0.27	<LOD	<LOD	<0.10–0.10	0.80–6.2	<0.10–0.20
Cape Fear River Basin, NC, USA(15)	Water	2006	–	–	<LOQ−7.38	<LOQ−38.7	<LOQ−43.4	<LOQ−33.6	<LOQ−22.1	<LOQ−10.4	<LOQ−2.17	–	–	<LOQ−2.58	<LOQ−7.29	<LOQ−31.2	–
Charleston estuary, SC, USA(16)	Sediment	2012	–	–	<0.02−0.189	<0.02−0.234	0.02−2.515	0.03−1.938	0.055−4.762	<0.02−2.445	<0.02−0.303	–	–	<0.02−0.271	<0.02−0.132	0.092−7.369	<0.02−0.136
Major river basins, Vietnam(17)	Water	2013-2015			<0.07–4.26	<0.45–7.81	<0.13–53.5	<0.06–4.81	<0.03–1.37	<0.03–0.33	<0.03–0.03	<0.05–0.06	<0.06	<0.30–8.28	<0.07–5.98	<0.03–40.2	<0.11
	Sediment				<0.16	<0.23	<0.20	<0.10	<0.06–0.17	<0.06–0.24	<0.05–0.16	<0.03–0.16	<0.13	<0.34–0.57	<0.04–16.4	<0.08–6.72	<0.30–0.85
Southern coast, India(18)	Water	2008	–	–	–	–	0.05–23	–	–	–	–	–	–	–	–	0.05–3.9	–
Sundarbans wetlands, India(19)	Sediment	2011	–	–	–	–	<0.5–14.09	–	–	–	–	–	–	–	–	<0.5	–
Coastal area, Bangladesh (This study)	Water	2015	<LOD–1.82	0.47–8.07	0.16–4.07	<LOD–5.12	3.17–27.83	0.46–4.17	<LOQ–5.72	<LOD–0.78	<LOD–3.3	<LOD–0.69	<LOD–1.66	<LOD–3.67	<LOD–2.59	<LOD–5.10	<LOD–0.86
	Sediment		<LOD–1.37	0.06–1.01	0.07–0.51	<LOD–0.51	0.09–1.49	0.04–0.53	<LOD–0.99	<LOD–0.55	<LOD–0.49	<LOD–0.12	<LOD–0.23	<LOD–0.87	<LOD–0.96	0.30–3.56	<LOD–0.99

(1) Wang et al., 2012; (2) Chen et al., 2011; (3) Pan et al., 2014; (4) Yamashita et al., 2005; (5) Zhao et al., 2014; (6) Zushi et al., 2011; (7) Zushi et al., 2010; (8) Naile et al., 2013; (9) Campo et al., 2015; (10) Gómez et al., 2011; (11) Ahrens et al., 2010; (12) Zhao et al., 2015; (13) Labadie and Chevreuil, 2011; (14) Thompson et al., 2011; (15) Nakayama et al., 2007; (16) White et al., 2015; (17) Lam et al., 2016; (18) Yeung et al., 2009; (19) Corsolini et al., 2012; LOD: limit of detection; LOQ: limit of quantification; '–': not analyzed;

4.3.2 Occurrence, concentrations and composition of PFAAs in sediment

The surface sediment samples collected from the coastal areas of Bangladesh showed detectable concentrations of all fifteen target PFAAs. The concentrations of the individual PFAAs are shown in **Figure 4-3** and Table B-6 (Appendix B). The total concentrations of the PFAAs in sediment ranged from 2.48 to 8.15 ng/g dw in winter, and from 1.07 to 3.81 ng/g dw in summer (**Table 4-1**). The mean ∑PFCAs of all the analyzed sediment samples was 2.77 ng/g dw (0.01–4.36 ng/g dw) in winter and 1.19 ng/g dw (0.64–1.77 ng/g dw) in summer, whereas the mean ∑PFSAs was 2.12 ng/g dw (0.86–4.31 ng/g dw) in winter and 0.84 ng/g dw (0.43–2.04 ng/g dw) in summer. The most frequently detected compounds were PFPeA, PFHxA, PFOA, PFNA, and PFOS, which had detection frequencies of 100% in both seasons, and their mean concentrations were 0.12–0.30 ng/g dw, 0.11–0.17 ng/g dw, 0.30–0.82 ng/g dw, 0.11–0.20 ng/g dw and 0.60–1.14 ng/g dw, respectively. The other, less frequently detected compounds were PFBA, PFHpA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, and PFDS, which had detection frequencies of 43–93%. The different occurrence patterns for the PFAAs may be related to their usages and physicochemical properties (Li et al., 2011). The relative compositional profiles of the 15 target PFAAs in surface sediment measured in both winter and summer are displayed in Figure B-1B (Appendix B). Our study revealed that PFOS was the most abundant PFAA and was detected in all sediment samples, and it contributed 23–30% of the total PFAAs in the sediments. Smaller contributions came from PFOA, between 15% and 17% of the total detectable analytes, followed by PFBS (7–11%) and PFBA (9–10%). The levels of PFOS were between 0.30 and 3.56 ng/g dw, and the levels of PFOA ranged from 0.09 to 1.49 ng/g dw. For PFBS and PFBA, the concentrations in sediments ranged from below the LOD to 0.87 ng/g dw and 1.37 ng/g dw, respectively (**Table 4-1**, **Figure 4-3**). In general, the concentration of PFOS was approximately 2–3 times higher than that of PFOA in sediment samples, which can be explained by its higher sorption potential in sediment (Higgins and Luthy, 2006). Furthermore, the large percentage of PFOS found in sediment in our study compared to water suggests a higher constant of distribution for PFOS in sediment (Pico et al., 2012). In contrast to our study, some previous studies showed opposite findings: PFOA was found to be higher in sediment samples than PFOS (Pan et al., 2014; Zhao et al., 2014; Corsolini et al., 2012). This may be due to the different physiochemical properties of the sediments and waters of different regions, or due to the different magnitudes of PFAA inputs and different geomorphological characteristics of the study areas.

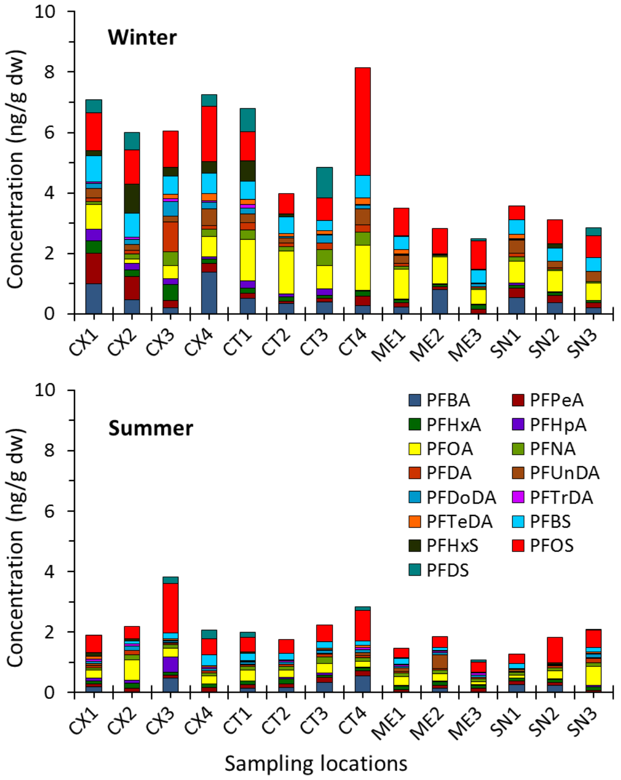

Figure 4-3. Concentrations of PFAAs in sediments from the Bangladeshi coastal area in two seasons.

In **Table 4-2**, the results of the present study are compared with previous reports. In particular, PFOS and PFOA were found to be the most abundant PFAAs in the surface sediments. The level of PFOS in the coastal sediment of Bangladesh (0.30–3.56 ng/g dw) was comparable to or less than the concentrations measured in the coastal area of China (<LOD–4.3 ng/g dw) (Wang et al., 2012), the west coast of Korea (<0.2–5.8 ng/g dw) (Naile et al., 2013), the Mediterranean area of Spain (0.15–11.4 ng/g dw) (Campo et al., 2015), the German Bight (0.023–5.4 ng/g dw) (Zhao et al., 2015), the Orge River basin of France (mean of 4.3 ng/g dw) (Labadie and Chevreuil, 2011), Sydney Harbor in Australia (0.80–6.2 ng/g dw) (Thompson et al., 2011), the Charleston Estuary in the USA (0.092−7.369 ng/g dw) (White et al., 2015), and the major river basins in Vietnam (<0.08−6.72 ng/g dw) (Lam et al., 2016). The PFOS level was higher than in the surface sediments of the Pearl River Delta in Hong Kong (0.08–0.36 ng/g dw) (Zhao et al., 2014), Tokyo Bay, Japan (<LOQ−1.66 ng/g dw) (Zushi et al., 2010), and the Cantabrian Sea of Spain (<LOD–0.02 ng/g dw) (Gómez et al., 2011). In the present study, the concentrations of PFOA (0.09–1.49 ng/g dw) in sediment samples were comparable to or less than those measured in the coastal area of China (Wang et al., 2012), the Pearl River Delta, Hong Kong (Zhao et al., 2014), the west coast of Korea

(Naile et al., 2013), the Mediterranean area of Spain (Campo et al., 2015), the Charleston Estuary in the USA (White et al., 2015) and Sundarbans wetlands in India (Corsolini et al., 2012), but the concentrations were higher than those reported from Tokyo Bay of Japan (Zushi et al., 2010), the Cantabrian Sea of Spain (Gómez et al., 2011), the German Bight (Zhao et al., 2015), and Sydney Harbor in Australia (Thompson et al., 2011) (**Table 4-2**).

In general, the levels of PFOS and PFOA in Bangladeshi coastal waters and sediments were lower than, comparable to or sometimes slightly higher than those reported in other developing and developed countries around the world, particularly China, Vietnam, Korea, Japan, Spain, France, Germany, Australia, and the USA. The differences may be due to the different amounts of PFOA and/or PFOS used in different countries. To draw a final conclusion, however, the specific sources of PFAAs in the Bangladeshi coastal environment should be examined because there is currently no information on the usage, production, import, and volumes of PFAAs in Bangladesh.

4.3.3 Seasonal and spatial variations and distributions of PFAAs in the coastal areas of Bangladesh

The seasonal variations and spatial distributions of PFAAs were investigated in surface water and sediment samples collected from the coastal area of Bangladesh, and the findings are presented in **Figure 4-4**. After screening the PFAAs in the Bangladeshi coastal area, no clear seasonal variation was found for their levels in surface water. In general, there were no obvious significant differences between the winter and summer for the target PFAAs in surface water ($p > 0.05$). However, some PFAAs, such as PFHpA, PFUnDA, and PFBS, had obvious seasonal variations ($p < 0.05$) (Table B-7, Appendix B). The concentration of PFHpA was higher in summer (<LOQ–5.12 ng/L) than in winter (<LOD–1.03 ng/L), whereas PFUnDA and PFBS were found to be higher in winter (<LOD–0.74 ng/L and <LOD–3.67 ng/L, respectively) than in summer (<LOD–0.70 ng/L and <LOD–1.5 ng/L, respectively). Simcik and Dorweiler (2005) found that concentrations could be higher due to atmospheric deposition of PFHpA in surface waters. Therefore, atmospheric deposition could be a source of PFHpA and other PFCAs in the surface water during times of heavy rainfall, such as the summer season. However, in the present study, the seasonal variations of these particular PFAAs (PFHpA, PFUnDA, and PFBS) in water might be due to their specific seasonal usage or applications in certain industries (e.g., the textile, paper, and leather industries) or the seasonal use of consumer products, although the exact reasons for this phenomenon are still

unknown. PFAA concentrations in water could be affected by many factors, such as water flow, water quality properties, weather (particularly rainfall), and PFAA usage and discharge in aquatic systems. In this study, in general, the mean concentration of total PFAAs (sum of 15 target PFAAs) in winter (21.5 ng/L) was slightly lower than in summer (24.0 ng/L). On the other hand, seven out of 15 target PFAAs, namely PFBA, PFPeA, PFOA, PFUnDA, PFBS, PFHxS, and PFOS, showed significant differences ($p < 0.05$) in their concentrations between winter and summer for the surface sediment (Table S8). In general, the mean concentration of total PFAAs in winter (4.89 ng/g dw) was more than two times higher than in summer (2.02 ng/g dw). This significant seasonal variation ($p < 0.05$) in PFAA concentrations in sediment samples can be explained by the following phenomenon: in summer, most of the coastal area of Bangladesh (> 90%) experiences heavy flooding and massive soil erosion occurs along the river banks due to heavy rainfall. Consequently, a huge amount of fresh soil (which may be less contaminated by PFAAs) is deposited as sediment in the aquatic environment by floodwaters and eroded soil in the estuarine and coastal areas. In winter, no such environmental disturbances occur, so the winter sediment samples showed significantly higher concentrations of PFAAs in the study area. In fact, environmental pollution monitoring with sediment samples can be considered as an indicator of accumulation over time. Core sediment analysis can be performed, and the environmental quality status at a given time can be determined by water monitoring. This is generally applicable for the complex and dynamic ecosystems of the coastal area of Bangladesh. Therefore, studies considering more sampling sites, diverse sampling times and sedimentary core analysis would be recommended to explore an in-depth scenario of PFAA contamination and to determine the fate and distribution of PFAAs in a complex ecosystem with diverse ecological features and resources.

Figure 4-4A shows the spatial distribution of PFAAs in surface water samples for both seasons. Concentrations of total PFAAs (∑PFAAs) were higher in the coastal waters at sampling locations CX1–CX4 (average of 30.1 and 30.3 ng/L in winter and summer, respectively), CT1–CT4 (average of 18.6 and 32.2 ng/L in winter and summer, respectively) and SN1–SN3 (average of 19.3 and 15.5 ng/L in winter and summer, respectively) in comparison to sampling locations ME1–ME3 (average of 16.1 and 13.1 ng/L in winter and summer, respectively), which indicates that the industrialized regions (Chittagong and Cox’s Bazar) in this area are potential sources of PFAAs and that economic development seems to be associated with the amount of PFAA emissions. Additionally, the lower concentrations of

PFAAs found in water taken from the Meghna Estuary (ME1−ME3) could result from the mixing or dilution effects of huge volumes of less contaminated inland freshwater (2.73–4.17 trillion m^3 per annum) with seawater.

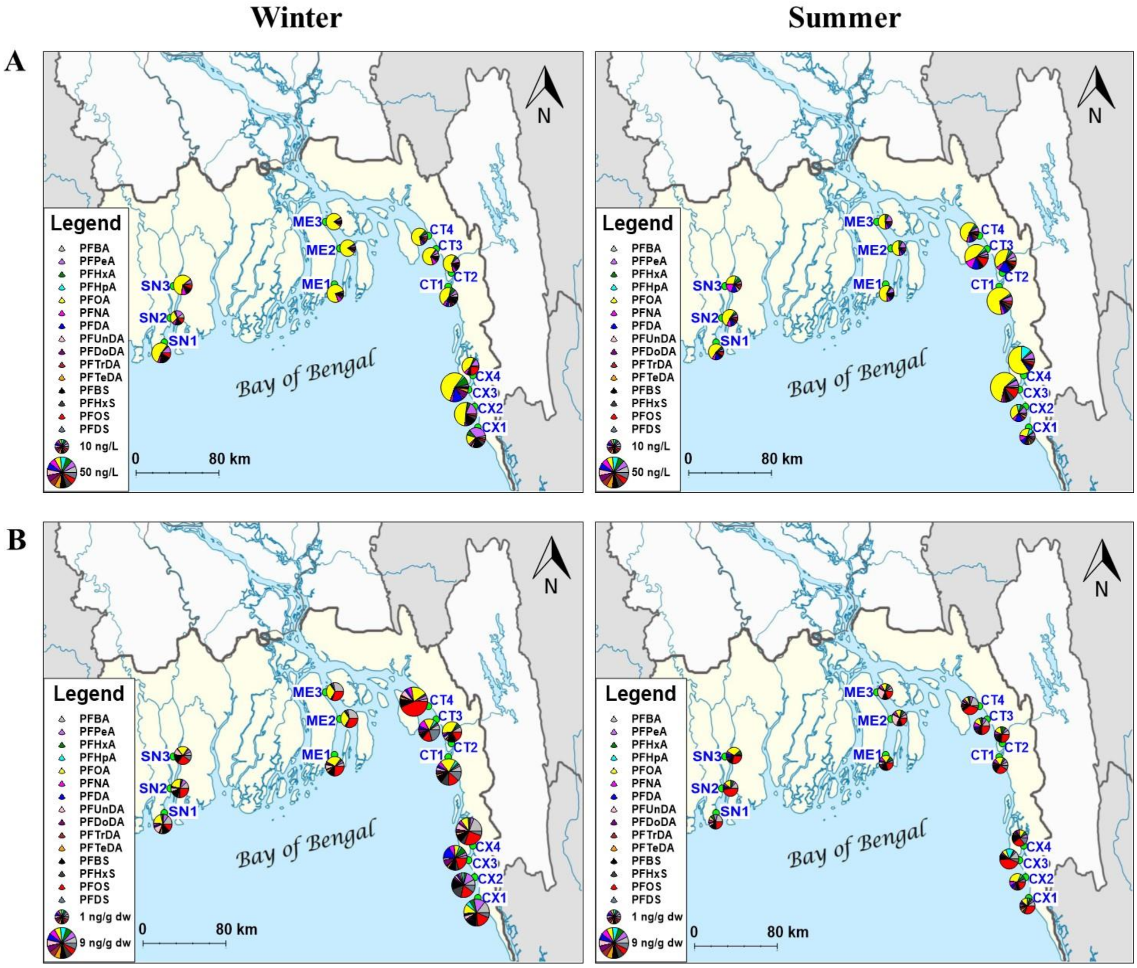

Figure 4-4. Distribution of PFAAs in surface water (A) and sediment (B) of the coastal area of Bangladesh. Samples with concentrations below the LOQ are represented as “0” for better visibility.

In particular, water samples at location CX3 (Bakkhali Estuary) showed the highest levels of PFAAs for both seasons (45.2 and 46.8 ng/L in winter and summer, respectively), indicating the existence of point source(s) in the area of Cox’s Bazar. Specifically, the Bakkhali Estuary receives huge discharges of untreated municipal and industrial effluents from the city, and the water is affected by activities such as intensive boating and fishing. Corsolini et al. (2012) found that the wastewater effluent is a point source of PFAA contamination in the Ganges River and the adjacent Sundarban mangrove wetlands in India.

The emissions from PFAAs-containing products used indoors and outdoors may also be the important sources for aqueous ecosystems (Zushi et al., 2009). PFAAs used indoors are discharged as sewage or municipal garbage. PFAAs used outdoors may leach out and flow into rivers, estuaries and coastal waters along with runoff. Airborn PFAAs also contribute significantly to the contamination of surface water environments (Dreyer et al., 2010). The main municipal garbage dump (mostly consumer products) of Cox's Bazar city is also located near CX3. Therefore, a significant amount of PFAAs might be emitted from this dumping site into the adjacent water body. Additionally, Cox's Bazar airport, which located very close to sampling location CX3, could be a potential source of PFAAs; Ahrens et al. (2015) described airport activities (such as fire training facilities) as a source of per- and polyfluorinated substances in nearby environmental matrices (water, sediment and fish). Furthermore, within Chittagong, site CT1 showed elevated levels of PFAAs in winter (22.1 ng/L) and summer (40.1 ng/L), respectively. This site is predominantly influenced by the port activities and also receives effluents from various industries that are located along the bank of the Karnaphuli River, which is the most polluted river in Chittagong division. Therefore, the Karnaphuli River should be considered as a potential PFAA source for the coastal areas of sampling stations CT1–CT2. Moreover, it was previously reported that the use of paints and grease repellents for ship and dock protection could contribute to the relatively high levels of PFAAs in port seawaters. Shipping and boat maintenance in the ports may also be responsible for the release of PFAAs into the surrounding waters (Paul et al., 2009).

The spatial distribution of PFAAs in surface sediment samples for both seasons is presented in **Figure 4-4B**. As for the water samples, Cox's Bazar and Chittagong showed higher levels of ∑PFAAs in sediment samples compared to the Sundarbans and the Meghna Estuary, which were at sampling locations CX1–CX4 (average of 6.59 and 2.49 ng/g dw in winter and summer, respectively). Lower levels were found at CT1–CT4 (average of 5.94 and 2.21 ng/g dw in winter and summer, respectively), followed by SN1–SN3 (average of 3.17 and 1.73 ng/g dw in winter and summer, respectively) and ME1–ME3 (average of 2.94 and 1.45 ng/g dw in winter and summer, respectively). Unlike the water samples, sediments from CT4 (ship breaking area) showed the highest concentration of total PFAAs (8.15 ng/g dw) in winter whereas CX3 (Bakkhali Estuary) showed the highest value (3.81 ng/g dw) in summer for sediment samples. Ship breaking activities along the coast of Chittagong may contribute to the PFAA pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PFAAs (Watkinson, 2013). There are approximately 100 e-waste recycling shops located very close to site CT4, generating approximately 2.7 million MT of toxic e-waste each year, the majority of which comes from the ship breaking sector. However, the waste generated from the e-waste recycling shops is discharged directly into the adjacent coastal water, which might be one of the most significant PFAA pollution sources in the study area. Additionally, a training institute for fire service and civil defense and the Chittagong international airport are located close to the study area and could also be sources of coastal pollution (Ahrens et al., 2015). Moreover, there are numerous multipurpose industrial establishments along the coast of Chittagong producing paper and pulp (e.g., the Karnaphuli paper mills), cement clinkers, fertilizers (e.g., Karnaphuli fertilizer), steel products, rubber and plastic, petroleum products (e.g., Super petrochemical), beverages, sugar, pharmaceuticals, tobacco, jute, textiles, fish products, tannery products, paint, rechargeable batteries, jewelry, plating, automobile engines, and electronics.

Apart from the Chittagong and Cox's Bazar sites, the other two sites, Sundarbans (SN1–SN3) and Meghna Estuary (ME1–ME3), did not show significant differences ($p >$ 0.05) in their total concentrations of PFAAs (∑PFAAs) in surface water or in sediment samples (**Figure 4-4**). In Sundarbans, higher PFAA concentrations were found near the mouth of the estuary leading into the Bay of Bengal, which might be attributed to a greater amount of deposition or accumulation of contaminated sediments transported by upstream waterways into the transitional zone of the estuary. The upstream estuary is mainly characterized by inputs of freshwater and is subject to daily tidal action. Therefore, the easily deposited particles, e.g., most of the coarse grain particles, tend to be deposited on the floor in the upstream area of the estuary due to the sudden change in the velocity of the water currents in this area. The fine particulates carried by rivers end up farther away in the estuary, where the rivers discharge their loads, or they can be carried offshore into the coastal area and beyond. In fact, the deposited sediments of fine particulates have higher accumulation and binding capacities for dissolved contaminants, particularly for hydrophobic organic compounds, such as PFAAs. Thus, fine particles might be a major carrier of PFAAs from the upstream source area to the mouth of the estuary (SN1). Interestingly, lower but detectable concentrations of PFAAs in surface waters and sediments from Meghna Estuary (**Figure 4-4**), an exclusively unindustrialized area, suggest that there are non-point sources of PFAAs (e.g., surface runoff) in the coastal area of Bangladesh. Furthermore, the Sundarbans and Meghna Estuary sites receive major volumes of water from the Ganges River of India through two main tributaries, the Gorhai River to the Sundarbans area via the Madhumoti River and the Yamuna River flowing down to the Meghna Estuary via the Mehgna River. Yeung et al. (2009) reported that the surface water of the Ganges River was contaminated by perfluoroalkyl chemicals (e.g., PFOA, PFDA, PFNA, PFPeA, PFHxA, and PFOS). The authors also noted that PFAA concentrations gradually declined due to dilution as the water mass increased downstream of the Ganges River. Thus, the coastal areas of Bangladesh might receive small but considerable amounts of PFAAs through the water flow of the Gorhai River and Yamuna River, which flow into the Sundarbans and Meghna Estuary sites. Generally speaking, the concentrations of ∑PFAAs in surface water and sediment at the Cox's Bazar (CX1–CX4) and Chittagong sites (CT1–CT4) were higher than those in the Meghna Estuary (ME1–ME3) and Sundarbans sites (SN1–SN3) because of greater development, including industrialization and urbanization activities.

4.3.4 Statistical analyses

Previous studies showed that the physicochemical properties of environmental matrices (e.g., water and sediment) can influence the fate and distribution of PFAAs in aquatic environments (Higgins and Luthy, 2006; Pan et al., 2014). Moreover, correlations between environmental contaminants might be indicative of common or multiple pollution sources in a specific study area. To identify such correlations and influences on the environmental parameters of PFAAs in the present study area, we calculated Spearman rank correlations ($p < 0.01$) between individual PFAAs and certain physical and chemical properties [pH, temperature (T), salinity, total suspended solids (TSS)] of the surface water samples. The results are presented in Table B-8 (for details of the physical and chemical properties of surface water and sediment samples, see Table B-4 in Appendix B). Significant correlations were found between PFPeA/[PFBS, PFOS], PFHxA/[PFDA, PFDoDA], and PFHpA/PFTeDA in winter as well as between PFBA/PFBS, PFHpA/PFTeDA, PFOA/PFBS, and PFOS in summer, indicating that the correlated compounds might originate from some shared pollution sources in the study area. In addition, the significant positive correlations between PFPeA, PFHxA, PFHpA, PFDA, PFDoDA and PFTeDA suggest a wide application range of PFCAs and/or sources arising from their potential precursor compounds such as fluorotelomer alcohols (FTOHs) in commercial products. Moreover, the concentrations of PFHpA, PFDoDA and PFTeDA were found to be correlated with water salinity in summer, which suggested that salinity could be affecting the pattern of relative contributions and the fates of waterborne PFAAs in the coastal or estuarine environments. However, no such correlations were found in winter samples of surface water, and such findings were expected because the concentration levels of most of the individual PFAAs and water properties did not show significant differences between the two seasons. Furthermore, Table B-9 (Appendix B) presents Spearman rank correlations ($p < 0.01$) for individual PFAAs and certain physical and chemical properties [pH, total organic carbon (TOC), black carbon (BC), total nitrogen (TN)] of sediment samples. Significant correlations between PFPeA/PFBS, PFHxA/[PFHpA, PFDA, PFBS], PFHpA/PFDoDA, and PFNA/[PFDA, PFDoDA] were found for winter samples, and PFOA/PFDA and PFOS/[PFDA, PFTeDA] were significantly correlated for summer samples. Additionally, the concentrations of PFHxA, PFNA, PFDA, PFDoDA, PFBS and PFOS were found to be correlated with TOC, BC and TN ($p < 0.01$), which is supported by previous findings reported by Pan et al. (2014). Interestingly, no such correlations were found in the summer samples may be because newly deposited coastal sediments (due to river bank erosion and/or runoff from heavy flooding and rainfall) contain lower amounts of organic compounds. The concentrations of sedimentary TOC (%) in winter (mean 0.61, range 0.06–1.28) were higher than in summer (mean 0.44, range 0.15–0.69) (Table B-4, Appendix B) which further confirms the hypothesis described above. Furthermore, sediment texture is considered to be one of the main factors affecting the accumulation profile of PFAAs in sediment. In the present study, sediment quality characteristics ranged from silt loam or clay to sandy loam (Table B-4, Appendix B), but the texture varied seasonally. We found that sites comprised of sandy loam or sandy clay loam had higher levels of PFAAs, whereas sites with silt loamy sediments generally had lower PFAAs concentrations in both seasons. Thus, it is more likely that the differences in PFAAs concentrations in the sediment samples are due to the influences of point and non-point sources.

The correlations among individual PFAAs between water and sediment, considering both seasons (Spearman rank correlation, $p < 0.01$, two-tailed) are presented in Table B-10 (Appendix B). The results indicate that some of the individual PFAAs in water samples (PFPeA, PFHxA, PFDA) have strong positive correlations/associations with some of PFAAs in sediment samples (PFPeA, PFHxA, PFDoDA), which suggests that these compounds probably had a common pollution source in the study area. This also supports the correlation results discussed in the previous section. However, this correlation analysis could be indicative of similar sources of associated PFAAs in surface waters and sediment samples in the aquatic environment, which are usually influenced by multiple input sources (Zhang et al., 2013). Moreover, a mass loading or mass balance calculation might determine the primary medium in which PFAAs will occur. In this study, concentrations were measured, but because the total masses of sediment and water were unknown, it was impossible to calculate the total mass of PFAAs in each medium. However, because concentrations of PFAAs in sediments were greater than those in water, sediments might be a useful integrating medium for pollution monitoring.

4.3.5 Partitioning behavior of PFAAs between sediment and water in a tropical environment

The partitioning of PFAAs between sediment and surface water is crucial for understanding the transport of PFAAs in the environment (Ahrens et al., 2010; Zhao et al., 2012; Zhu et al., 2014). The distribution coefficient (K_D) is the most commonly used parameter in the evaluation of the partitioning of organic pollutants in water. We calculated site-specific values of K_D (L/kg) as follows:

$$K_D = \frac{C_S}{C_W} \qquad (4.1)$$

C_S and C_W are concentrations of PFAAs in the sediment and surface water, with units of ng/kg dw and ng/L, respectively. Previous studies showed that the fraction of organic carbon (f_{OC}) has a significant influence on the sorption and transport of PFAAs in sediment (Higgins and Luthy, 2006). Hence, the organic carbon normalized partition coefficient (K_{OC}) provides a better indication of the partitioning behavior by reducing the variability between samples. K_{OC} (L/kg) was calculated as

$$K_{OC} = K_D \times \frac{100}{f_{OC}} \qquad (4.2)$$

where f_{OC} is the organic carbon fraction in sediment (%).

Average values of log K_D and log K_{OC} for PFAAs are shown in **Table 4-3**. In the present study, log K_D values were in the range of 1.63–2.63 and 2.26–2.88 for PFCAs and PFSAs, respectively, and they agreed well with the field-based K_D values reported by Campo et al. (2015). They were slightly higher than the values reported by Zhu et al. (2014) and Ahrens et al. (2010) (**Table 4-3**). In this study, the K_D values of PFOS (2.87±0.47) were much higher than those of PFOA (1.63±0.44), which explains why PFOA was more abundant in the water and PFOS predominated in the sediments. On the other hand, the present study revealed a higher value of log K_{OC} for PFOA (4.02±0.54) than in the Bohai coastal watersheds (2.68 ± 0.51) in China (Zhu et al., 2014) and Tokyo Bay (1.9±0.1) in Japan (Ahrens et al., 2010), but it agreed well with the reported values (3.76-4.83) from Spain (Campo et al., 2015). A much higher value of log K_{OC} (5.11±0.60) for PFOS was found than in previous field reports of log K_{OC} values (2.09−4.07; 3.75 ± 0.44; 3.8±0.1) (Campo et al., 2015; Zhu et al., 2014; Ahrens et al., 2010). Moreover, Guo et al. (2015) estimated log K_D (PFCAs: 0.65–2.49; PFSAs: 1.82–2.24) and log K_{OC} (PFCAs: 2.53–4.48; PFSAs: 3.77–4.15) for the Taihu lake, which is one of the most important and significant freshwater ecosystems in China (**Table 4-3**). Our log K_D values had similar ranges, but our log K_{OC} values were slightly higher than those reported by Guo et al. (2015). However, the differences in log K_D and log K_{OC} values calculated here and those reported in other studies were expected because the physicochemical parameters of the environmental matrices (sediment and water) of a tropical coastal area (present study) might be quite different from the other study areas, which include temperate and sub-tropical regions. Moreover, variation in the basic elements of climate (e.g., rainfall, temperature, sunlight and microbial degradation) between tropical and temperate regions might also influence the dissipation and/or accumulation of chemical pollutants in the aquatic environments. The environmental parameters of a tropical climate (e.g., heavy rainfall, higher temperature and greater microbial activities) may contribute to the levels of PFAAs in the environmental matrices (water and sediment), which eventually affects their partitioning behavior in the environment.

In this study, the average log K_{OC} ranged from 4.02 (PFOA) to 4.68 (PFBA and PFTeDA) for PFCAs, and 4.54 (PFBS) to 5.16 (PFDS) for PFSAs. The average log K_{OC} was directly proportional to the number of carbons in the PFAAs, which was consistent with previous reports (Higgins and Luthy, 2006; Ahrens et al., 2010). Exceptions occurred for PFBA and PFOA; PFBA had a relatively high value (4.68±0.52), and PFOA had a relatively low value (4.02±0.54). There are many factors influencing the fate of PFAAs in aquatic systems, but data on their distribution between water and sediment is still scarce. Because longer carbon chains increase hydrophobicity of PFAAs and enhance their interactions with carbonaceous materials, the log K_{OC} values of PFAAs increase as their carbon chains become longer. We also found that log K_{OC} increases linearly with increasing chain length, from PFOA to PFTeDA and from PFBS to PFDS (**Figure 4-5**), which agrees well with the trends reported by Kwadijk et al. (2010) and Labadie and Chevreuil (2011). However, the slopes of the regression lines for both PFCAs and PFASs (**Figure 4-5**) differ by several orders of magnitude from these two studies, which might be attributable to differences in sediment properties or to the fact that different PFAAs were considered.

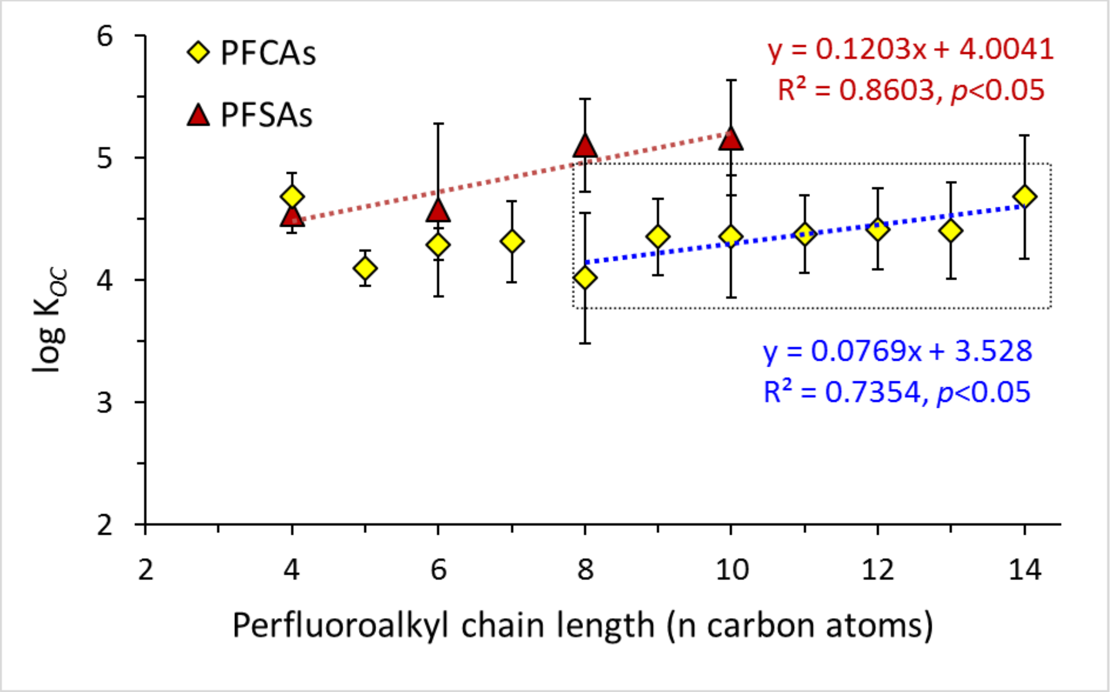

Figure 4-5. Correlation between perfluoroalkyl chain length (n carbon atoms) and log K_{OC}.

The sediment properties (e.g., organic carbon content) and water conditions (e.g., pH and salinity) may affect the partitioning of PFAAs between sediment and water in real environments (Higgins and Luthy, 2006). In this study, the influence of salinity and pH on PFAA partitioning was not significant ($p > 0.05$, data not shown) because their differences at most sampling locations did not vary significantly, with the exception of Meghna Estuary (ME1−ME3) (Table B-4, Appendix B). However, the organic fraction (f_{OC}) and other characteristics of the sediment might influence the partitioning of PFAAs. Correlations between K_Ds and f_{OC} were evaluated for individual PFAAs and are presented in **Table 4-3**. Significant correlations were found between f_{OC} and K_Ds of PFNA, PFTrDA, PFBS, PFHxS and PFOS ($p < 0.05$). This result is in agreement with previous laboratory research by Higgins and Luthy (2006). In contrast, some field research found no significant correlations between f_{OC} and K_Ds for PFAAs ($p > 0.05$) (Kwadijk et al., 2010; Hong et al., 2013).

Table 4-3. Calculated average log K_D (L/kg) and log K_{OC} (L/kg) for individual PFAAs and comparison with values reported in the literature.

PFAAs	Coastal area, Bangladesh (This study)					Llobregat river basin, NE Spain (Campo et al., 2015)		Bohai coastal watersheds, China (Zhu et al., 2014)		Tokyo Bay, Japan (Ahrens et al., 2010)		Taihu Lake, China (Guo et al., 2015)	
	C[a]	N[b]	log K_D	log K_{OC}	Correlation between log K_D and f_{OC} (r^2)[c]	log K_D	log K_{OC}	log K_D	log K_{OC}	log K_D	log K_{OC}	log K_D	log K_{OC}
PFBA	4	5	2.63±0.39	4.68±0.52	0.52 ($p > 0.05$)	2.33	4.27	1	3.02			1.64±0.24	3.58±0.29
PFPeA	5	14	1.99±0.24	4.1±0.42	0.19 ($p > 0.05$)	2.4	4.71	0.46	2.4			1.29±0.24	3.17±0.2
PFHxA	6	14	2.24±0.19	4.29±0.43	-0.20 ($p > 0.05$)			0.59	2.55			0.86±0.34	2.64±0.44
PFHpA	7	7	2.25±0.51	4.32±0.58	0.25 ($p > 0.05$)			0.45	2.33			1.43±0.43	3.36±0.47
PFOA	8	14	1.63±0.44	4.02±0.54	0.10 ($p > 0.05$)	2	4.02	0.63	2.68	0.04	1.9	0.65±0.30	2.53±0.41
PFNA	9	14	2.01±0.35	4.35±0.34	0.53 ($p < 0.05$)			1.03	3.07	0.6	2.4	1.88±0.44	3.80±0.52
PFDA	10	12	2.03±0.49	4.36±0.55	0.16 ($p > 0.05$)	2.51	4.55	1.6	3.59	1.8	3.6	2.16±0.31	4.07±0.40
PFUnDA	11	4	2.2±0.46	4.38±0.55	0.60 ($p > 0.05$)			2.25	4.19			2.47±0.41	4.37±0.56
PFDoDA	12	6	2.22±0.38	4.42±0.50	0.02 ($p > 0.05$)							2.49±0.13	4.48±0.18
PFTrDA	13	7	2.28±0.42	4.41±0.40	0.69 ($p < 0.05$)								
PFTeDA	14	6	2.38±0.47	4.68±0.51	0.05 ($p > 0.05$)								
PFBS	4	7	2.3±0.34	4.54±0.43	0.67 ($p < 0.05$)	2.88	4.92	2.18	4.24				
PFHxS	6	3	2.26±0.63	4.57±0.56	0.79 ($p < 0.05$)			2.24	4.15			1.82±0.36	3.77±0.51
PFOS	8	12	2.87±0.47	5.11±0.60	0.59 ($p < 0.05$)	1.3	3.26	1.7	3.75	2.1	3.8	2.24±0.61	4.15±0.69
PFDS	10	3	2.88±0.51	5.16±0.47	0.69 ($p > 0.05$)								

[a] Number of carbon atoms in PFAAs carbon chain; [b] The available number of sampling sites in which particular PFAAs were detected from both surface water and sediment;
[c] Spearman rank correlation, $p < 0.05$;

We found a similar result for PFAA homologs except for PFNA, PFTrDA, PFBS, PFHxS and PFOS. In addition to the difference in sediment characteristics, this variation may be partially attributed to the sediment-water equilibrium status (Zhang et al., 2012), which is rarely achieved in such a complex coastal aquatic ecosystem. However, if we assume that the water environment of the study area was in equilibrium conditions at the time of sampling because the samples were collected when the water was quiet and calm, PFAAs might be exchanged between the two phases during the water and sediment interaction process. For example, Pan et al. (2014) reported that, due to its high affinity for sediment, PFOS may serve as an internal reservoir that may later be released back into the water column when environmental conditions change (e.g., re-suspension of the sediment and diffusion as a result of the dilution of water). Even if the environmental inputs of PFOS and/or PFOA into the aquatic systems were reduced, the aqueous phase PFOS may be buffered to some extent due to the internal loads in the sediment. The understanding of the fate of PFAAs in aqueous environment is still incomplete, and relevant laboratory studies have found different results than some of the field-based distribution coefficient studies (Higgins and Luthy, 2006; Ahrens et al., 2010; Kwadijk et al., 2010). Because of high variability in environmental parameters, field studies do not always find the same correlations as studies conducted under laboratory conditions. It is almost impossible to determine all physicochemical parameters of a complex coastal ecosystem, so the partitioning behavior may be obscured by other properties of the system that we do not pay attention to in field studies.

4.3.6 Preliminary environmental hazard assessment

A preliminary assessment was conducted to determine the ecotoxicological risk to aquatic organisms, including fish, birds, and other invertebrates, due to exposure to selected PFAAs (PFOA and PFOS, the two most frequently detected PFAAs in aquatic environments worldwide). The assessment was based on concentrations of the compounds in surface water and sediment. PFOS and PFOA can cause both acute and chronic toxicity in fish and invertebrates (Giesy et al., 2010; Zhu et al., 2014). Multiple approaches are available to derive environmental quality values. The most commonly used methods are described by the Great Lakes Initiative guidelines of the USEPA (Sanchez-Avila et al., 2010). The values are calculated using reported acute and subacute toxicity values of PFOS and PFOA to aquatic organisms and birds (Giesy et al., 2010). Recently, Giesy et al. (2010) calculated suggested criteria maximum concentrations (CMC) for the most sensitive aquatic species for PFOS (21 μg/L) and PFOA (25 mg/L). Criteria continuous concentrations (CCC) for PFOS (5.1 μg/L) and PFOA (2.9 mg/L) and avian wildlife values (AWV) for PFOS (47 ng/L) were also suggested. In this study, an evaluation of the PFOA and PFOS burden hazard to surface water-dwelling aquatic animals, calculated as a hazard quotient (HQ), was performed by comparing their concentrations with the suggested water quality criteria mentioned above. Conversely, the ratio of the measured environmental concentration (MEC) to the predicted no-effect concentration (PNEC), termed the hazard quotient (HQ), was used to evaluate the ecological risks posed by PFOA and PFOS to the sediment-dwelling benthic and bentho-pelagic fish and other aquatic animals of the study area. An HQ value below 1 indicates no potential risk. The PNEC of sediment can be derived from: a) the no observed effect concentration (NOEC) or the median effect/lethal concentration (E/LC_{50}) with an assessment factor (AF); and b) the equilibrium partitioning (EqP) method when the toxicity data are unavailable (Lepper, 2005). Because the PNECs of PFOA and PFOS for benthic animals in the Bay of Bengal coast are not available, the PNEC selected in this study was derived from a study by Zhao et al. (2013); it was calculated by using K_D values and the equilibrium partitioning method (van Vlaardingen and Verbruggen, 2007). The PNECs for PFOS and PFOA were 4.9 ng/g and 86 ng/g in riverine and estuarine sediments, and they were 0.49 ng/g and 8.6 ng/g in deep sea marine sediments, respectively (Zhao et al., 2013). However, K_D varies with sediment conditions, such as the concentrations of organic compounds and ionic substances (Higgins and Luthy, 2006; Zushi et al., 2010). Thus, further development of the site-specific PNEC in sediment is required.

In the present study, all the HQ values for PFOA and PFOS were less than 1 (**Table 4-4**), indicating little or insignificant risks to the pelagic, benthic and benthopelagic organisms. More recently, Valsecchi et al. (2016) reported the national EQSs (Environmental Quality Standards) termed as AA-QSs (Annual Average Quality Standards) aiming to protect the pelagic aquatic organisms for PFOA (3 µg/L) and selected short chain perfluoroalkyl acids, such as PFBA (11 µg/L), PFPeA (3.2 µg/L) and PFBS (37 µg/L), and these EQSs were adopted in the Italian legislation (Italian Legislative Decree 172/2015). Our results revealed that the concentrations of PFAAs in water samples were much lower than the corresponding AA-QSs of the Italian EQSs. Conversely, the European Commission (EC) recently included PFOS in the list of priority substances, setting an EQS of 0.13 ng/L for saltwater to protect the aquatic pelagic organisms from prolonged exposure to water-phase PFOS (Directive 2013/39/EC). In the present study, 93% of the surface water samples exceeded the EC guideline for PFOS, suggesting a potential long-term risk to the pelagic organisms in the coastal area of Bangladesh.

Table 4-4. Hazard quotient (HQ) of PFOA and PFOS in surface water and sediment of the coastal area of Bangladesh.

Sites	Hazard quotients (HQ)						
	Water					Sediment	
	PFOA/CCC	PFOS/CCC	PFOA/CMC	PFOS/CMC	PFOS/AWV	PFOA/PNEC$_{PFOA}$	PFOS/PNEC$_{PFOS}$
Winter							
CX1	<0.001	<0.001	<0.001	<0.001	0.022	0.010	0.260
CX2	<0.001	<0.001	<0.001	<0.001	0.019	0.002	0.236
CX3	<0.001	<0.001	<0.001	<0.001	0.003	0.005	0.249
CX4	<0.001	0.001	<0.001	<0.001	0.091	0.008	0.380
CT1	<0.001	<0.001	<0.001	<0.001	0.009	0.016	0.202
CT2	<0.001	<0.001	<0.001	<0.001	0.010	0.017	0.141
CT3	<0.001	<0.001	<0.001	<0.001	0.010	0.009	0.156
CT4	<0.001	<0.001	<0.001	<0.001	0.017	0.017	0.741
ME1	<0.001	<0.001	<0.001	<0.001	0.005	0.011	0.191
ME2	<0.001	<0.001	<0.001	<0.001	0.006	0.010	0.179
ME3	<0.001	<0.001	<0.001	<0.001	0.005	0.005	0.197
SN1	<0.001	<0.001	<0.001	<0.001	0.043	0.008	0.092
SN2	<0.001	<0.001	<0.001	<0.001	0.030	0.008	0.163
SN3	<0.001	<0.001	<0.001	<0.001	0.034	0.007	0.151
Summer							
CX1	<0.001	<0.001	<0.001	<0.001	0.013	0.003	0.124
CX2	<0.001	–	<0.001	–	–	0.008	0.088
CX3	<0.001	0.001	<0.001	<0.001	0.109	0.003	0.342
CX4	<0.001	<0.001	<0.001	<0.001	0.036	0.003	0.111
CT1	<0.001	<0.001	<0.001	<0.001	0.037	0.004	0.103
CT2	<0.001	<0.001	<0.001	<0.001	0.036	0.003	0.095
CT3	<0.001	0.001	<0.001	<0.001	0.091	0.003	0.117
CT4	<0.001	<0.001	<0.001	<0.001	0.015	0.002	0.209
ME1	<0.001	–	<0.001	–	–	0.003	0.065
ME2	<0.001	<0.001	<0.001	<0.001	0.011	0.003	0.072
ME3	<0.001	<0.001	<0.001	<0.001	0.027	0.001	0.073
SN1	<0.001	<0.001	<0.001	<0.001	0.025	0.001	0.062
SN2	<0.001	<0.001	<0.001	<0.001	0.019	0.003	0.178
SN3	<0.001	<0.001	<0.001	<0.001	0.015	0.007	0.118

CCC: Criteria continuous concentration; CMC: Criteria maximum concentration; AWV: Avian wildlife value; PNEC: Predicted no-effect concentration;

Moreover, PFOS concentrations were several orders of magnitude less than the guideline value for the protection of avian wildlife (HQ < 1), which suggests that the present level of PFOS in surface water is unlikely to cause harm to avian wildlife (**Table 4-4**). However, it should be noted that more toxicological data will be needed to undertake a more comprehensive risk assessment. Specifically, estimation of the concentrations of PFAAs in biota samples will allow further assessment of the risks to ecological systems due to exposure to these compounds. Low but persistent concentrations of PFAAs in water and sediment could lead to higher levels in fish and shellfish, some of which (e.g., Hilsa shad, Pomfret, Shrimp, and Crab) are consumed by the local population.

4.4 Conclusions

This study provides national baseline data on the occurrence of perfluoroalkyl acids (PFAAs) in the surface waters and sediments of the coastal area of Bangladesh. All 15 target PFAAs were detectable in both matrices with different compositional patterns and detection frequencies. The total concentration of PFAAs (∑PFAAs) in surface water and sediment samples was in the range of 10.6–46.8 ng/L and 1.07–8.15 ng/g dw, respectively. PFOA in water (range of 3.17–27.8 ng/L) and PFOS in sediment samples (range of 0.60–1.14 ng/g dw) were found to be the most abundant PFAAs; they had high concentrations and were detected in 100% of samples. Comparisons with the literature showed that PFOA and PFOS concentrations in surface water and sediment were comparable to or less than most other values reported, particularly those reported in China, Japan, Korea and Spain. Anthropogenic activities (e.g., urbanization, industrialization, ship breaking, port activities, boating and fishing) were identified as potential sources of PFAA contamination in the coastal area of Bangladesh. This study also provides field-based distribution coefficients (K_D and K_{OC}) for a wide range of PFAAs in a tropical coastal environment that is very dynamic and complex. Such data are still scarce, and more data are needed for a better understanding of the fate and behavior of PFAAs in global aquatic ecosystems. Preliminary environmental hazard assessment indicated that none of the PFOA or PFOS concentrations exceed the thresholds of acute hazard, suggesting little potential risk to aquatic organisms at different trophic levels regarding the possible effects from short term concentration peaks. Conversely, prolonged exposure to PFOS might be an ecological threat to the pelagic organisms. However, the ubiquitous presence of PFAAs in surface waters and sediments is an indicator of their widespread distribution in the coastal area of Bangladesh, which should not be ignored. Immediate control measures are recommended, particularly to prevent the discharge of untreated wastewater effluents and the dumping of e-waste and municipal garbage into aquatic environments.

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Chapter 5

OCCURRENCE AND ASSESSMENT OF PERFLUOROALKYL ACIDS (PFAAs) IN COMMONLY CONSUMED SEAFOOD FROM THE COASTAL AREA OF BANGLADESH

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Occurrence and assessment of perfluoroalkyl acids (PFAAs) in commonly consumed seafood from the coastal area of Bangladesh

Abstract

Dietary intake is the most important route of perfluoroalkyl acids (PFAAs) exposure and seafood is the major dietary component for the coastal populations. It is, therefore, an urgent need to monitor the levels of PFAAs in seafood. This study reports the first evidence of the occurrence of PFAAs in commonly consumed seafood from the coastal area of Bangladesh (a tropical ecosystem). Fifteen target PFAAs in 48 seafood samples collected in winter and summer of 2015 were measured by HPLC-MS/MS. The ∑PFAAs (ng/g wet weight) in finfish and shellfish were in the range of 0.32–14.58 and 1.31–8.34, respectively. Perfluorooctanesulfonate (PFOS) in finfish (0.1–3.86 ng/g ww) and shellfish (0.1–1.99 ng/g ww), whereas perfluoroctanoic acid (PFOA) in shellfish (0.07–2.39 ng/g ww) were found to be the most abundant PFAAs. The results were comparable with other studies worldwide, particularly from China, Spain, Sweden, and USA. Majority of monitored PFAAs showed no obvious seasonal variation. However, seafood from the southeast area (Cox's Bazar and Chittagong) showed relatively higher levels of PFAAs. Moreover, the estimated daily intakes (EDI, ng/kg body weight/day) of PFOA and PFOS in seafood ranged from 0.06 to 0.85 and 0.94 to 2.16 for adults, whereas it was 0.08 to 1.32 and 1.08 to 2.85 for children, respectively. Nevertheless, the EDI from seafood is much lower than the tolerable daily intake (TDI) recommended by the European Food Safety Authority in 2008 (PFOA: 1500 ng/kg-bw/day; PFOS: 150 ng/kg-bw/day), indicating low health risk of PFAAs exposure via seafood consumption among the coastal populations in Bangladesh.

5.1 Introduction

Perfluoroalkyl acids (PFAAs) are a new class of emerging organic pollutants with both hydrophobic and lipophobic/oleophobic properties. Because of high surface activity, thermal and acid resistance, weak intermolecular interactions, and water and oil repellency, PFAAs have been extensively used in industrial and consumer applications as surfactants and surface protectors (Kissa, 2001, Giesy and Kannan, 2002). PFAAs have been detected globally in the environmental matrices of riverine, estuarine and coastal ecosystems including water, sediments, and biota (Senthil et al., 2009; Zushi and Masunaga, 2009; Zushi et al., 2010; Naile et al., 2013; Ahrens et al., 2015). In addition, PFAAs were also found in food (Gulkowska et al., 2006; Ye et al., 2008), human blood (Falandysz et al., 2006; Fromme et al., 2007; Yeung et al., 2008; Zhang et al., 2011) and human milk (So et al., 2006; Tao et al., 2008; Liu et al., 2010). Perfluorinated carboxylic acids (PFCAs) and perfluorinated sulfonic acids (PFSAs) are the two most prevalent groups of PFAAs in the environment (Pan et al., 2014). In particular, long chain (more than seven fully fluorinated carbon atoms, e.g. perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS)) compounds are known to be bioaccumulative (Martin et al., 2003), and they have received great attention from the public and scientific community due to their potential adverse impacts on the ecosystem and human health (Peters and Gonzalez, 2011; Lau, 2012). PFOS with its precursor, perfluorooctanesulfonyl fluoride (PFOSF), were listed as persistent organic pollutants (POPs) in Annex B of the Stockholm Convention in 2009 (UNEP, 2009). Production of PFOS and similar perfluorooctyl products was phased out in USA and Europe in years 2000–2002 (OECD, 2002), but production continues elsewhere, particularly in developing countries (Wang et al., 2009).

Sources of human exposures to PFAAs have not yet been fully characterized and are still under debate. The Canadian Total Diet Study suggest that diet was an important source of PFAAs exposure along with other routes such as air, water, dust, treated carpeting and apparel (Tittlemier et al., 2007). Seafood is a major dietary source of protein and healthy lipids. In particular, the long chain omega-3 fatty acids have beneficial roles in human health. Despite the benefit of a seafood diet, a concern of frequent seafood consumption is the potential risk from exposure to toxic chemicals (Domingo et al., 2012). Some other previous studies of different regions around the world have shown that seafood (finfish and shellfish) accounted for >50% of PFOS exposures along with other PFAAs in the exposed populations in Sweden (Berger et al., 2009), Spain (Ericson et al., 2008), Canada (Fromme et al., 2007), Poland (Falandysz et al., 2006), China (Zhang et al., 2011; Wu et al., 2012), Japan (Fujii et al., 2015). Moreover, consumption of fish and seafood has been associated with high levels of PFAAs in human blood (Holmström et al., 2005; Falandysz et al., 2006; Fromme et al., 2007; Zhang et al., 2011). Studies concluded that the PFAAs contamination levels found in seafood may pose a significant health risk to human and wildlife, which requires immediate assessment (Berger et al., 2009; Ericson et al., 2008; Fromme et al., 2007; Falandysz et al., 2006; Zhang et al., 2011; Wu et al., 2012). Besides, the pollutant levels in aquatic biota should correlate with concentrations in their environment (Das and Das, 2004).

Bangladesh is an exclusively riverine agricultural country that is undergoing rapid industrialization, urbanization and economic development in recent years, particularly in the coastal regions and continues to bring about increasing amounts of municipal, industrial and agricultural wastes into the water body. The environmental quality in the estuarine and nearby coastal areas has been adversely affected by intense industrial and urban activities. The main sources of PFAAs in the aqueous environment are direct discharge of industrial or municipal wastewater and effluents containing these compounds, runoff from contaminated soil by precipitation, accidental spills or by the release of non-treated or semi-treated discharges, landfills, dumping of municipal garbage (Zhu et al., 2014; Campo et al., 2015; Ahrens et al., 2015). Bangladesh has a highly irregular deltaic marshy coastline of 580 kilometers, fissured by many rivers and streams to the Bay of Bengal. The increase in population and industrial pressure along rivers, and estuarine and coastal areas of Bangladesh pose an important threat to the coastal ecosystems since contaminants such as PFAAs can be accumulated in the coastal or marine food chain and also local population can be exposed via contaminated seafood consumption. About 42 million people (30 % of the total population) live in the coastal area (47,211 km^2; 32 % of the total land area) of Bangladesh, of which about 5 million are engaged directly in commercial fishing (BOBLM, 2011). Seafood is the most important and one of the major dietary components of the coastal populations. Therefore, it is an urgent need to assess the potential health risk that might be posed from the dietary exposure of PFAAs through seafood consumption. However, to the best of our knowledge, no study regarding PFAAs contamination has been carried out so far in this region. Hence, the present study was initiated, for the first time in Bangladesh, to explore the possible occurrence of PFAAs in seafood (finfish and shellfish) from the coastal areas of Bangladesh. The compositional pattern and spatiotemporal variations of PFAAs in the seafood samples were examined. Moreover, we also contrasted the concentrations of PFAAs in seafood from other countries and analyzed the possible sources of the contamination. Using the data, we estimated the average daily intake of PFAAs via seafood consumption by the coastal residents (adults and children) of Bangladesh and figured out the health effects by comparing the intake data with the health criteria recommended by the international authorities.

5.2 Materials and methods

5.2.1 Study area and sample collection

In the present study, seafood samples were collected from four main fish landing centers located in the four major coastal areas of Bangladesh (Cox's Bazar, Chittagong, Bhola and Sundarbans; Figure 5-1) which contribute approximately 70–80% of the total catchment of the seafood. A total of 48 seafood samples (5 finfish and 2 shellfish species) were collected in winter and summer of 2015, of which the finfish species included Ilish (*Tenualosa ilisha*), Rupchanda (*Pampus argentius*), Loitta (*Harpadon nehereus*), Sole (*Cynoglossus lingua*), and Poa (*Otolithoides pama*), whereas the shellfish species included shrimp (*Penaeus indicus*) and crab (*Scylla serrata*). The varieties were the most commonly consumed seafood in the coastal area of Bangladesh. The edible portion of the collected seafood samples were homogenized, weighed, freeze-dried for about 48 h until the constant weight was attained.

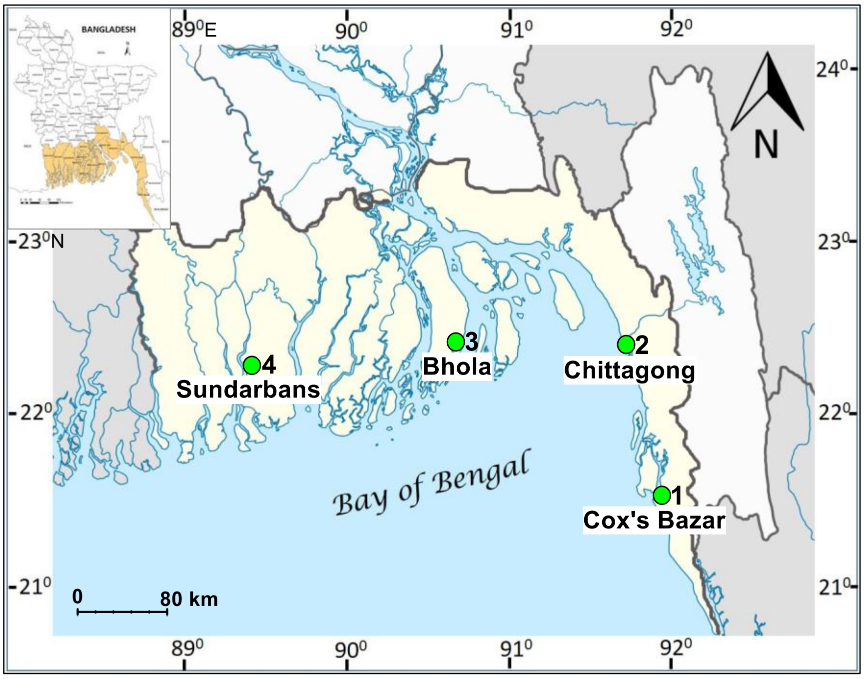

Figure 5-1. Map showing the four major coastal areas of Bangladesh along with the position of four main fish landing centers from where seafood samples were collected in the present study; (1): Fish Landing and Wholesale Fish Market, Cox's Bazar; (2): Chittagong fishing harbor, Chittagong; (3): Fish Landing and Wholesale Fish Market, Bhola; (4): Fish Processing and Marketing Centre, Sundarbans area in Mongla. Colored area in the inset map represents the coastal area of Bangladesh.

The species-specific information with their biometric data and pretreatment are shown in Table A-2 (Appendix A). All the processed samples were brought to Yokohama National University, Japan for further chemical analysis. Detailed description of the study area and sampling procedure was discussed in Chapter 3 of this thesis.

5.2.2 Target PFAAs compounds

Samples were analyzed for 15 PFAAs, including perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS) and perfluorodecane sulfonate (PFDS). Description about the chemicals and reagents is detailed in Chapter 4.

5.2.3 Sample extraction and clean-up

The alkaline digestion method was suggested to be an effective and reliable extraction method for biological samples (Taniyasu et al., 2005; Ye et al., 2008; Campo et al., 2015; Hong et al., 2015). The extraction procedure for seafood samples were adopted from Ye et al. (2008) and Campo et al. (2015) with some modifications. Briefly, 0.5 g of homogenized freeze dried sample was transferred to a 50 mL PP tube and spiked with 800 µL of 5 ng/mL of $^{13}C_4$-PFBA, $^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_2$-PFDoDA and $^{13}C_4$-PFOS as internal standards, to which 10 mL of 10 mM NaOH in methanol (0.1% water by volume) was added and sonicated for 30 min in a water bath at 60 °C followed by shaking at 250 rpm on an orbital shaker for 16 h. The samples were then centrifuged at 4000 rpm for 5 min and 5 mL of the supernatant was transferred into a new 50 mL PP tube. After diluting to 30 mL with Mili-Q water, vortexed, and subsequently loaded onto an Oasis® WAX SPE cartridge that had been preconditioned with 4 mL of 0.1% ammonium hydroxide (in methanol), 4 mL of methanol, and 4 mL of Milli-Q water at a rate of one drop per second. The cartridge was then washed with 4 mL of 50% methanol in Milli-Q water and centrifuged for 10 min at 3000 rpm to remove the residual water. Target compounds were eluted with 4 mL of methanol and 4 mL of 0.1% ammonium hydroxide (in methanol) followed by an ENVI-Carb™ tube (1mL, 100mg) for additional clean-up and collected in a 15 mL PP centrifuge tube. Finally, the eluate was concentrated to 400 µL under a gentle stream of high-purity nitrogen gas at 40°C for injection.

5.2.4 Instrumental analysis and Quality assurance/quality control (QA/QC)

The PFAAs in the extract were analyzed by a high-performance liquid chromatography-tandem mass spectrometry system (HPLC-MS/MS) that consisted of an HP 1100 HPLC system (Agilent Technologies) interfaced with a Micromass Quattro Ultima Pt mass spectrometer (Waters Corp., Milford, MA) operated in the electrospray negative ionization mode. The separation was achieved on an Agilent Zorbax XDB-C18 column (150 mm × 2.1 mm, aperture size 5 µm) after the injection of a 10 µL aliquot of sample extract. The column was maintained at 40 °C during the sample analysis. Quantification of all PFAAs was conducted using an internal standard calibration curve consisting of a concentration gradient (0.01, 0.1, 5, 10, 30, 50 µg/L) with the deviation of every point from the regression line less than 20% from its theoretical value. The standard calibration curve showed strong linearity (correlation coefficients, $r^2 > 0.99$). The linearity and repeatability of these calibration curves were confirmed prior to each set of determinations. The limit of detection (LOD) for each analyte was evaluated as the smallest mass of compound resulting in a S/N ratio that was equal to or greater than 3, and the limit of quantitation (LOQ) was defined as the higher either the concentration in a standard yielding a S/N ratio of 10:1. The LODs and LOQs for target PFAAs ranged from 0.01 to 0.05 ng/g wet weight (ww) and 0.03 to 0.19 ng/g ww, respectively. Instrumental blanks and procedural blanks were below the limit of quantification (LOQ). Analyte recovery was determined using spiked samples to determine the accuracy of the methods. Matrix spike recovery was determined by spiking of target compounds into the samples (n = 3) at a level of 10 ng/g, followed by the extraction and analysis as described in the previous section. The mean recoveries of PFAAs spiked into the seafood samples were 82%–119%. Information about the instrumental analysis and quality control are detailed in Appendix B.

5.2.5 Statistical analysis

Statistical analyses were performed with IBM SPSS software (Version 23.0, IBM Corp., NY, USA). The significance level was set at $p = 0.05$. Before analyzing, concentration values lower than the LODs were set to LOD/2 (Succop et al., 2004). A statistical distribution test called *P–P* plots was carried out to test for normality. Spearman rank correlation analysis was used to examine possible correlations among various PFAAs in the samples. A one-way

ANOVA was performed to determine the significant difference between the concentrations of PFAAs detected in the seafood samples from the coastal area of Bangladesh and seasonal variations.

5.3 Results and discussion

5.3.1 Occurrence, concentrations and composition of PFAAs in Seafood

In the present study, we first demonstrated the presence of PFAAs in the Bangladeshi seafood. A summary of the concentrations and detection frequency (%) of PFAAs in the investigated seafood samples is shown in **Table 5-1**. All the fifteen target PFAAs were detectable in the finfish samples for both seasons. On the contrary, PFPeA, PFHxA and PFHxS in winter, and PFHxA and PFHpA in summer were not detected in shellfish samples. The detection frequencies of PFAAs in this study were higher than those reported in some previous studies (Gulkowska et al., 2006; Ye et al., 2008; Zhao et al., 2011), which might be attributed to the relatively lower LOQs of the present study (Table B-11, Appendix B). Among the PFCAs, PFBA, PFPeA, PFHxA and PFHpA had lower detection frequencies (0–44%) and mean concentrations (<LOD–0.21 ng/g ww) than the other PFCAs, which can be attributed to the greater water solubility and lower bioaccumulation potential of short-chain PFCAs (Holmstrom and Berger, 2008). PFDoDA and PFTrDA in the finfish samples, whereas PFOA, PFNA, PFDA, PFDoDA and PFTrDA in the shellfish samples were detected with higher frequencies (50–100%) and mean concentrations (0.1–0.83 ng/g ww), which could be explained by the fact that the environmental occurrence of long-chain carboxylates may partly be a result of impurities of long-chain compounds in the technical products of PFOA and PFNA and partly a result of the degradation of other long-chain fluorinated chemicals, such as fluorotelomer alcohols (Ellis et al., 2004; Holmstrom and Berger, 2008). On the other hand, among the PFSAs, PFBS and PFOS were the most frequently detected PFAAs in seafood samples of the present study with the higher detection frequencies (63–100%) and mean concentrations (0.44–1.23 ng/g ww) than the other PFSAs (**Table 5-1**). In general, the total concentrations of the PFAAs (sum of 15 PFAAs, ∑PFAAs) in the finfish ranged from 0.32 to 8.71 ng/g ww in winter, and 0.80 to 14.58 ng/g ww in summer, whereas in the shellfish it was 1.31 to 7.18 ng/g ww in winter and 1.41 to 8.34 ng/g ww in summer (**Table 5-1**). Thus, the present results indicated that the finfish were contaminated with PFAAs slightly higher than the shellfish, which were in agreement with the findings reported in some

Table 5-1. Concentrations (ng/g ww) and detection frequency (%) of PFAAs in seafood (finfish and shellfish) in two seasons (winter and summer) collected from the coastal area of Bangladesh.

Sample	Season		PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFAAs
Finfish	Winter	Minimum	<LOD[1]	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.21	<LOD	0.32
		Maximum	0.60	0.80	0.73	1.12	0.21	0.29	0.50	0.76	1.90	1.77	0.77	0.73	0.09	2.41	1.46	8.71
		Mean	0.13a[4]	0.21a	0.12a	0.15a	0.05a	0.08a	0.06a	0.15a	0.39a	0.39a	0.08a	0.29a	0.02a	0.64a	0.20a	2.97a
		Median	0.56	0.34	0.42	0.99	0.09	0.24	0.12	0.71	0.14	0.31	0.43	0.51	0.07	0.45	0.72	2.28
		DF[3]	19	44	25	13	50	25	19	25	75	75	13	69	13	100	25	
	Summer	Minimum	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.10	<LOD	0.80
		Maximum	0.72	0.65	0.65	1.08	0.40	1.56	1.20	0.88	2.01	1.98	1.34	1.47	0.69	3.86	2.02	14.58
		Mean	0.13a	0.18a	0.12a	0.21a	0.08a	0.32b	0.34b	0.13a	0.43a	0.43a	0.28a	0.49a	0.20b	1.23b	0.22a	4.78a
		Median	0.60	0.38	0.35	1.07	0.09	0.45	0.41	0.45	0.28	0.19	0.38	0.73	0.48	0.80	0.57	3.50
		DF	19	44	25	19	44	56	69	25	63	81	44	63	50	100	25	
Shellfish	Winter	Minimum	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.12	<LOD	<LOD	<LOQ[2]	<LOD	<LOD	<LOD	0.21	<LOD	1.31
		Maximum	0.64	<LOD	<LOD	0.66	1.26	2.11	1.34	0.98	0.31	0.49	<LOD	0.80	<LOD	1.61	0.71	7.18
		Mean	0.12a	<LOD	<LOD	0.11a	0.58b	0.55b	0.47b	0.19a	0.09a	0.14a	<LOD	0.38a	<LOD	0.76a	0.11a	3.59a
		Median	0.64	<LOD	<LOD	0.66	0.42	1.02	0.20	0.72	0.10	0.21	<LOD	0.40	<LOD	0.74	0.40	2.71
		DF	13	0	0	13	100	50	100	25	63	50	0	88	0	100	25	
	Summer	Minimum	<LOD	<LOD	<LOD	<LOD	0.24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.10	<LOD	1.41
		Maximum	0.66	0.19	<LOD	<LOD	2.39	0.64	0.50	1.17	0.24	0.54	0.12	1.58	0.36	1.99	1.08	8.34
		Mean	0.17a	0.06b	<LOD	<LOD	0.83b	0.29b	0.23b	0.23a	0.11a	0.17a	0.03a	0.51a	0.05a	0.44a	0.21a	3.39a
		Median	0.57	0.17	<LOD	<LOD	0.67	0.35	0.28	0.56	0.15	0.21	0.12	0.75	0.36	0.25	0.22	2.30
		DF	25	25	0	0	100	75	75	38	75	63	13	63	13	100	50	

[1] Limit of detection (LOD): PFBA (0.05), PFPeA (0.03), PFHxA (0.03), PFHpA (0.04), PFOA (0.01), PFNA (0.03), PFDA (0.02), PFUnDA (0.02), PFDoDA (0.01), PFTrDA (0.02), PFTeDA (0.03), PFBS (0.02), PFHxS (0.01), PFOS (0.02), PFDS (0.02) (ng/g ww);

[2] Limit of quantification (LOQ): PFBA (0.19), PFPeA (0.07), PFHxA (0.10), PFHpA (0.13), PFOA (0.03), PFNA (0.10), PFDA (0.07), PFUnDA (0.04), PFDoDA (0.03), PFTrDA (0.05), PFTeDA (0.08), PFBS (0.09), PFHxS (0.03), PFOS (0.07), PFDS (0.07) (ng/g ww);

[3] Detection frequency (%);

[4] Values with vertically different letters differ significantly ($p < 0.05$; Tukey-HSD test).

previous studies conducted elsewhere (Gulkowska et al., 2006; Ericson et al., 2008; Yang et al., 2012; Wu et al., 2012).

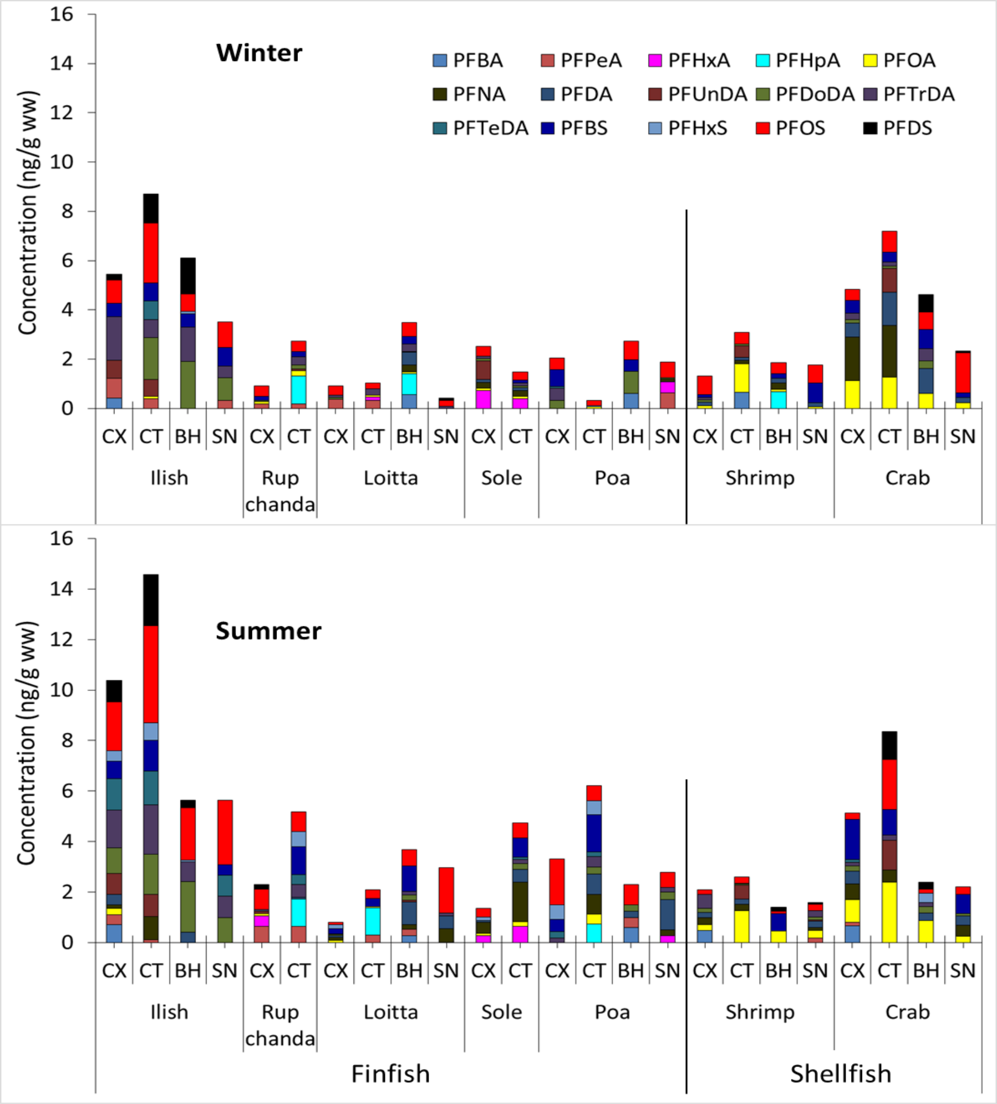

Figure 5-2. Concentrations of PFAAs in seafood (finfish and shellfish) of Bangladeshi coastal area in the two seasons. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans.

For both the finfish and shellfish samples, majority of the monitored PFAAs did not show clear seasonal variation, excepting the concentrations of PFNA, PFDA and PFOS in finfish which differed significantly between seasons ($p < 0.05$) with relatively higher concentrations in summer samples (**Table 5-1**). Concentrations of PFAAs in aquatic organisms is strongly dependent on the concentrations of PFAAs in water (Hong et al., 2015).

Therefore, the elevated levels of these long-chain PFAAs (PFNA, PFDA and PFOS) might be due to the higher concentration of PFAAs in water in summer, which probably due to their specific seasonal usage or applications in certain industries (e.g., the textile, paper, and leather industries) or the seasonal use of consumer products, although the exact reasons for this phenomenon are still unknown. Moreover, studies have shown that the long-chain PFAAs (e.g. PFNA, PFDA and PFOS) are more bioaccumulative than the other short-chain PFAA isomers (Hong et al., 2015).

Detailed results of the concentrations of PFAAs in finfish and shellfish for both seasons are presented in **Figure 5-2** and Table B-12 & B-13 (Appendix B). The PFAAs concentrations in seafood varied, depending on the location and the species. In general, the highest levels of total PFAAs were found in Ilish (8.71 ng/g ww in winter, 14.58 ng/g ww in summer) among the finfish species and Crab (7.18 ng/g ww in winter, 8.34 ng/g ww in summer) among the shellfish species, while both of the samples were collected from Chittagong. In finfish, the concentrations of total PFAAs (∑PFAAs) showed the following trend: Ilish > Sole > Rupchanda > Poa > Loitta in both seasons. In particular, considering both seasons, the highest concentrations of PFOA, PFNA, PFDA, PFDoDA and PFTrDA among the finfish species were found in Poa (0.40 ng/g ww) from Chittagong, Sole (1.56 ng/g ww) from Chittagong, Poa (1.20 ng/g ww) from Sundarbans, Ilish (2.01 ng/g ww) from Bhola and Ilish (1.98 ng/g ww) from Chittagong, respectively. The maximum concentrations of PFHxS and PFOS in finfish samples were observed in Ilish (0.69 and 3.86 ng/g ww) from Chittagong. The reasons why Ilish (Hilsa shad) exhibited relatively higher concentrations of PFAAs could be explained by the following phenomena: [1] Ilish is a carnivorous species that tends to concentrate contaminants to a higher degree than other species (Das and Das, 2004); [2] It is an anadromous species which migrates from the sea to the rivers for spawning. Consequently, this species is habitated to different ecosystems (marine, estuarine, brackish and freshwater) and exposed to various degree of contamination; [3] Ilish has relatively a larger body surface area with a very thin layer of skin and a larger gill surface area that facilitate the process of accumulation of the contaminants like PFAAs from water column into the fish body. Butt et al. (2010) reported that PFAAs can undergo bioconcentration via gills in fish although the main uptake is through their diet. Among the shellfish species, the highest concentrations of PFOA, PFNA and PFDA were found in Crab (2.39, 2.11 and 1.34 ng/g ww) from Chittagong, whereas the maximum concentrations of PFDoDA and PFTrDA were found in Crab (0.31 ng/g ww) from Bhola and Shrimp (0.54 ng/g ww) from Cox's

Bazar, respectively. The highest concentrations of PFBS, PFOS and PFDS in shellfish were respectively found in Crabs from Cox's Bazar (1.58 ng/g, ww), Chittagong (1.99 ng/g, ww) and Chittagong (1.08 ng/g, ww) (**Figure 5-2**). In general, the concentrations of total PFAAs (∑PFAAs) in Crabs were higher than that in Shrimps for both seasons. This can be explained as Crab is a typical benthic organism, also known as a scavenger that tends to feed on detritus which is the most contributing factor to the high pollution in crabs (Ip et al., 2005). It also feed on algae and other benthic organisms including mollusks, worms, other crustaceans, fungi and bacteria. The benthic feeders are likely to be exposed to contaminants from a variety of sources such as sediments and pore waters, along with prey species (Oosterom et al., 2010). The bottom dwelling organisms were found to be more prone to bioaccumulation of chemical contaminants than pelagic organisms as sediments have capacity to trap contaminants. Martin et al. (2004) reported that sediments must be a major source of perfluorinated contaminants which may be a result of the sorption of PFAAs to organic matter, followed by sedimentation and subsequent uptake by benthic invertebrates, particularly crabs. Moreover, crabs also possess gills with a relatively larger surface area which could facilitate the accumulation of PFAAs into crab's body since gill can continuously transfer the organic pollutants from both water and suspended particles onto its surface that are subsequently distributed throughout the whole body via blood (Yang et al., 2007).

Patterns of the relative contribution of individual PFAA (% composition) to the total PFAAs in the Bangladeshi seafood are presented in **Figure 5-3**. In finfish, PFOS contributed 11–30% of the total PFAAs followed by PFBS (5–22%), PFDoDA (3–15%), PFTrDA (5–13%) and PFNA (3–10%) whereas in shellfish, PFOS contributed 5–56% followed by PFBS (7–35%) and PFOA (7–29%), while the other PFAAs contributed fewer percentages to the total PFAAs (**Figure 5-3**). The mean concentrations of PFOS and PFOA ranged from 0.64 to 1.23 and 0.5 to 0.08 ng/g ww in finfish, whereas 0.44 to 0.76 and 0.58 to 0.83 ng/g ww in shellfish, respectively. The present study revealed that PFOS was the dominant PFAA in both finfish and shellfish, whereas PFOA was the predominant PFAA in shellfish (**Figure 5-3**) and the similar trends were also observed in many related studies (Gulkowska et al., 2006; Ericson et al., 2008; Ye et al., 2008; Wu et al., 2012; Fang et al., 2014). However, the different composition of PFAAs between finfish and shellfish in the present study might be attributed to the different bioaccumulation potential of PFAAs in different seafood species according to their position in the trophic level, feeding habit, physiological activities and contaminant physical-chemical properties (Gulkowska et al., 2006; Zhou et al., 2007; Wu et al., 2012; Fang et al., 2014). Therefore, more in-depth studies on the bioaccumulation characterization of PFAAs in different seafood species might be fruitful for better understanding of the present results.

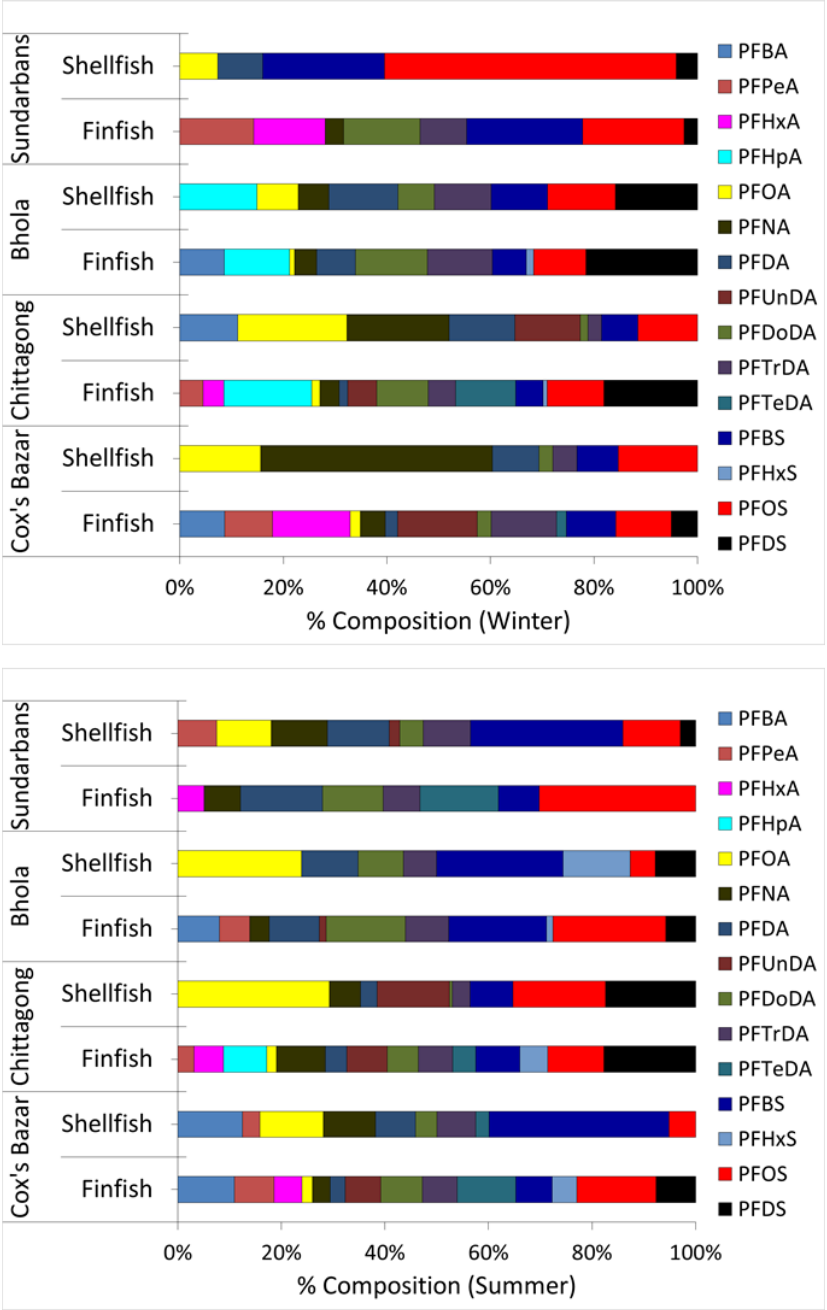

Figure 5-3. Relative contribution of individual PFAAs (% composition) to the total PFAAs in seafood (finfish and shellfish) of the coastal area of Bangladesh in the two seasons.

5.3.2 Distribution of PFAAs in Seafood

The spatial distributions of PFAAs were investigated in seafood collected from the coastal area of Bangladesh and the variations in the average of total PFAAs are presented in **Figure 5-4**. In finfish, the highest mean concentration of total PFAAs were found in samples from Bhola (4.11 ng/g ww) in winter, whereas samples from Chittagong showed the highest value (6.56 ng/g ww) in summer. On the other hand, shellfish samples from Chittagong exhibited the maximum mean levels of total PFAAs in winter (5.14 ng/g ww) and summer (5.47 ng/g ww), respectively. In general, the mean concentration of total PFAAs (ng/g ww) in the investigated seafood seemed to have the following trends: Chittagong > Bhola > Cox's Bazar > Sundarbans, and Chittagong > Cox's Bazar > Bhola > Sundarbans in winter and summer, respectively (Figure B-2, Appendix B).

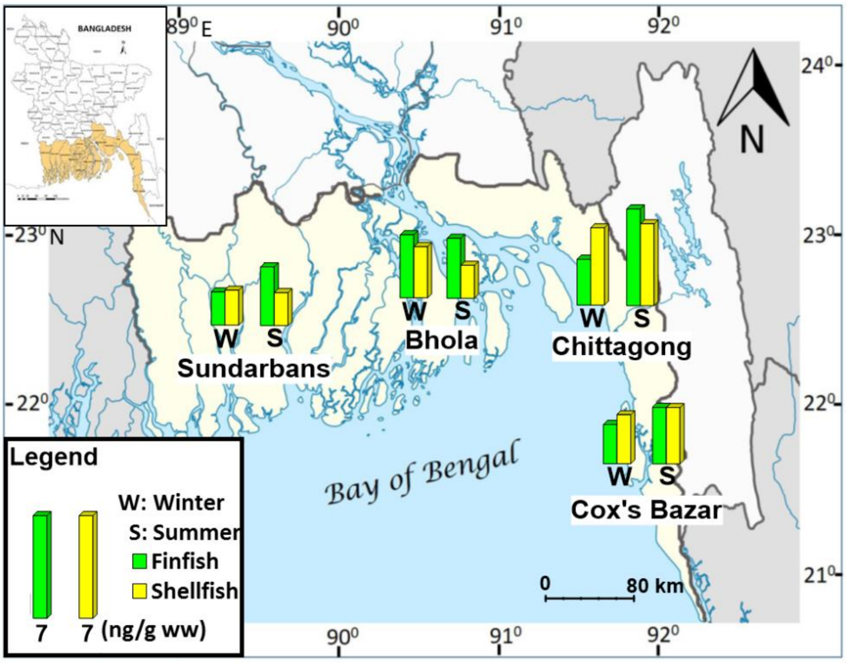

Figure 5-4. Seasonal and spatial variation of average ∑PFAAs in seafood (finfish and shellfish) from the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh.

The level of contamination in seafood with the PFAAs is significantly affected by environmental factors such as water and sediment (Wu et al., 2012), since the aquatic organisms are primarily exposed to the contaminants like PFAAs from water and sediment. The aquatic environment of Chittagong area receives a huge amount of untreated or semi-treated urban, industrial, agricultural wastewater and sludge either directly or indirectly through the Karnaphuli river, the most polluted river in Chittagong division. Also, this area is severely influenced by the port activities, fire training facilities by the fire service department and airport, ship breaking industries and numerous multifarious industries located along the Chittagong coast. The ship breaking activities may contribute to the PFAAs pollution in the adjacent areas as dismantling ships could originate various kinds of inorganic and organic pollutants including PFAAs (Watkinson, 2013). Furthermore, the coastal area of Cox's Bazar is also getting polluted day by day due to unplanned urbanization and industrialization in recent years. Municipal and industrial wastewaters without any treatment are being discharged directly to the Bay of Bengal through the Bakkhali Estuary. As a consequence, water and sediment contaminated by PFAAs might be attributed to the higher mean concentrations of ∑PFAAs in seafood from these areas. However, the mean ∑PFAAs concentrations in seafood showed that high concentrations are not only found in locations in proximity to the fastest growing industrial areas, as is the case of Chittagong and Cox's Bazar, but might also occur in less industrialized areas. This is clearly the case for seafood samples from Bhola which is an unindustrialized or less industrialized and exclusively an estuarine area (Meghna Estuary). It should be noted, however, that a large volume of municipal and industrial effluents from the major cities of the middle and upper part of the country through numerous inland rivers finally find their way to the Bay of Bengal in the Meghna Estuary, and thus polluting the environmental matrices (water, sediment and biota) in this area. In general, the present study revealed that the seafood of the southeast part (Chittagong and Cox's Bazar) of the Bangladeshi coastal area was more contaminated with PFAAs than the south (Bhola, Meghna Estuary) and southwest part (Sundarbans) because of higher intensity of recent developments in the coastal areas of Bangladesh which are characterized by the modern industrialization and urbanization activities.

5.3.3 Correlation analysis

Spearman rank correlations ($p < 0.01$) among the individual PFAAs for both the finfish and shellfish and all seafood samples are presented in Table B-14 (Appendix B). PFNA/PFDA, PFUnDA/PFDS, PFTrDA/[PFDoDA, PFTeDA], PFOS/[PFDoDA, PFTrDA], PFBS/[PFTrDA, PFTeDA] in finfish, and PFOA/[PFNA, PFUnDA], PFDoDA/[PFDA, PFTrDA], PFOS/PFBS in shellfish, whereas PFOA/[PFNA, PFDA], PFUnDA/PFDS, PFTrDA/[PFDoDA, PFOS], PFBS/[PFTeDA, PFOS] (considering both the finfish and shellfish samples as a whole, i.e. seafood) were found to be significantly correlated ($p < 0.01$), with the correlation coefficient of (0.502–0.715), which suggested that these compounds probably had a common pollution source in the same region while the others might be originated from different or multiple sources.

5.3.4 Comparison with previous studies worldwide

In the environmental research, comparison of monitoring data with other related studies is a very insightful and informative attempt/task which could give an overall idea about the degree of contamination and sources of the contaminants as well, particularly for a newly explored study. Contrary, species to species comparison is very difficult since the same species might not be available everywhere around the world. The distribution of a particular species is solely dependent on the geographical conditions. In this case, we could consider the category or sub-category of species rather than a single species, e.g. seafood (finfish and shellfish). Moreover, the fate and distribution of chemical contaminants like PFAAs might be influenced by the geomorphological characteristics or climatic conditions of the aquatic environments of different regions, such as tropical, sub-tropical and temperate. Taking these into account, we tried to make a rough comparison of the levels of PFAAs in seafood samples of the present study with the reported data worldwide and summarized in **Table 5-2**. In general, **Table 5-2** reveals that PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFHxS, and PFOS are the compounds that could be detected more frequently in seafood samples around the world, while PFOS and PFOA were the most abundant PFAAs. The concentration range of PFOS in finfish (0.10–3.86 ng/g ww) from our research was comparable or lower than those observed in Baltic sea, Sweden (Berger et al., 2009), Mediterranean finfish (Vassiliadou et al., 2015), Ohio, Missouri, and Mississippi rivers, USA (Ye et al., 2008), Canadian marine fish (Tittlemier et al., 2007), Guangzhou and Zhoushan (Gulkowska et al., 2006), but higher than that from Catalan market, Spain (Ericson et al., 2008), Hong Kong (Zhao et al., 2011), Nunavut in Canada (Ostertag et al., 2009), and coastal areas of China (Wu et al., 2012). Moreover, the levels of PFOS in shellfish (0.10–1.99 ng/g ww) were comparable or less than those measured from Mediterranean shellfish (Vassiliadou et al., 2015), retail shellfish in USA (Young et al., 2013), Guangzhou and Zhoushan (Gulkowska et al., 2006), municipal market in China (Zhang et al., 2011), but higher than that from coastal areas of Catalonia (Dominigo et al., 2012), Nunavut in Canada (Ostertag et al., 2009), coastal areas of China (Wu et al., 2012) and Catalan market, Spain (Ericson et al., 2008).

Table 5-2. Comparison of PFAAs concentrations (ng/g ww) in seafood (finfish and shellfish) samples of the present study with the reported data worldwide.

Sampling site	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	References
Finfish																
Baltic Sea, Sweden	–	–	–	–	<0.10–0.39	<0.08–0.47	<0.08–0.34	<0.08–0.61	<0.08–0.15	<0.10–0.38	<0.15	–	<0.02–0.20	0.47–3.34	–	Berger et al., 2009
L. Vättern, Sweden	–	–	–	–	<0.10–0.25	<0.08–0.71	0.13–0.81	0.09–0.89	<0.08–0.53	<0.10–1.83	<0.15–0.65	–	<0.02–0.80	0.97–23.1	–	
Catalan Market, Spain	–	–	–	<0.002	<0.065	–	–	–	–	–	–	–	–	0.271–0.654	–	Ericson et al., 2008
Mediterranean finfish (Greece)	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD–0.60	<LOD–0.65	<LOD–1.50	<LOD–1.86	<LOD	<LOD	<LOD–0.45	<LOD	<LOD–20.37	<LOD	Vassiliadou et al., 2015
Hong Kong marine fish	–	–	–	–	<LOD	<LOD–0.65	<LOD	<LOD–0.71	<LOD	<LOD–0.75	–	–	–	<LOD–1.5	–	Zhao et al., 2011
Three rivers of USA	–	–	<4.00–18.4	<0.20–4.03	<0.20–2.10	<0.20–5.89	<0.20–9.01	<0.40–48.0	<0.20–4.13	–	–	<0.20–0.64	<0.20–8.14	<10.0–1250	–	Ye et al., 2008
Marine fish, Canada	–	–	–	<0.4	<0.5	<1	<2	<1	<0.8	–	<5	–	–	2.6	–	Tittlemier et al., 2007
Inuit in Nunavut (Canada)	–	–	–	–	<0.2	–	–	–	–	–	–	–	–	<0.5	–	Ostertag et al., 2009
Guangzhou and Zhoushan, China	–	–	<0.25	<0.25	<0.25	<0.25	<0.25	0.35–0.65	–	–	–	–	<0.25	0.38–2.93	–	Gulkowska et al., 2006
Coastal areas in China	–	–	–	–	<0.005–7.543	<0.003–0.588	–	<0.002–1.126	–	–	–	–	–	<0.001–1.627	–	Wu et al., 2012
Coastal area, Bangladesh	<LOD–0.72	<LOD–0.80	<LOD–0.73	<LOD–1.12	<LOD–0.40	<LOD–1.56	<LOD–1.20	<LOD–0.88	<LOD–2.01	<LOD–1.98	<LOD–1.34	<LOD–1.47	<LOD–0.69	0.10–3.86	<LOD–2.02	*This study*
Shellfish																
Coastal areas of Catalonia (Spain)	–	–	<LOD	<LOD	<0.068–0.098	0.03–0.26	<LOD	<0.045–0.22	<LOD	<LOD	<LOD	<LOD	<LOD	<0.045–0.54	<LOD	Dominigo et al., 2012
Catalan Market, Spain	–	–	–	<0.002	<0.029	–	–	–	–	–	–	–	–	0.148	–	Ericson et al., 2008
Mediterranean shellfish (Greece)	<LOD	<LOD–4.94	<LOD	<LOD	<LOD	<LOD–1.27	<LOD–1.73	<LOD–2.76	<LOD–1.36	<LOD	<LOD	<LOD–1.37	<LOD	<LOD–5.15	<LOD	Vassiliadou et al., 2015
Retail shellfish, USA	–	–	–	–	–	<LOD–1.2	<LOD–3.40	<LOD–6.54	<LOD	–	–	–	<LOD	<LOD–6.29	–	Young et al., 2013
Inuit in Nunavut (Canada)	–	–	–	–	<0.4	–	–	–	–	–	–	–	–	<0.2	–	Ostertag et al., 2009
Guangzhou and Zhoushan, China	–	–	<0.25–0.29	<0.25	<0.25–1.67	<0.25–0.61	<0.25–0.30	0.27–0.93	–	–	–	–	<0.25	0.33–13.9	–	Gulkowska et al., 2006
Municipal market in China	–	–	<0.10	<0.10–0.32	<0.10–1.68	<0.10–0.49	<0.10–0.81	<0.10–2.94	<0.10–0.37	–	–	–	<0.10–0.13	<0.10–1.94	–	Zhang et al., 2011
Coastal areas in China	–	–	–	–	<0.005–7.543	<0.002–0.588	–	<0.002–0.509	–	–	–	–	–	<0.001–0.593	–	Wu et al., 2012
Coastal area, Bangladesh	<LOD–0.66	<LOD–0.19	<LOD	<LOD–0.66	<LOD–2.39	<LOD–2.11	<LOD–1.34	<LOD–1.17	<LOD–0.31	<LOD–0.54	<LOD–0.12	<LOD–1.58	<LOD–0.36	0.10–1.99	<LOD–1.08	*This study*

In the present study, PFOA level in finfish (<LOD–0.40 ng/g ww) was comparable or lower than those reported from L. Vättern and the Baltic Sea, Sweden (Berger et al., 2009), Catalan market, Spain (Ericson et al., 2008), Mediterranean finfish (Vassiliadou et al., 2015), Hong Kong marine fish (Zhao et al., 2011), Ohio, Missouri, and Mississippi rivers, USA (Ye et al., 2008) and coastal areas of China (Wu et al., 2012).

Furthermore, the concentration range of PFOA in shellfish (<LOD–2.39 ng/g ww) from our study was higher than that of coastal areas of Catalonia (Dominigo et al., 2012), Guangzhou and Zhoushan (Gulkowska et al., 2006), municipal market in China (Zhang et al., 2011), Catalan market, Spain (Ericson et al., 2008), but lower than the level of PFOA observed in shellfish (<0.005–7.543 ng/g ww) from coastal areas of China (Wu et al., 2012) (**Table 5-2**). The concentration of PFNA and PFDA in the present study was higher than those measured in L. Vättern and the Baltic Sea, Sweden (Berger et al., 2009), Mediterranean seafood (Vassiliadou et al., 2015), Hong Kong marine fish (Zhao et al., 2011), coastal areas of China (Wu et al., 2012), coastal areas of Catalonia (Dominigo et al., 2012), retail fish in USA (Young et al., 2013), Guangzhou and Zhoushan (Gulkowska et al., 2006), municipal market in China (Zhang et al., 2011), while lower than that of Ohio, Missouri, and Mississippi rivers, USA (Ye et al., 2008). Moreover, the concentrations of PFUnDA, PFDoA and PFHxS in the present study were comparable to or lower than those reported in other studies (Berger et al., 2009; Vassiliadou et al., 2015; Zhao et al., 2011; Ye et al., 2008; Gulkowska et al., 2006; Wu et al., 2012; Dominigo et al., 2012; Zhang et al., 2011) (**Table 5-2**). The information about the concentrations of other PFAAs (e.g. PFBA, PFPeA, PFHxA, PFHpA, PFTrDA, PFTeDA, PFBS, and PFDS) in seafood (finfish and shellfish) from other studies was scarce (**Table 5-2**). However, as mentioned above, it should be taken into account that the characteristics and habitats or position in the trophic levels for different species may be attributed to different bioaccumulation levels of a particular PFAA compound. To address this phenomenon, more comprehensive studies would be necessary by giving emphasis to the source- or compound-, or concentration-specific bioaccumulations of PFAAs against various habitats and environments. Wu et al. (2012) also concluded that the environmental concentrations of PFAAs in different countries were not comparable, which could be considered as another factor for the difference of the comparisons mentioned above.

5.3.5 *Exposure assessment of dietary PFAAs from seafood consumption*

Seafood (finfish and shellfish) consumption has been suggested as the major source of human dietary PFAAs exposure (Ericson et al., 2008; Berger et al., 2009; Hlouskova et al., 2013). Haug et al. (2010) reported that the intake of seafood was the largest contributor to the total dietary intake of PFAAs, representing 81% for PFOS and 38% for PFOA of the estimated daily intake. To assess potential impacts on human health resulting from the consumption of contaminated seafood a risk assessment based upon a defined tolerable daily intake (TDI) is needed. Although there are no internationally agreed TDIs for PFAAs, some suggested values for PFOS and PFOA are still available that have been proposed by several national or international organizations. The UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT) recommended a TDI of 300 ng/kg for PFOS, while for PFOA, a TDI of 3000 ng/kg was suggested (COT, 2006). On the other hand, oral reference dose (RfD) values for most PFAAs have not been established yet by any government or regulatory agency. However, provisional RfDs for PFOS and PFOA have been estimated on the basis of a rat chronic carcinogenicity study and a rat multigenerational study, respectively. On this basis, the provisional RfDs would be 25 and 333 ng/kg/day for PFOS and PFOA, respectively (Thayer, 2002; Gulkowska et al., 2006). Furthermore, for PFOS, the Scientific Panel on Contaminants in the Food Chain (CONTAM) identified 30 µg/kg-bw/day as the lowest no-observed-adverse-effect level (NOAEL) (effect: changes in lipids and thyroid hormones) from a subchronic study in Cynomolgus monkeys to derive a provisional TDI of 150 ng/kg-bw/day by applying an overall uncertainty factor (UF) of 200 to the NOAEL (EFSA, 2008). Moreover, after reviewing a number of toxicological studies on PFOA, the CONTAM Panel established a TDI for PFOA of 1500 ng/kg/day by applying an overall UF of 200 to the lower confidence limit of the benchmark dose for a 10% effect size ($BMDL_{10}$) of 0.3 mg/kg/day which was derived from a subchronic study in male rats with the endpoints of hepatocellular hypertrophy and increased liver weight (EFSA, 2008). The recommended TDIs for PFOS and PFOA (EFSA, 2008) of 150 and 1500 ng/kg/day, respectively, are remarkably higher than the suggested provisional RfDs.

Dietary intake is an imperative approach for the exposure assessment of PFAAs to human. The estimated daily intake (EDI) (ng/kg-bw/day) of PFOA, PFOS and total PFAAs via seafood consumption were calculated according to the following equation:

$$EDI=\frac{C\times FCR}{ABW} \qquad (5.1)$$

where C is the concentration (ng/g ww) of PFOA, PFOS and total PFAAs in seafood (finfish and shellfish) samples, FCR is the seafood consumption rate (g/day, wet weight basis), and ABW is the average body weight (60 kg for adults and 25 kg for children). The daily consumption data of seafood for the adults (≥18 years) and children (6–17 years) in the coastal area of Bangladesh were obtained from a questionnaire based dietary survey during our sampling campaign. The seafood consumption data and the estimated daily intake (EDI) of PFOA, PFOS and total PFAAs by the adults and children in the coastal area of Bangladesh for both seasons (winter and summer) are presented in **Table 5-3**.

Table 5-3. The estimated daily intake (EDI) of PFOA, PFOS and total PFAAs via seafood consumption for the coastal residents (adults and children) of Bangladesh.

Location	Type	Mean concentration (ng/g ww)			Seafood consumption[a] (g/day)		EDI (ng/kg–bw/day)					
							Adult[b]			Children[b]		
		PFOA	PFOS	Total PFAAs	Adult	Children	PFOA	PFOS	Total PFAAs	PFOA	PFOS	Total PFAAs
Winter												
Cox's Bazar	Finfish	0.04	0.52	2.57	85	39	0.06	0.74	3.65	0.07	0.81	4.01
	Shellfish	0.62	0.61	3.79	21	12	0.22	0.21	1.33	0.30	0.29	1.82
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.28	0.95	4.97	0.36	1.10	5.83
Chittagong	Finfish	0.11	0.73	3.08	87	42	0.16	1.05	4.47	0.18	1.22	5.18
	Shellfish	1.21	0.66	6.70	24	14	0.48	0.26	2.68	0.68	0.37	3.75
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.64	1.32	7.15	0.86	1.59	8.93
Bhola	Finfish	0.03	0.69	4.28	67	31	0.03	0.77	4.78	0.03	0.85	5.30
	Shellfish	0.36	0.59	4.04	18	10	0.11	0.18	1.21	0.14	0.23	1.62
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.14	0.94	5.99	0.18	1.08	6.92
Sundarbans	Finfish	0.01	0.64	2.15	69	33	0.01	0.74	2.47	0.01	0.85	2.84
	Shellfish	0.15	1.18	3.97	19	11	0.05	0.37	1.26	0.07	0.52	1.75
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.06	1.11	3.73	0.08	1.37	4.59
Summer												
Cox's Bazar	Finfish	0.11	1.01	3.29	88	41	0.16	1.48	4.82	0.18	1.65	5.39
	Shellfish	0.56	0.23	3.73	19	13	0.18	0.07	1.18	0.29	0.12	1.94
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.33	1.55	6.00	0.47	1.77	7.33
Chittagong	Finfish	0.13	1.24	5.36	85	44	0.18	1.75	7.60	0.23	2.18	9.44
	Shellfish	1.82	1.12	5.64	22	15	0.67	0.41	2.07	1.09	0.67	3.38
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.85	2.16	9.67	1.32	2.85	12.82
Bhola	Finfish	0.01	1.17	3.43	70	33	0.01	1.37	4.00	0.01	1.55	4.53
	Shellfish	0.67	0.13	2.12	16	13	0.18	0.04	0.56	0.35	0.07	1.10
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.19	1.41	4.57	0.36	1.62	5.63
Sundarbans	Finfish	0.01	1.64	2.28	71	35	0.01	1.94	2.70	0.01	2.30	3.19
	Shellfish	0.27	0.28	2.08	21	11	0.09	0.10	0.73	0.12	0.13	0.91
	EDI$_{(\mathit{Finfish+Shellfish})}$						0.10	2.04	3.42	0.13	2.42	4.10

[a] Data obtained from dietary questionnaire survey; [b] Average body weight was 60 kg for adults and 25 kg for children.

No obvious seasonal variation was found concerning the seafood consumption rate for the Bangladeshi coastal residents ($p > 0.05$). The data revealed that the EDI of PFOA was relatively higher from shellfish consumption and the EDI of PFOS was higher from finfish consumption for both the adults and children (**Table 5-3**), while the differences in EDI values were related to the amounts of seafood consumed as well as the concentrations of PFOA and PFOS in the investigated samples. In general, the EDIs (ng/kg-bw/day) of PFOA and PFOS from seafood consumption ($EDI_{Finfish+Shellfish}$) ranged from 0.06 to 0.85 and 0.94 to 2.16 for adults, whereas it was 0.08 to 1.32 and 1.08 to 2.85 for children, respectively (**Table 5-3**). Thus, the children of Bangladeshi coastal area were exposed slightly higher to the dietary PFAAs in seafood than that for the adults, which might be due to the relatively lower average body weight for the children since the EDI values were calculated for per kg body weight of the consumers. Furthermore, the highest EDI of PFOA, PFOS and total PFAAs were found in Chittagong area for both the adults and children in the both seasons (**Table 5-3**), elucidating that the coastal residents in the Chittagong area were more prone to PFAA burden health hazard from seafood consumption.

Table 5-4. The estimated daily intake (EDI) of PFOA and PFOS in seafood for the adults from other studies.

Study area	Year	EDI (ng/kg-bw/day)		Reference
		PFOA	PFOS	
Guangzhou (China)	2004	0.514	3.22	Gulkowska et al., 2006
Zhoushan (China)	2004	0.6	2.13	Gulkowska et al., 2006
Tarragona County (Catalonia, Spain)	2006	-	0.541	Ericson et al., 2008
Baltic Sea (Sweden)	2001	-	0.15–0.62	Berger et al., 2009
Coastal and inland areas (Norway)	2003	0.6–1.4	4.7–5.9	Haug et al., 2010
Tianjin Binhai New Area (China)	2008	-	20.6–62.5	Chen et al., 2011
Hong Kong	2009	3.3 ± 0.54	2.4 ± 2.9	Zhao et al., 2011
Tianjin, Nanchang, Wuhan, and Shenyang (China)	2009	0.13–0.28	0.10–1.58	Zhang et al., 2011
Coastal areas in China	2007	0.008–0.914	0.037–0.694	Wu et al., 2012
Attika (Greece)	2011	0.05–0.20	0.42–10.48	Vassiliadou et al., 2015
Coastal area, Bangladesh	2015	0.06–0.85	0.94–2.16	*This study*

Note: Sign ‘-’ denotes ‘not available’.

The EDI (ng/kg-bw/day) of PFOA and PFOS in seafood for adults from other related studies worldwide are shown in **Table 5-4**. The results from the present study revealed that the $EDI_{Finfish+Shellfish}$ of PFOA for the Bangladeshi coastal adults were comparable to or lower than the values reported from Norway (Haug et al., 2010), Hong Kong (Zhao et al., 2011), and coastal areas in China (Wu et al., 2012), but higher than the data obtained from

Guangzhou and Zhoushan in China (Gulkowska et al., 2006), Tianjin, Nanchang, Wuhan, and Shenyang in China (Zhang et al., 2011), and Greece (Vassiliadou et al., 2015). On the other hand, the $EDI_{Finfish+Shellfish}$ of PFOS were higher than the EDI values reported from Spain (Ericson et al., 2008), Sweden (Berger et al., 2009) and China (Zhang et al., 2011; Wu et al., 2012), but similar to or lower than that from Zhoushan in China (Gulkowska et al., 2006), Norway (Haug et al., 2010), Hong Kong (Zhao et al., 2011), and Greece (Vassiliadou et al., 2015). Furthermore, Wu et al. (2012) reported an EDI of total PFAAs from seafood consumption at the range of 0.115 to 2.513 pg/kg-bw/day for the adults living in the coastal areas in China which were much lower than the results from our study. There was little information about the EDI of PFAAs in seafood for the children from other studies. However, Zhang et al. (2011) reported EDIs of PFOA (0.17 to 0.37 ng/kg-bw/day) and PFOS (0.13 to 2.11 ng/kg-bw/day) for the children (6 to 17 years) from consumption of fish and seafood in three cities of China (Nanchang, Wuhan, and Shenyang) which were comparable or slightly lower than our results (PFOA: 0.08 to 1.32 ng/kg-bw/day; PFOS: 1.08 to 2.85 ng/kg-bw/day). The major reasons to explain the discrepancy of EDIs in the different researches mentioned above are related to the amounts of seafood consumed, PFAAs levels measured in seafood samples as well as variation in seafood species investigated from each study.

The present study revealed that the EDI of PFOA and PFOS through seafood consumption were much lower than the EFSA recommended TDIs for PFOA and PFOS at 1500 ng/kg/day and 150 ng/kg/day, respectively (EFSA, 2008). Furthermore, assuming the toxicological concern for the other PFAAs (e.g. PFCAs: PFBA, PFPeA, PFHxA, PFHpA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, and PFTeDA; PFSAs: PFBS, PFHxS, and PFDS) similar to the health hazard of dietary PFOA or PFOS, the EDIs of total PFAAs through seafood consumption by the Bangladeshi coastal residents (3.42–9.67 ng/kg-bw/day for adults and 4.10–12.82 ng/kg-bw/day for children) are still far below the EFSA recommended TDIs or other guidelines currently available as mentioned above. Therefore, this study elucidated that the coastal residents of Bangladesh were not significantly exposed to PFAAs via the consumption of seafood yearly. However, the data mentioned above were calculated from the mean estimated daily intakes. Thus, the potential health risk should not be ignored for the residents with much higher seafood consumption.

5.4 Conclusion

This study provides national baseline data on the contamination status of PFAAs in several species of commonly consumed seafood from the coastal area of Bangladesh collected in winter and summer, 2015. The results indicated that there still existed a variety of PFAAs in seafood. PFOS was the most abundant PFAA in all samples. The majority of the monitored PFAAs did not show clear seasonal variation. However, the levels and composition profiles of PFAAs in seafood varied depending on the location and the species. The seafood from southeast part of the Bangladeshi coastal area (Cox's Bazar and Chittagong) exhibited relatively higher levels of PFAAs than that from the south (Bhola) and southwest part (Sundarbans). Among the seafood species, Ilish (hilsa sad) in the finfish group and Crab (mud crab) in the shellfish group showed the highest concentrations of total PFAAs, both of which were collected from Chittagong. According to the result from the dietary exposure assessment, most of the coastal residents in Bangladesh were not significantly exposed to PFAAs burden health hazard from seafood consumption depending on the recommended levels of dietary intake at least for those PFAAs for which information is currently available. Since this is a preliminary health risk assessment, investigations should be promoted by performing with larger numbers of species with different origins, and gender, age, occupation and eating habit of the consumers need to be taken into consideration. In addition, the elevated level of PFAAs in seafood as well as relatively higher dietary PFAAs exposure from seafood consumption in the southeast coastal area of Bangladesh warrants an attention that should be paid to the environment quality in this rapidly industrialized and urbanized area with more studies on the sources, fate and distribution of PFAAs into the coastal aquatic environment and overall, the countermeasures to improve the environmental quality in the study area is recommended.

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Chapter 6

POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN SURFACE WATER, SEDIMENT AND SEAFOOD FROM COASTAL AREA OF BANGLADESH: LEVELS, DISTRIBUTION AND RISK ASSESSMENT

Polycyclic aromatic hydrocarbons (PAHs) in surface water, sediment and seafood from coastal area of Bangladesh: levels, distribution and risk assessment

Abstract

Levels, distribution, and potential risks of 16 USEPA priority polycyclic aromatic hydrocarbons (PAHs) in the surface water, sediment, and seafood from the coastal areas of Bangladesh were investigated. Samples were collected in winter and summer, 2015. The total concentration of PAHs (∑PAHs) in surface water, sediment, and seafood were 855.4–9653.7 ng/L, 349.8–11058.8 ng/g dry weight (dw) and 184.5–2806.6 ng/g wet weight (ww) in winter, and 679.4–12639.3 ng/L, 199.9–17089.1 ng/g dw and 117.9–4216.8 ng/g ww in summer, respectively. The levels of ∑PAHs were comparable or relatively high in comparison with other coastal areas around the world. No significant seasonal variation was observed in the levels of ∑PAHs in either medium. Spatial distribution revealed that the areas with recent urbanization and industrialization (Chittagong, Cox's Bazar and Sundarbans) were more contaminated with PAHs than the unindustrialized area (Meghna Estuary). Molecular ratios suggested mixed sources of PAHs in the Bangladeshi coastal area with a slight imposition of pyrolytic over the petrogenic inputs closely related to shipping and fishing activities, and industrial and municipal sewage discharge. From the ecotoxicological point of view, PAH concentrations in the Bangladeshi coastal area exceeded some of the existing national and international environmental quality guidelines/standards, suggesting potential threat to the aquatic organisms and to the human health as well through biomagnification. In addition, the coastal residents are sufficiently exposed to the dietary PAHs through seafood consumption which would obviously cause severe health risk including carcinogenic effects. The results from this study may be useful for the design of appropriate risk communication strategies in Bangladesh.

6.1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of persistent organic pollutants formed by two or more fused aromatic rings of carbon and hydrogen atoms. They are ubiquitous in the environment due to their persistence, long range transport, bioaccumulation and known to be toxic, carcinogenic and mutagenic (Lotufo and Fleeger,1997; Wan et al., 2007; Callén et al., 2013; Gu et al., 2013). Based on the evidences of their high toxicological risk, 16 selected PAHs have been listed as priority pollutants by the United States Environmental Protection Agency (USEPA), seven of which are potentially carcinogen according to the International Agency for Research on Cancer (IARC).

PAHs are generally formed by natural processes and anthropogenic activities (Abdel-Shafy and Mansour, 2016) and introduced into the environments via various routes (Durand et al., 2004; Xu et al., 2006). Environmental PAHs are mainly from two sources: petrogenic source, which is composed by PAHs associated with spills of crude and refined oil, and pyrogenic source, which includes compounds formed by fossil fuel combustion, waste incineration, biomass burning, and asphalt production (Zheng et al., 2016). Furthermore, it should be taken in mind that the source profiles and spatial distributions of different sources of PAHs in the environment may vary seasonally (Deshmukh et al., 2013).

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 (Oros et al., 2007; Counihan et al., 2014; Yancheshmeh et al., 2014; Zhang et al., 2016 and references therein). PAHs enter the marine environment through several mechanisms including discharge of industrial and municipal sewage, marine transport, shipping and fishing activities, ship building and ship breaking activities, terrestrial surface and riverine runoff, atmospheric deposition, and petroleum spills, etc. Owing to their hydrophobic properties, PAHs in the water body tend to be absorbed rapidly by suspended particulate matter and to be subsequently deposited on sediments, which can be generally regarded as a sink for PAHs. In addition, PAHs are also readily taken up and bio-accumulated by marine organisms, and affect negatively not only the growth and reproduction of native wildlife species, but also human health via consumption of contaminated seafood (Gu et al., 2013; Bandowe et al., 2014; Zhao et al., 2014; Sun et al., 2016). Therefore, PAHs have always been of great concern, and it is very important to monitor their pollution levels and to evaluate their potential toxicity in the environment.

Bangladesh is an exclusively riverine agricultural country that is undergoing rapid industrialization, urbanization and economic development in recent years, particularly in the coastal regions. The country has a highly irregular deltaic marshy coastline of 580 kilometers, divided by many rivers and streams that enter the Bay of Bengal, which is characterized by a tropical climate. The coastal areas of Bangladesh suffers from environmental degradation due to rapid human settlement, the development of industrial hubs, tourism and transportation, extensive ship breaking and port activities, illegal and mismanaged dumping of e-waste, the operation of an excessive number of mechanized boats and fishing trawlers, deforestation, and increasing agriculture and aquaculture activity and large discharges of untreated and semi-treated domestic and municipal sewage. There are also effluents containing heavy loads of organic and inorganic pollutants from many large and small local multipurpose industries and factories. With the increase of population and industrial pressures, the coastal areas of Bangladesh may face threats from contaminants such as PAHs, which can accumulate in the coastal or marine food chains. Besides the adverse impact on the ecological integrity, local populations can also be exposed to these chemicals via contaminated seafood consumption. About 42 million people (30 % of the total population) live in the coastal area (47,211 km^2; 32 % of the total land area) of Bangladesh, of which about 5 million are engaged directly in commercial fishing (BOBLM, 2011). Seafood is the most important and one of the major dietary components of the coastal populations. Therefore, it is an urgent need to assess the potential health risk that might be posed through PAH-contaminated seafood consumption.

In the last few decades, environmental PAHs and their distribution, sources and environmental risk to ecological systems including humans have been extensively studied in the coastal regions worldwide (Wang et al., 2001; Nakata et al., 2003; Lim et al., 2007; Horii et al., 2009; Moon et al., 2010; Yim et al., 2014; Sun et al., 2016; Ohiozebau et al., 2016; Zheng et al., 2016; Li et al., 2016; Goswami et al., 2016). However, there are few studies monitoring concentrations of PAHs in the Bangladeshi environments (Zuloaga et al., 2013; Nøst et al., 2015), and no comprehensive studies have been conducted so far in this country. Hence, we aimed to investigate the levels, distribution, and sources of PAHs in surface water, sediments and commonly consumed seafood in the coastal area of Bangladesh. The ecological and human health risk assessment of PAHs was also conducted to provide scientific data for organic pollution control of the Bangladeshi coastal areas. Furthermore, the data from this study may be helpful in policy making on sustainable development and marine environmental conservation in the Bay of Bengal.

6.2 Materials and Methods

6.2.1 Study area and collection of samples

A sampling map with sampling sites and locations is shown in **Figure 6-1**. A total of 28 surface water, 28 sediment, and 48 seafood samples were collected in winter and summer 2015. The seafood species are commonly consumed by the Bangladeshi coastal residents which includes 5 finfish [Ilish (*Tenualosa ilisha*), Rupchanda (*Pampus argentius*), Loitta (*Harpadon nehereus*), Sole (*Cynoglossus lingua*), and Poa (*Otolithoides pama*)] and 2 shellfish [Shrimp (*Penaeus indicus*) and Crab (*Scylla serrata*)]. For detailed description of the study area and collection of samples, please see section 3.2.1 (Chapter 3) and Appendix A.

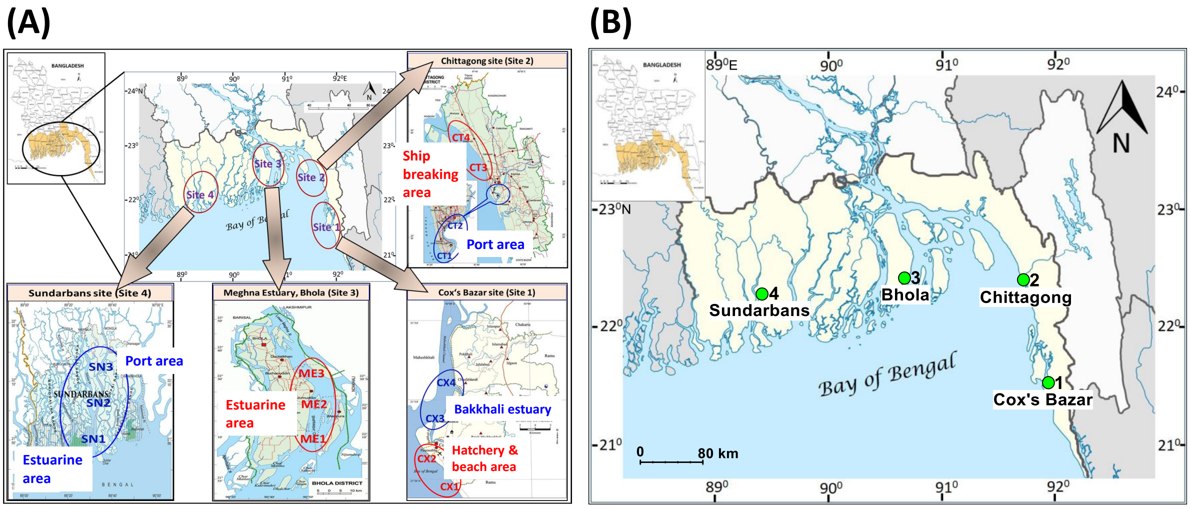

Figure 6-1. Maps showing (A). four sampling sites with 14 sampling locations in the coastal area of Bangladesh from where surface water and sediment samples were collected; (B). four main fish landing centers in the coastal areas of Bangladesh from where seafood samples were collected- (1): Fish Landing and Wholesale Fish Market, Cox's Bazar; (2): Chittagong fishing harbor, Chittagong; (3): Fish Landing and Wholesale Fish Market, Bhola; (4): Fish Processing and Marketing Centre, Sundarbans area in Mongla. Yellow colored area in the inset map represents the coastal area of Bangladesh.

6.2.2 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 Pure Chemical Ind. (Osaka, Japan). Milli-Q (>18.2 MΩ) water was used throughout the experiment and was generated by using an ultrapure water purification system (Millipore, Billerica, MA, USA). Filter membranes (0.45 μm, 47 mm i.d.) were obtained from ADVANTEC® (Tokyo, Japan).

6.2.3 Sample pretreatment

Surface 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 aliquots of dichloromethane, then twice by 10 mL aliquots of methanol and then 10 mL distilled water. One liter of filtered water samples was mixed well and trapped through the SPE tubes with a flow rate at 10 mL/min under vacuum. After the extraction, the cartridges were dried under vacuum for 10 min. The cartridges were then transported in dark, air-tight containers to the Laboratories of Yokohama National University in Japan for elution and analysis of PAHs. 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.

Freeze-dried sediment samples were homogenized with a silica mortar and pestle, then sieved through a 2-mm mesh sieve to remove debris and remove the coarse fraction (> 2 mm), which has low or negligible binding capacity for many contaminants (IAEA, 2003). After that, they were extracted by a Quick, Easy, Cheap, Effective, Rugged and Safe (QuEChERS) method validated by Salem et al. (2016) following some modifications. Briefly, an aliquot of 5 g of sediment was weighed into a polypropylene tube (50 mL capacity). Then, 5 mL of ultrapure water was added, the tube was manually shaken and spiked with 100 µL of 500 ng/mL of Ace-D_{10} and BaP-D_{12} as internal standards (IS) for quantification. Fifteen (15) mL of extraction solvent (hexane:acetone:dichloromethane ≈ 1:1:1, v/v/v) and two ceramic bars (Agilent p/n 5982–9313) were added to aid in sample extraction and the tube was shaken vigorously by hand for 5 min. Afterwards, the QuEChERS salts ((4 g of magnesium sulfate ($MgSO_4$), 1 g of sodium chloride (NaCl), 1 g of trisodium citrate dihydrate ($Na_3C_6H_5O_7$) and 0.5 g of disodium hydrogen citrate sesquihydrate ($C_6H_8Na_2O_8$); Agilent p/n 5982–6650) was then added and the tube was immediately shaken for 1 min to avoid agglomeration of salts followed by ultrasonic agitation for 20 min. Finally, the tube was centrifuged for 3 min at 3500 rpm and 10 mL of the supernatant were transferred to a QuEChERS dispersive solid phase extraction (d-SPE) clean-up tube (15 mL) containing 0.9 g of anhydrous magnesium sulfate, 0.15 g of primary secondary amine (PSA) and 0.15 g of C18EC (Agilent p/n 5982–5156). One ceramic bar (Agilent p/n 5982–9312) was added and the tube was shaken vigorously by hand for 1 min followed by centrifugation for 3 min at 3500 rpm. The extracts were then treated with activated copper for desulphurization and transferred into a glass test tube and then evaporated to near dryness under a gentle stream of high-purity nitrogen. The residue was finally re-dissolved in 1 mL n-hexane and stored at −20 °C until GC-MS/MS analysis.

Sample preparation for the extraction of PAHs from seafood involves a modified QuEChERS method. Briefly, an aliquot of 5 g of homogenized sample was weighed into a polypropylene tube (50 mL capacity). Then, 5 mL of ultrapure water was added, the tube was manually shaken and spiked with 100 µL of 500 ng/mL of Ace-D_{10} and BaP-D_{12} as an internal standards (IS) for quantification. Ten (10) mL of extraction solvent (hexane:acetone ≈ 1:1, v/v) and two ceramic bars (Agilent p/n 5982–9313) were added to aid in sample extraction and the tube was shaken vigorously by hand for 5 min. Afterwards, the QuEChERS salts (6 g of $MgSO_4$ and 1.5 g of sodium acetate; Agilent p/n 5982–5755) was added and the tube was immediately shaken for 1 min to avoid agglomeration of salts followed by ultrasonic agitation for 20 min. The tube was centrifuged for 5 min at 3500 rpm and 10 mL of the supernatant were transferred to a 15 mL centrifuge tube containing 1 g EMR–Lipid sorbent (Agilent p/n 5982–1010). One ceramic bar (Agilent p/n 5982–9312) was added and the tube was shaken vigorously by hand for 1 min followed by centrifugation for 3 min at 4000 rpm. The entire supernatant was decanted into a second 15 mL polishing tube containing 2 g mixture of 4:1 (w/w) anhydrous $MgSO_4$:NaCl (Agilent p/n 5982–0101), and vortexed immediately to disperse, followed by centrifugation at 4000 rpm for 3 min. The extracts were then transferred into a glass test tube and evaporated to near dryness under a gentle stream of high-purity nitrogen. The residue was finally re-dissolved in 1 mL n-hexane and kept at −20 °C until GC-MS/MS analysis.

6.2.4 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 and a computer with MassHunter software (version B.05.00412) 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 C-1 (Appendix C). 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 or volume (depending on media type). Concentrations of PAHs are given in nano gram per liter (ng/L) for water, nano gram per gram dry weight (ng/g dw) for sediment and nano gram per gram wet weight (ng/g ww) for seafood samples.

6.2.5 Quality assurance and quality control (QA/QC)

The experiments were operated under strict quality control procedures. During the pretreatment procedure, all containers and equipment were pre-cleaned with methanol followed by acetone.

The limit of detection (LOD) for each analyte was defined as the smallest mass of compound resulting in an S/N ratio that was equal to or greater than 3, and the limit of quantitation (LOQ) was defined as the concentration in a standard yielding an S/N ratio of 10:1. The LOQs were in the range of 0.04 to 0.59 ng/L in water, 0.06 to 0.82 ng/g dw in sediment, and 0.03 to 0.21 ng/g ww in seafood. Instrumental blanks (solvent without internal standard) were analyzed every five to seven samples to monitor instrumental background. Instrumental blanks gave signal-to-noise (S/N) values of less than three. The procedural blanks (method blanks) prepared with Milli-Q water were spiked internal standards. Procedural blanks were analyzed with every batch of samples. Instrumental blanks and procedural blanks were below the LOQ. For each matrix, analyte recovery was determined by using spiked samples to validate the accuracy of the methods. Matrix spike recovery (n = 3) was determined by spiking the target compounds into the water (10 ng/L), sediment (10 ng/g dw) and seafood (10 ng/g ww) samples, followed by extraction and analysis as described in the previous section. The mean recoveries of PAHs spiked into the water, sediment and seafood samples were 76%−114%, 70%−112% and 71%−113%, respectively. The detailed QA/QC data are given in Table C-2 (Appendix C).

6.2.7 *Data analysis*

Statistical analyses were performed with IBM SPSS software (Version 23.0, IBM Corp., NY, USA). The significance level was set at p = 0.05. Before analyzing, concentration values lower than the LODs were set to LOD/2 (Succop et al., 2004). A statistical distribution test called *P–P* plots was carried out to test for normality. Descriptive statistics (range: minimum-maximum, mean, and median) was calculated by using the Microsoft® Excel® 2016 MSO Windows program. A one-way ANOVA was performed to determine the significant differences between the concentrations of PAHs detected in the coastal area of Bangladesh and to examine seasonal variations. The spatial distributions of PAHs were analyzed using MapViewer™ software (Version 8, Golden Software Inc., CO, USA).

6.3 Results and discussion

6.3.1 PAHs in surface water

6.3.1.1 Concentration of PAHs in surface water and global comparison

All of the sixteen target PAHs were detectable in surface waters of the coastal areas of Bangladesh. The concentrations of PAHs are summarized in **Table 6-1** and illustrated in **Figure 6-2**, while the detailed data are presented in Table C-3 and C-4 (Appendix C). The total concentrations of the ∑PAHs (sum of 16 USEPA PAHs) in the water phase 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 6-1**).

Table 6-1. 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); [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 C-2 (Appendix C) for the LOD values of investigated PAHs.

Among the 16 target PAHs, Nap, Flu, Phe, Ant, Flt, and Pyr were the most frequently detected compounds; they were found in 100% of samples 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 in the water phase were identified both by occurrence and abundance by combining the detection rate and mean relative concentration of each compound across samples. Regardless of season, top three PAH compounds were Nap, Flu and Phe, comprising up to 36–89% (mean: 68%) of ∑PAHs by sites, and highly correlated with ∑PAHs ($r = 0.96$; $p < 0.05$), 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).

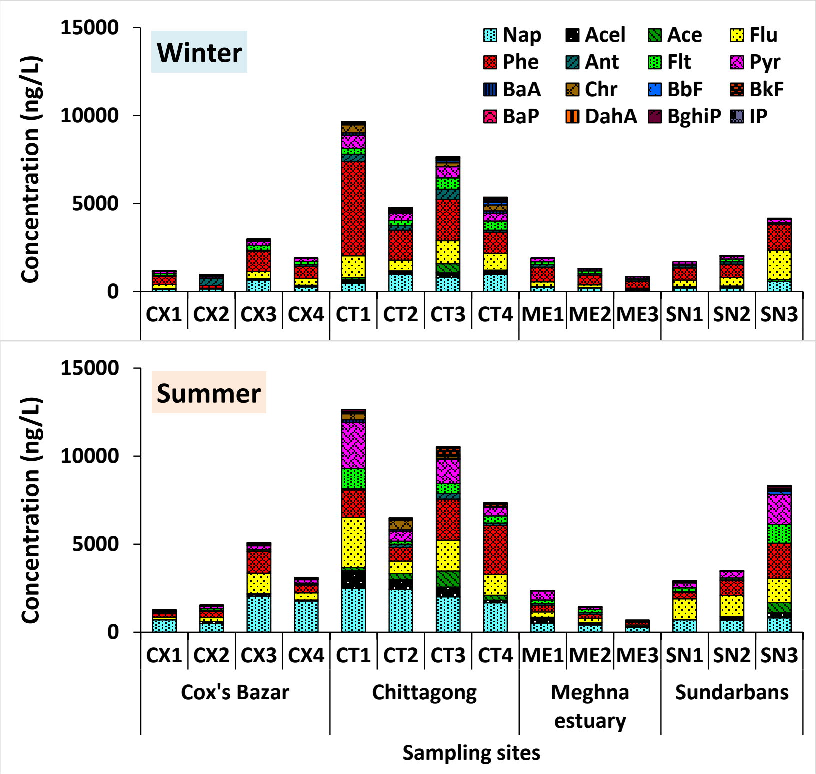

Figure 6-2. Concentrations of PAHs in the surface water of the Bangladeshi coastal area in winter and summer.

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 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 6-1**). Particularly, BaP (the best known potentially carcinogenic PAH) was detected in 50% and 64% samples in winter and summer, respectively. Due to absence of threshold levels 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 in the study area.

Table 6-2. 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	71.12–4255.43	Zheng et al., 2016
Yangpu Bay, China	2013	16	582.8–2208.3 (W)[b] 952.4–1201.7 (S)[b]	Li et al., 2015
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., 2012a
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) 679.4–12639.3 (S)	This study

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

Table 6-2 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., 2012a) 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 6-2**). 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.

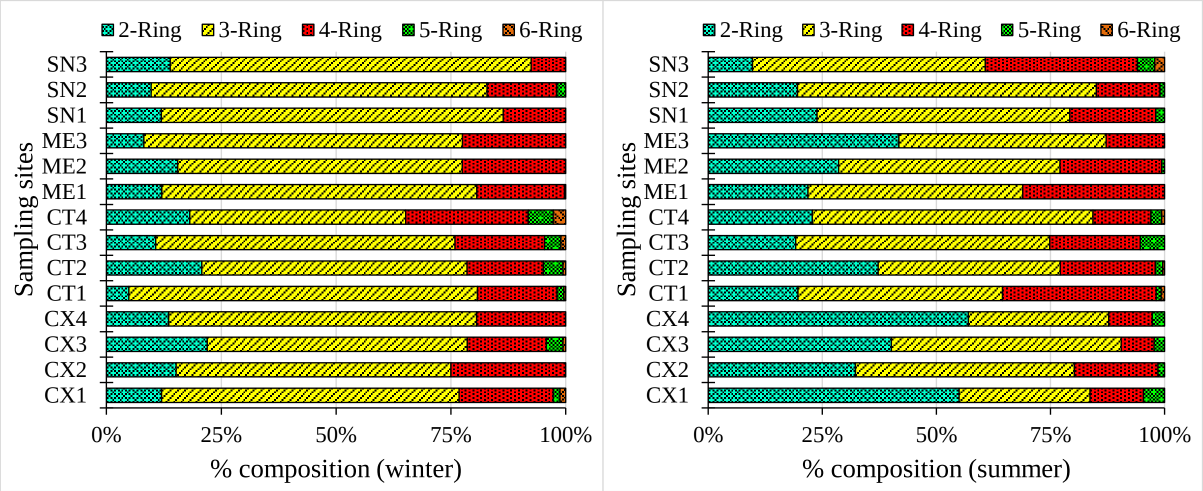

Figure 6-3. Compositional profiles of PAH compounds in surface waters taken in winter and summer.

6.3.1.2 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 **Figure 6-3**. Interestingly, the compositions of PAHs in different sampling sites are quite 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 C-3 and C-4, Appendix C). 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).

Similarity of PAHs components suggests that the PAH contamination in the Bangladeshi coastal water comes from similar sources. In general, the sources of LMW PAHs may be from petrogenic origin (e.g., incomplete combustion of fossil fuel, petroleum products, and biomass), whereas HMW PAHs are mainly derived from pyrogenic sources (high temperature combustion) (Fernandes et al., 1997). The predominant percentage of LMW PAHs in surface water signifies the importance of petrogenic sources in the Bangladeshi coastal area.

The ratio of the concentration of the parent PAHs in the environment is often used to determine the initial sources of the PAHs (Zhang et al. 2003; Yunker et al., 2002; Katsoyiannis and Breivik, 2014). Due to the difference in volatility, solubility, and adsorption, the concentration of PAHs would change from the sources to the environment, and the parent PAHs with similar molecular weight and stability were selected as molecular markers to distinguish the PAH sources (Yunker et al. 2002). 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 (**Figure 6-4**) and that have been proven to effectively differentiate PAH sources (Jiang et al., 2009; Martins et al., 2010; Xue et al., 2013).

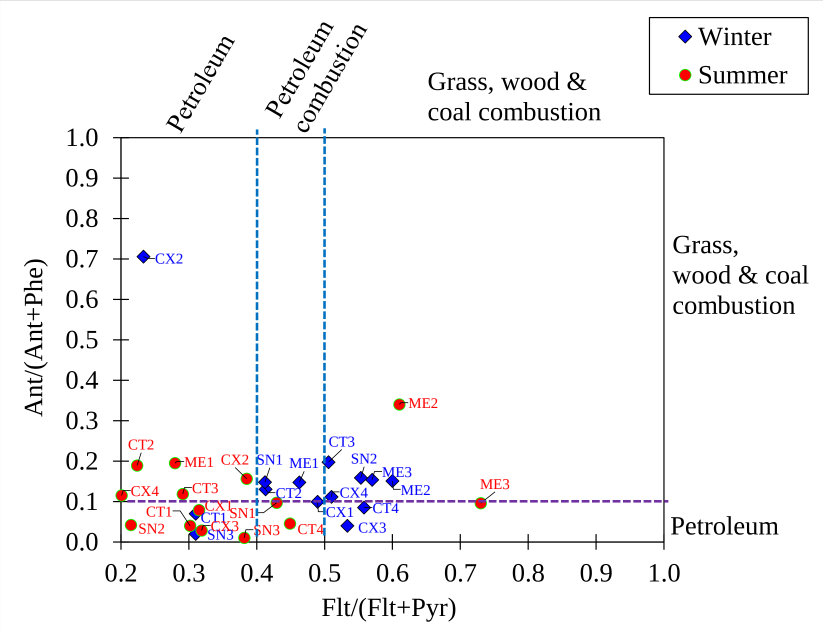

Figure 6-4. PAH cross-correlations for the ratios of Flt/(Flt+Pyr) and Ant/(Ant+Phe).

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 coastal area.

6.3.1.3 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 **Figure 6-5**. Small variations were observed in the levels of water phase PAH between the two seasons, although the difference was not statistically significant (p > 0.05). The concentration of ∑PAHs detected in summer were slightly higher than that in winter, suggesting less inputs during dry period. Precipitation and pollution sources are considered to be the main factors causing fluctuations in surface water quality. In summer (wet period), PAHs previously buried in the surface soil of heavily contaminated sites and accumulated in dry weather were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. Furthermore, the concentrations of PAHs in the gas phase increase in summer or in general 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 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, after screening the PAHs in the Bangladeshi coastal area, no clear seasonal variation was demonstrated for most of the target compounds 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, atmospheric deposition could be a major source of Nap in the surface water during times of heavy rainfall, such as the summer season. However, in the present study, the seasonal variations of this particular PAH in water might also be related to its seasonal emissions from land based sources or anthropogenic activities, although the exact reasons for this phenomenon are still unknown.

Figure 6-5 shows the spatial distribution of PAHs in surface water samples. 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 at sampling locations CT1–CT4 (average of 6862.2 and 9249.4 ng/L in winter and summer, respectively), SN1–SN3 (average of 2636.7 and 4916.8 ng/L in winter and summer, respectively) and CX1–CX4 (average of 1762.3 and 2760.6 ng/L in winter and summer, respectively) in comparison to sampling locations ME1–ME3 (average of 1355.3 and 1493.9 ng/L in winter and summer, respectively), which indicates that the industrialized regions (Chittagong, Cox’s Bazar and Sundarbans) in this area are potential sources of PAHs and that economic development seems to be associated with the amount of PAH emissions.

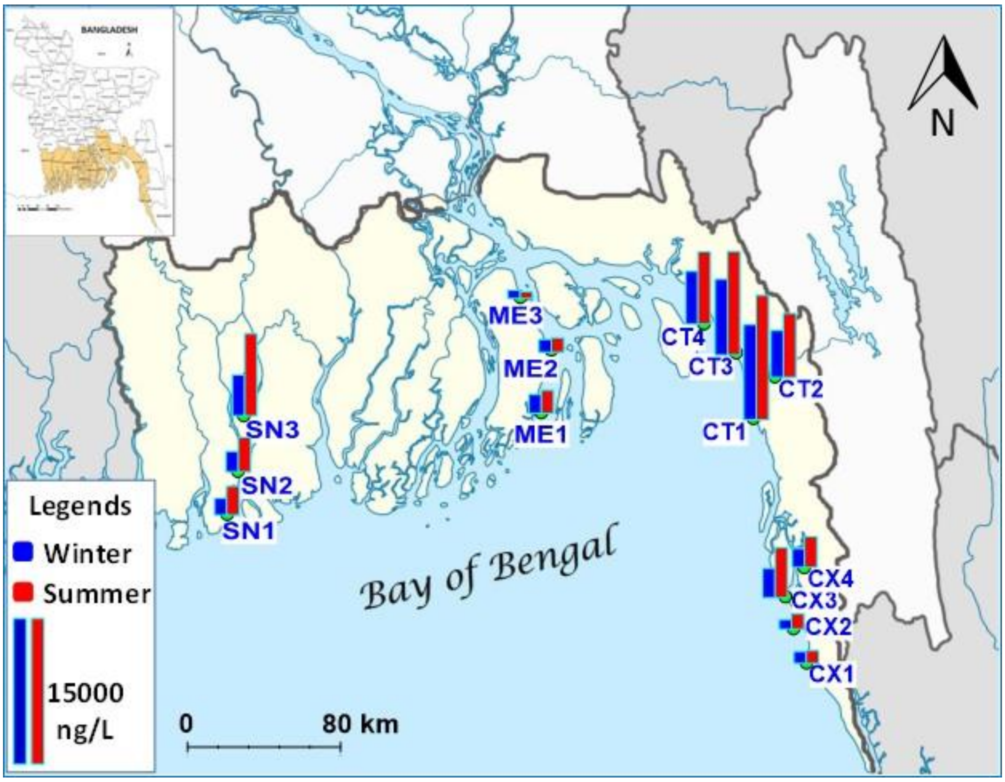

Figure 6-5. Distribution of total PAHs in surface water of the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh.

In particular, water samples at location CT1 showed the highest levels of PAHs for both seasons (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. These sites are predominantly influenced by the port activities. 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., 2016a). CT3 and CT4 are located very close to Chittagong ship breaking yard. Ship breaking activities along the coast of Chittagong may contribute to the PAH pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PAHs (Neşer et al., 2012; Siddiquee et al., 2012). Various activities in ship breaking operations including cutting, blasting, tank cleaning activities, bilge and ballast water discharges, asbestos removal, burning of electrical cables and plastic materials 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. Moreover, there are numerous multipurpose industrial establishments along the coast of Chittagong producing paper and pulp (e.g., Karnaphuli paper mills), cement clinkers, fertilizers (e.g., Karnaphuli fertilizer), steel products, rubber and plastic, petroleum products (e.g., Super petrochemical), beverages, sugar, pharmaceuticals, tobacco, jute, textiles, printing and dyeing, fish products, tannery products, paint, rechargeable batteries, jewelry, plating, automobile engines, and electronics. 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. Ship breaking and oil seepage from industrial activities lead to high concentrations of total PAHs in Mumbai harbor line in Indian coast of the Arabian Sea (Dhananjayan et al., 2012).

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., 2016a). 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, the main municipal garbage dump (mostly consumer products) of Cox's Bazar city is also located near CX3. Sometimes these garbage are openly burnt at this site. Therefore, a significant amount of PAHs might be emitted from this dumping and burning site into the adjacent water body.

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 untreated or partially 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, comparatively lower but detected PAHs concentrations in water from this site suggest that there are non-point sources of PAHs (e.g., surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc.) in the coastal area of Bangladesh. Furthermore, the Meghna Estuary sites receive major volumes of water from the GBM river systems (Ganges, Brahmaputra and Meghna River) of India through the Padma and Yamuna River flowing down to the Meghna Estuary via the Mehgna River. Chakraborty et al. (2014) reported that the surface water of these rivers were contaminated with PAHs at a level up to 31000 ng/L. The authors also noted that PAH concentrations gradually declined due to the dilution effects and ended at the Bay of Bengal. Thus, the coastal areas of Bangladesh to some extent might receive PAHs through the water flow of these upstream rivers, which flow into the Meghna Estuary sites. Generally speaking, the concentrations of ∑PAHs in surface water at the Chittagong sites (CT1–CT4), Cox's Bazar (CX1–CX4) and Sundarbans sites (SN1–SN3) were higher than those in the Meghna Estuary (ME1–ME3) and because of greater development, including industrialization and urbanization activities.

6.3.1.4 Ecological risk assessment of water-borne PAHs

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

$$RQ=\frac{C_{PAHs}}{C_{QV}} \qquad (6.1)$$

where C_{PAHs} was the concentration of certain PAHs in water samples and C_{QV} was 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:

$$RQ_{NCs} = \frac{C_{PAHs}}{C_{QV(NCs)}} \qquad (6.2)$$

$$RQ_{MPCs} = \frac{C_{PAHs}}{C_{QV(MPCs)}} \qquad (6.3)$$

where $C_{QV(NCs)}$ was the quality values of the NCs of PAHs in water and $C_{QV(MPCs)}$ was the quality values of the MPCs of PAHs in water. According to this, the ecological risk of 16 USEPA priority PAHs could be assessed (Zheng et al., 2016). 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).

Table 6-3. 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

Risk classification of individual PAHs and ∑PAHs is presented in **Table 6-3** (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 6-4** and the detailed data are provided in Table C-4 (Appendix C). 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 ecosystem was at high risk and suffered from severe toxicity, and remedial actions should be undertaken as soon as possible. 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, DahA, BghiP, and IP in summer were < 1 and RQ_{NCs} > 1, showing moderate risk to the ecosystems which should not be ignored indeed. 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 C-5, Appendix C). Site ME3 exhibited the lowest concentrations of ∑PAHs in both seasons (855.4 ng/L in winter and 679.4 ng/L in summer). By comparing RQs for ∑PAHs with the classification system given in **Table 6-3**, it is therefore illustrated that risk associated with ∑PAHs at all sites is of high level except for ME3 which is under moderate risk$_2$. In terms of season, it appears that 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. It is, therefore, suggested that intensive and long term survey of water quality should be conducted to develop effective management strategies and that should be utilized effectively to control the contamination of PAHs in Bangladesh.

Table 6-4. 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

6.3.2 PAHs in surface sediment

6.3.2.1 Concentration of PAHs in surface sediment

All of the sixteen USEPA priority PAHs were measured in the surface sediment of the coastal areas of Bangladesh. The concentrations of PAHs are summarized in **Table 6-5** and illustrated in **Figure 6-6**, while the detailed data are presented in Table C-6 and C-7

(Appendix C). The total concentrations of the ∑PAHs (sum of 16 PAHs) in the sediment samples ranged from 349.8 to 11058.8 ng/g dw (mean: 4571.0 ng/g dw; median: 4515.0 ng/g dw) in winter, and from 199.9 to 17089.1 ng/g dw (mean: 5729.0 ng/g dw; median: 4108.7 ng/g dw) in summer (**Table 6-5**).

Table 6-5. Concentrations of PAHs in sediments from the coastal area of Bangladesh and standard pollution criteria of PAH components for sediment matrix (ng/g dw).

PAHs	Winter				Summer				Guideline values	
	Range	Mean	Median	D.F.[a]	Range	Mean	Median	D.F.	ERL[d]	ERM[d]
Nap	11.5–202.2	69.3	54.1	100	18.5–1092.3	330.5	262.2	100	160	2100
Acel	4.2–246.8	50.0	23.2	100	7.8–420.53	127.9	78.1	100	44	640
Ace	0.5–106.7	26.8	23.05	100	2.1–816.7	170.2	76.2	100	16	500
Flu	1.2–126.1	52.3	45.0	100	0.6–545.6	97.7	39.1	100	19	540
Phe	17.3–1439.1	508.0	407.6	100	14.7–1538.3	482.6	502.7	100	240	1500
Ant	4.7–334.2	91.6	32.2	100	11.3–564.3	103.0	58.1	100	853	1100
Flt	59.9–3479.2	1118.2	950.6	100	30.6–3664.7	1135.2	816.9	100	600	5100
Pyr	46.4–2619.4	1195.8	1162.1	100	4.6–6190.3	1828.1	1125.9	100	665	2600
BaA	13.6–483.2	154.3	100.8	100	14.7–473.4	173.4	105.4	100	261	1600
Chr	28.9–1973.9	565.3	421.5	100	10.8–2907.9	565.9	193.5	100	384	2800
BbF	7.9–1108.6	308.9	262.1	100	4.6–657.2	147.4	54.0	100	NA	NA
BkF	1.8–347.3	110.6	109.0	100	4.7–584.9	159.4	75.2	100	NA	NA
BaP	4.4–373.5	132.1	110.3	100	17.8–352.7	102.9	65.2	100	430	1600
DahA	0.6–314.4	54.2	23.1	100	6.4–295	74.2	41.9	100	63.4	260
BghiP	3.4–271	77.4	33.4	100	2.6–207.8	60.0	34.8	100	NA	NA
IP	2.8–185.2	56.0	34.3	100	3.0–1475.5	170.5	20.2	100	NA	NA
∑C-PAHs[b]	84.5–3638.2	1381.4	1430.7	100	76.9–4908.8	1393.7	643.6	100	NA	NA
∑PAHs[c]	349.8–11058.8	4571.0	4515.0	100	199.9–17089.1	5729.0	4108.7	100	4000	44792

[a] Detection frequency (%); *n=14* for each season; [b] Sum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA and IP); [c] Sum of 16 USEPA PAHs; [d] The effects range low (ERL) and the effects range median (ERM) (Long et al., 1995); NA: not available;

All of the 16 target PAHs were detected in 100% of samples in both seasons. However, identifying indicator congeners of one certain class of environmental substances from various sources could be useful for predicting total pollutant contents by the determination of only indicator congeners (Liu et al., 2015). In this study, we identified the dominant PAH compounds in the sediment both by occurrence and abundance by combining the detection rate and mean relative concentration of each compound across samples. Regardless of season, top four PAH compounds were Flt, Pyr, Chr and Phe, comprising up to

30–90% (mean: 65%) of ∑PAHs by sites, and highly correlated with ∑PAHs (*r* = 0.98; $p < 0.05$), well representing the ∑PAHs in the surface sediment of the Bangladeshi coastal area.

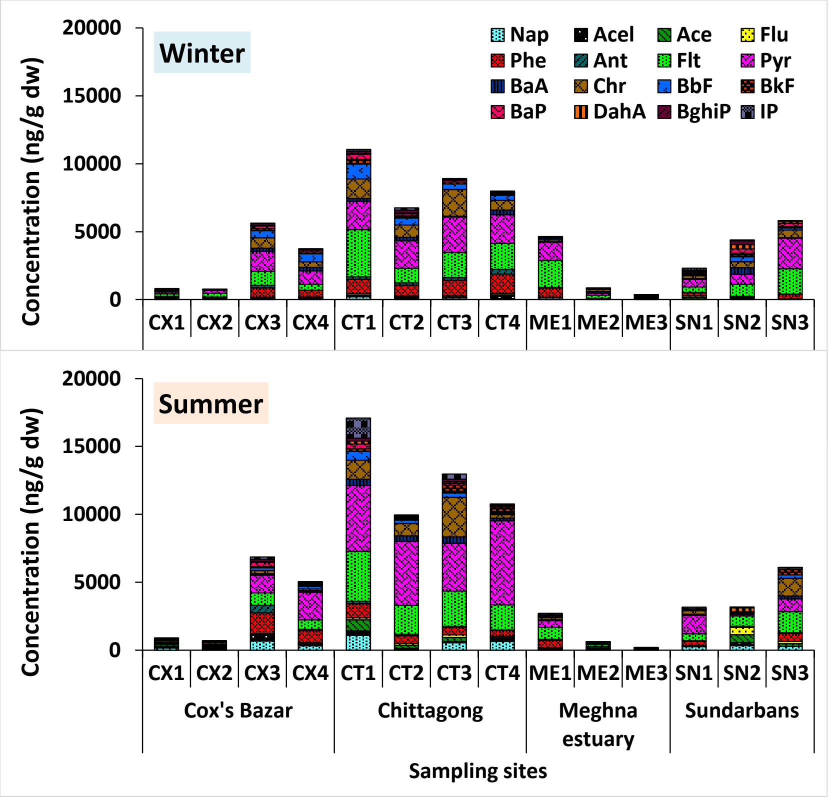

Figure 6-6. Concentrations of PAHs in the surface sediment in the Bangladeshi coastal area in winter and summer.

In particular, Flt was the most abundant PAH compound in winter with a contribution of 12–43% (mean: 24%) of ∑PAHs, whereas Pyr was the most abundant PAH compound in summer contributing up to 2–57% (mean: 22%) of ∑PAHs. However, the dominance of these PAH compounds were also reported in the coastal and estuarine sediments in several studies (Pozo et al., 2011; Gu et al., 2016; Goswami et al., 2016).

A comparison of total PAH concentrations in surface sediment samples collected from different coastal and estuarine regions around the world are shown in **Table 6-6**. To some degree, the results of comparison could at least reflect the pollution levels of PAHs in sediment, although in number and type of PAH compounds analyzed, the sediment fraction screened, and geological characteristics; so the comparison of total PAH levels from different studies have to be treated with caution.

Table 6-6. Concentrations of total PAHs in sediment (ng/g dw) from various estuary and coastal regions in the world.

Locations	Sampling year	*N*[a]	∑PAHs	Pollution levels	References
China coastal zones	2014	16	195.9–4610.2 (W)[b] 98.2–2796.5 (S)[b]	Low to high	Li et al., 2016
Kaohsiung Harbor, Taiwan	2006	17	34–16700	Low to very high	Chen et al., 2013
Jinhae Bay, Korea	2010	16	12.4–2430	Low to high	Yim et al., 2014
Tokyo Bay, Japan	2004	16	109–1170	Moderate to high	Horii et al., 2009
Boston Harbor, USA	1999	16	7300–358000	Very high	Wang et al., 2001
Entire continental shelf, Sweden	1995–2006	16	120–9600	Moderate to very high	Sánchez-García et al., 2010
Santander Bay, Northern Spain	–[c]	16	20–25800	Low to very high	Viguri et al., 2002
Genoa-Voltri Harbor, Italy	2002	16	4500–20800	Moderate to very high	Salvo et al., 2005
Norwegian Harbor, Norway	–	16	2000–76000	Moderate to very high	Oen et al., 2006
Gemlik Bay, Turkey	–	14	50.8–13482	Low to very high	Ünlü and Alpar, 2006
Mediterranean coastal zone, Egypt	–	16	3.51–14100	Low to very high	Barakat et al., 2011
Lenga Estuary, Chile	2002	16	290–6118	Moderate to very high	Pozo et al., 2011
Bahía Blanca estuary, Argentina	2004–2005	18	15–10260	Low to very high	Arias et al., 2010
Klang Strait, Malaysia	2011–2012	16	100.3–3446.9	Moderate to high	Sany et al., 2014
Estuarine and riverine area, Chennai, India	2014	16	13–31425	Low to very high	Goswami et al., 2016
Coastal area of Bangladesh	2015	16	349.8–11058.8 (W) 199.9–17089.1 (S)	Moderate to very high	This study

[a] Number of PAHs; [b] W: Winter, S: Summer; [c] Sampling year not mentioned.

In general, the ∑PAHs concentrations in the present study were comparable or lower than those measured in the Kaohsiung Harbor (Taiwan), Boston Harbor (USA), Santander Bay (Northern Spain), Genoa-Voltri Harbor (Italy), Norwegian Harbor (Norway), and Estuarine and riverine area (Chennai, India). However, the levels were higher than those reported in the China coastal zones, Jinhae Bay (Korea), Tokyo Bay (Japan), Entire continental shelf (Sweden), Gemlik Bay (Turkey), Mediterranean coastal zone (Egypt), Lenga Estuary (Chile), Bahía Blanca estuary (Argentina), and Klang Strait (Malaysia) (**Table 6-6**). Furthermore, the levels of sediment contaminations evaluated by PAH concentrations could be classified into four categories: (a) low, 0–100 ng/g; (b) moderate, 100–1000 ng/g; (c) high, 1000–5000 ng/g; (d) very high, >5000 ng/g (Baumard et al., 1998). Therefore, based on the global comparison along with the proposed contamination classification the surface sediment of the Bangladeshi coastal area could be classified as moderately to heavily polluted by PAHs. More specifically, sediments from this study area can be characterized as moderate for sites CX1, CX2, ME2, and ME3, high for ME1, SN1, and SN2, and very high for sites CX3, CX4, CT1, CT2, CT3, CT4, and SN3 in terms of PAH contamination.

6.3.2.2 PAH composition in sediments and source characterization

The composition patterns and relative abundance of PAHs by the number of aromatic rings in the Bangladeshi coastal surface sediments in winter and summer are illustrated in **Figure 6-7**. Interestingly, the compositions of PAHs in different sampling sites were almost similar and did not varied significantly between seasons ($p > 0.05$). In particular, 4-ring PAHs were the dominant compounds accounting 52–81% and 30–82% of ∑PAHs in winter and summer, respectively, followed by 5-ring PAHs (4–29% in winter and 4–18% in summer). Two-, 3- and 6-ring PAHs contributed lesser percentages of the ∑PAHs (Table C-6 and C-7, Appendix C). In general, regardless of source and season, the patterns of PAHs contamination on the basis of ring number were in order of 4-ring > 5-ring > 3-ring > 2-ring > 6-ring. The results are consistent with the typical PAHs composition in the surface sediments reported in other studies (Ünlü and Alpar, 2006; Sánchez-García et al., 2010; Pozo et al., 2011; Chen et al., 2013; Sany et al., 2014; Goswami et al., 2016; Li et al., 2016; Qian et al., 2016). About anthropogenic PAHs, low molecular weight (LMW, 2–3 rings) PAHs have both petrogenic and low-temperature combustion sources, whereas high molecular weight (HMW, 4–6 rings) PAHs have a predominantly high-temperature pyrolytic source (Mai et al., 2003). The dominance of HMW PAHs was attributed to their resistant to degradation and with lower aqueous solubility (hydrophobic), and hence they associate with particulate matter and eventually deposit in sediments (Nagy et al., 2014; Sinaei and Mashinchian, 2014; Yun et al., 2016). On the contrary, LMW PAHs have a tendency of long range transport and high aqueous solubility (Aziz et al., 2014), and thus tend to retain mainly in the dissolved phase. As a result, the higher concentrations of HMW PAHs than those of LMW PAHs have been commonly observed in sediments. In this study, the contribution of HMW PAHs (44–94%) was significantly higher than the LMW PAHs (6–56%). Zhou and Maskaoui (2003) reported that pyrolysis/combustion of fossil materials yields such PAH assemblages, which are subsequently introduced into the marine environment by coastal and river runoff, and by direct dry and wet precipitation from the atmosphere. Industrial and domestic wastes/sewerage were also suggested as another important local source.

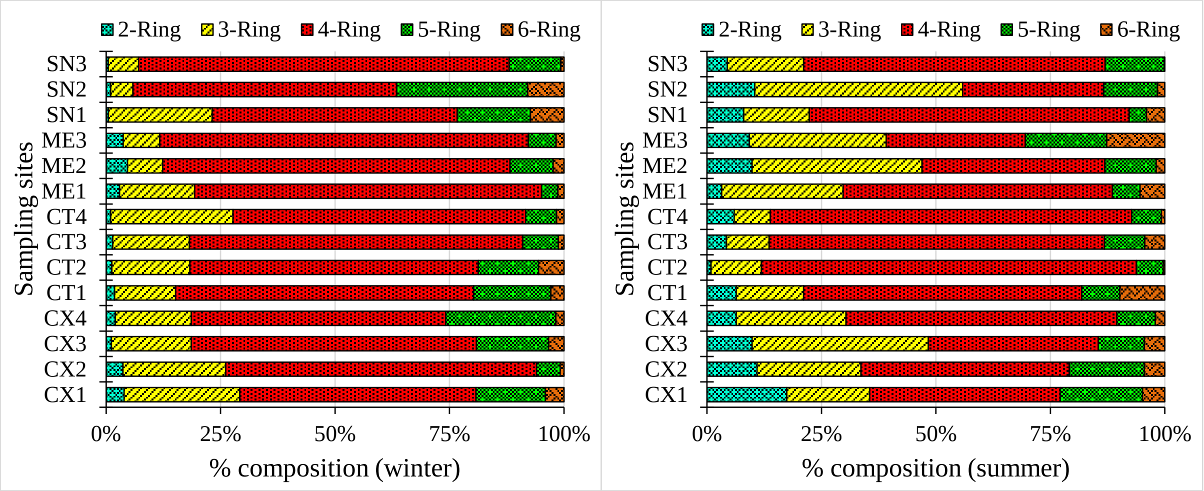

Figure 6-7. Compositional profiles of PAH compounds in surface sediments taken in winter and summer.

Source identification of PAHs is important to reveal the transport and fate processes of PAHs in the environment (Wang et al., 2009). As a general practice, ratios of various individual PAH concentrations are used to distinguish the possible sources of PAHs (Yunker et al., 2002; Katsoyiannis and Breivik, 2014). Among these molecular ratios, Ant/(Ant + Phe), Flt/(Flt + Pyr), BaA/(BaA + Chr) and IP/(IP + BghiP) are more widely used (Yunker et al., 2002; Tobiszewski and Namiesnik, 2012). Although the use of PAH diagnostic ratios for source characterization has been criticized (Katsoyiannis et al., 2007; Galarneau, 2008), it has been used widely and proven to effectively differentiate PAH sources (Jiang et al., 2009; Martins et al., 2010; Xue et al., 2013) and consequently, in the present study it is utilized as an indicative tool of PAHs source and distribution.

The criteria for PAH ratios as indicators of PAH sources are listed in **Table 6-7** and the plotted isomeric ratios of: Ant/(Ant + Phe) vs. Flt/(Flt + Pyr), and BaA/(BaA + Chr) vs. IP/(IP + BghiP) used in this study are shown in **Figure 6-8**. The values of Ant/(Ant + Phe) were 0.02–0.83, 86% of which were higher than 0.1; the ratios of Flt/(Flt + Pyr) were less than 0.4 in 14%, 0.4–0.5 in 29% and higher than 0.5 in 57% of samples; the values of BaA/(BaA + Chr) were less than 0.2 in 25%, 0.2–0.35 in 32% and higher than 0.35 in 43% of samples; the ratios of IP/(IP + BghiP) were less than 0.2 in 4%, 0.2–0.5 in 21% and higher than 0.5 in 75% of samples (**Figure 6-7**). The above data demonstrated that the PAHs in sediments from the Bangladeshi coastal area originated from both the petrogenic and pyrogenic sources including crude petroleum (e.g. gasoline/diesel), petroleum combustion, and combustion of grass, wood and coal. However, more than 60% of the values calculated from the PAH molecular ratios skewed towards the values for indication of pyrolytic origin.

Table 6-7. The criteria for PAH ratios as indicator of PAH source (Yunker et al., 2002).

PAH ratio	Sources
Ant/(Ant + Phe) < 0.1	Petroleum
Ant/(Ant + Phe) ≥ 0.1	Grass, wood & coal combustion
Flt/(Flt + Pyr) <0.4	Petroleum
0.4 ≤ Flt/(Flt + Pyr) < 0.5	Petroleum combustion
Flt/(Flt + Pyr) ≥ 0.5	Grass, wood & coal combustion
BaA/(BaA+Chr) < 0.2	Petroleum
0.2 ≤ BaA/(BaA+Chr) < 0.35	Petroleum combustion
BaA/(BaA+Chr) ≥0.35	Grass, wood & coal combustion
IP/(IP+BghiP) < 0.2	Petroleum
0.2 ≤ IP/(IP+BghiP) < 0.5	Petroleum combustion
IP/(IP+BghiP) ≥ 0.5	Grass, wood & coal combustion

In addition, the LMW/HMW ratio was less than 1 for most sites, suggesting a pyrolytic origin of PAHs was prominent at these sites (mean 0.35; range 0.06–1.26). However, PAH contamination in this area from petrogenic origin should not be ignored as well. Interestingly, it was seen from **Figure 6-8** that the ratios were more scattered in summer season compared to winter, indicating a considerable input of PAH might be occurred through atmospheric wet and dry deposition and/or land runoff during times of heavy rainfall, and that are most probably originated from non-point defuse sources, such as vehicular exhaust from cities, high intensity of fishing by motorized boats and vessels, emission mainly as black smoke from seasonally operated factories and industries (e.g. brick kiln) from nearshore or far shore areas, etc.

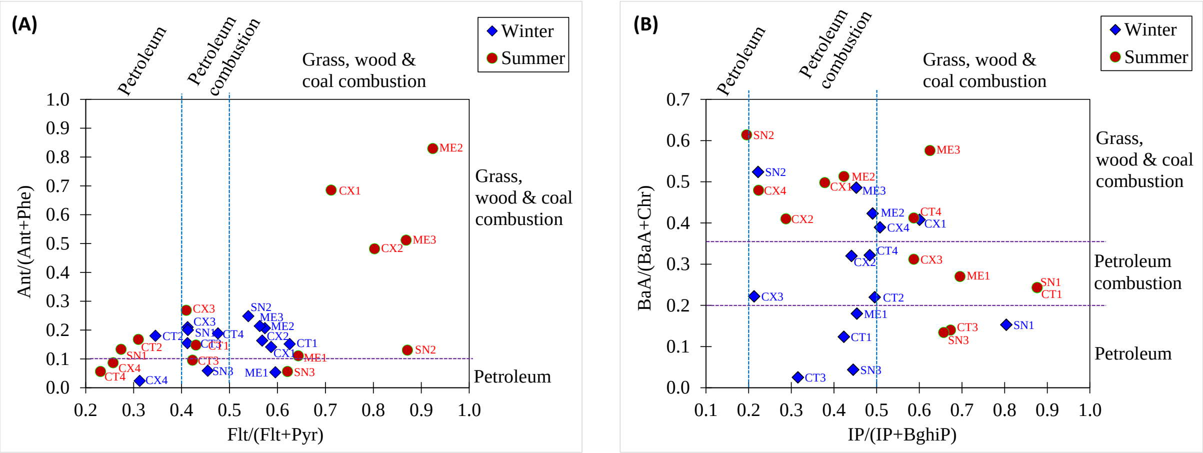

Figure 6-8. PAH cross-correlations for the ratios of (A) Ant/(Ant+Phe) and Flt/(Flt+Pyr), (B) BaA/(BaA+Chr) and IP/(IP+BghiP).

6.3.2.3 Seasonal and spatial distribution of sedimentary PAHs

The seasonal variations of sedimentary PAHs were investigated. As a whole, the levels of ∑PAH between the two seasons did not differ significantly ($p > 0.05$). However, seasonal variations to an extent were noticeable depending on the locations. For example, CX1, CX3, CX4, CT1, CT2, CT3, CT4, SN1 and SN2 showed slightly higher concentrations in summer, whereas the rest of the locations exhibited the opposite trends. In summer (wet period), PAHs previously buried in the surface soil of heavily contaminated sites and accumulated in dry weather were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. Furthermore, the concentrations of PAHs in the gas phase increase in summer or in general in tropical regions (Abdel-Shafy et al., 2016) that could be redeposited through the atmospheric wet and dry deposition. Additionally, increased shipping and fishing activities during summer in the study area might also be another crucial factor for the seasonal variations. On the contrary, elevated levels of PAHs in winter samples were probably due to either co-evaporation of PAHs with water or intense sedimentation from less mixing effects due to comparatively weaker tidal movement or wave action and lower inflow from upstream rivers in the dry season as well. Overall, the pattern of seasonal variation of ∑PAHs concentrations in this study was in agreement with other studies (Zhang et al., 2012; Tian et al., 2014), but contrary to the temporal variations of ∑PAHs concentrations in sediment from China coastal zones (Li et al., 2016). These discrepancies might be attributed to the impact of regional hydrological conditions and local pollution sources among the study areas. However, in terms of individual PAHs, concentrations of 12 PAHs (Nap, Acel, Ace, Flu, Phe, Ant, Flt, Pyr, BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IP) were found to be higher in summer, whereas the other 4 PAHs (Phe, BbF, BaP, and BghiP) showed the reverse trends, although the differences were statistically significant only for Nap (average of 69.3 and 330.5 ng/g dw in winter and summer, respectively) and Ace (average of 26.8 and 170.2 ng/g dw in winter and summer, respectively) ($p < 0.05$). The seasonal variations of these particular PAHs might be related to its seasonal emissions from land-based or sea-based sources or due to impact of anthropogenic activities during this particular time. This variation might also be influenced by hydrological conditions of the site, the physico-chemical properties of the deposited sediments, absorption-desorption mechanism, degradation of PAH compounds along with their other properties. However, the exact reasons are yet to be investigated in the study area. In general, regardless of sampling sites the concentration of ∑PAHs in summer was slightly higher than in winter.

Figure 6-9 shows the spatial distribution of PAHs in surface sediment samples. Levels of PAHs in surface sediment differed significantly between the four coastal regions ($p < 0.05$), indicating the PAHs contamination mainly influenced by the local/regional source inputs in the study areas. However, the pattern of ∑PAHs distribution between sites were more or less similar in winter and summer. Concentrations of ∑PAHs were higher in the coastal sediments at sampling locations CT1–CT4 (average of 8676.5 and 12693.6 ng/g dw in winter and summer, respectively), SN1–SN3 (average of 4171.1 and 4138.9 ng/g dw in winter and summer, respectively) and CX1–CX4 (average of 2734.3 and 3374.5 ng/g dw in winter and summer, respectively) in comparison to sampling locations ME1–ME3 (average of 1946.0 and 1172.5 ng/g dw in winter and summer, respectively), which indicates that the industrialized urban regions (Chittagong, Cox's Bazar and Sundarbans) in this area are potential sources of PAHs and that the economic development seems to be associated with the amount of PAH emissions. Likewise, the pattern is thus well correlated with the degree of energy consumption between areas.

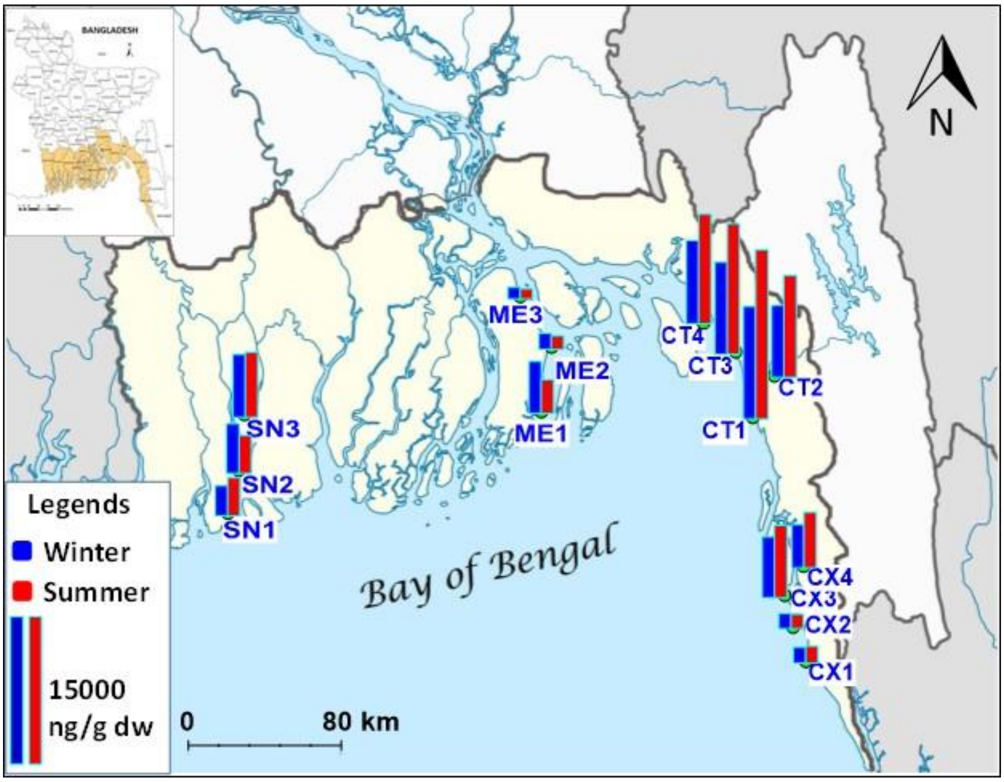

Figure 6-9. Distribution of total PAHs in surface sediment of the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh.

In particular, sediment samples at location CT1 showed the highest levels of ∑PAHs for both seasons (11058.8 and 17089.1 ng/g dw in winter and summer, respectively) followed by CT3 (8906.6 and 12968.0 ng/g dw 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 and heavy traffic of tankers and cargo ships is common nearby. These sites are predominantly influenced by the port and fishing harbor activities. 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, and that are in well agreement with other studies (Dudhagara et al., 2016; Wang et al., 2016a). CT3 and CT4 are located very close to Chittagong ship breaking yard. Ship breaking activities along the coast of Chittagong may contribute to the PAH pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PAHs (Neşer et al., 2012; Siddiquee et al., 2012; Dudhagara et al., 2016). Various activities in ship breaking operations including cutting, blasting, tank cleaning activities, bilge and ballast water discharges, asbestos removal, burning of electrical cables and plastic materials 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 the adjacent aquatic environment through wet deposition and air/water exchange processes. Moreover, there are numerous multipurpose industrial establishments along the coast of Chittagong producing paper and pulp (e.g., Karnaphuli paper mills), cement clinkers, fertilizers (e.g., Karnaphuli fertilizer), steel products, rubber and plastic, petroleum products (e.g., Super petrochemical), beverages, sugar, pharmaceuticals, tobacco, jute, textiles, printing and dyeing, fish products, tannery products, paint, rechargeable batteries, jewelry, plating, automobile engines, and electronics. 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. Furthermore, Chittagong is the second largest city in Bangladesh and has a population of approximately 4.5 million people. Therefore, higher energy consumption as well as vehicular emission and resulted street runoff from this densely populated city may partly contribute to the observed elevated levels of PAHs. Previous studies have identified that the ship breaking activities, leakage of diesel fuel, crude oil used in ship engines and coal from ship and heavy transport vehicles and oil seepage from industrial activities lead to high concentrations of total PAHs in the coastal sediments (Dhananjayan et al., 2012; Dudhagara et al., 2016).

Within Cox's Bazar, water samples from CX3 (Bakkhali Estuary) showed elevated concentration of ∑PAH (5630.0 and 6861.4 ng/g dw in winter and summer, respectively), followed by CX4 (3741.5 and 5046.5 ng/g dw in winter and summer, respectively). It was expected because these sites receive huge residential and industrial waste from the surrounding area, and this coastal waterbody is predominantly 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., 2016a). Combustion or incomplete combustion of wood and wooden materials, and coal that are used by several small factories and in some cases at households in nearby areas might also be a potential emission source of PAH in this area. In addition, the main municipal garbage dump (mostly consumer products) of Cox's Bazar city is also located near CX3. Sometimes these garbage are openly burnt at this site. Therefore, a significant amount of PAHs might be emitted from this dumping and burning site into the adjacent water body. In Sundarbans area, the highest concentration of ∑PAHs was found at SN3 (5815.0 and 6081.3 ng/g dw 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 untreated or partially 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. Burning of coal and other petroleum products, oil spillage from motorized boats and adjacent road traffics were observed as very common sources of PAHs contaminations in estuarine environments (Duan et al., 2015; Keshavarzifard et al., 2015; Goswami et al., 2016).

The levels of ∑PAHs in sediment taken from the Meghna estuary were significantly lower than that from other areas ($p < 0.05$). A clear downward increasing trend was noticeable in this area. The increase in PAH concentrations near the mouth of the estuary (ME1) might be attributed to the river-sea boundary zone sedimentation or marginal filter effects (Wang et al., 2016b). A marginal filter functions as a barrier that contributes to sorption and sedimentation (Yang et al., 2012) resulting in high contamination. Besides, fine particles carried by the inland rivers to the zone where the rivers discharge their loads might be a major carrier of PAHs from the upstream to the mouth of the estuary. In addition, ships and boats for fishing and transportation travel in this area, which might be partly associated with the increase of PAHs as well. It is to be noted that the Meghna estuary is an exclusively unindustrialized area. In particular, ME3 is located in an unindustrialized rural area less or not affected by cities, which led to the lowest concentration of PAHs (349.8 and 199.9 ng/g dw in winter and summer, respectively). However, comparatively lower but detected PAHs concentrations in sediments from this site suggest that there are non-point sources of PAHs, such as surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc. In addition, the differences of regional fossil energy consumption could partly result in the spatial-seasonal variations in PAH contamination of the surface sediments along the coastal areas of Bangladesh. Generally speaking, the coastal sediments in the Chittagong area were the most contaminated by PAHs followed by Cox's Bazar and Sundarbans, whereas Meghna Estuary was the least contaminated area and because of greater development, including industrialization and urbanization activities.

6.3.2.4 Impacts of sediment properties on PAHs distribution

The physicochemical properties of sediment (e.g. grain size/texture, organic matter) have been recognized as a major influential factor in determining the distribution and transport of hydrophobic organic compounds (OC) like PAHs in the aquatic environments (Wang et al., 2001; Sánchez-García et al., 2010; Chen and Chen, 2011; Chen et al., 2013; Li et al., 2016). Among them total organic carbon (TOC) is crucial for their sorption, a key process in trapping and transporting PAHs in sediments (Oen et al., 2006; Chen and Chen, 2011).Within total organic moieties, black carbon (BC) with the highest sorption capacity has been shown to large extent affect the distribution of OCs in sediments (Accardi-Dey and Gschwend, 2002). PAHs may be extensively adsorbed onto condensed carbonaceous geo-sorbents such as BC owing to its condense and aromatic structure (Lohman et al., 2005; Wang et al., 2014). Moreover, correlations between environmental contaminants might be indicative of common or multiple pollution sources in a specific study area. To identify such correlations and influence of sediment properties on the distribution of PAHs in the present study area, we performed a Spearman rank correlation analysis and the results are shown in **Table 6-8**.

Table 6-8. Spearman rank correlations among the individual PAHs and some physical chemical properties [total organic carbon (TOC), black carbon (BC), total nitrogen (TN), pH, grain size (sand, silt, clay)] of sediment samples (*n=28*).

Variables	Nap	Acel	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	IP	∑PAHs	TOC	BC	TN	pH	Clay	Sand	Silt
Nap	**1**																							
Acel	**0.64**	**1**																						
Ace	**0.54**	0.38	**1**																					
Flu	**0.50**	0.31	**0.84**	**1**																				
Phe	**0.55**	0.38	0.44	**0.60**	**1**																			
Ant	0.45	0.45	0.20	0.37	**0.74**	**1**																		
Flt	**0.56**	0.32	**0.53**	**0.71**	**0.79**	**0.52**	**1**																	
Pyr	**0.51**	0.32	**0.53**	**0.62**	**0.77**	**0.49**	**0.88**	**1**																
BaA	0.44	0.48	0.37	0.44	0.40	0.35	0.48	**0.50**	**1**															
Chr	0.39	0.15	0.43	**0.62**	**0.70**	**0.54**	**0.80**	**0.80**	**0.66**	**1**														
BbF	0.36	0.30	0.38	**0.53**	**0.77**	**0.54**	**0.73**	**0.72**	**0.71**	**0.88**	**1**													
BkF	**0.50**	0.34	**0.55**	**0.59**	**0.61**	**0.48**	**0.75**	**0.77**	**0.75**	**0.89**	**0.81**	**1**												
BaP	0.28	0.24	**0.55**	**0.54**	**0.59**	**0.49**	**0.58**	0.47	0.47	**0.68**	**0.75**	**0.66**	**1**											
DahA	0.40	**0.49**	**0.71**	**0.50**	0.24	0.15	0.43	0.45	0.36	0.34	0.38	**0.52**	**0.63**	**1**										
BghiP	0.48	**0.57**	0.47	**0.54**	**0.65**	**0.61**	**0.64**	**0.61**	**0.61**	**0.59**	**0.72**	**0.63**	**0.67**	**0.67**	**1**									
IP	0.41	**0.52**	0.37	0.43	**0.58**	**0.51**	**0.52**	**0.53**	0.40	**0.55**	**0.58**	**0.56**	**0.57**	**0.52**	**0.76**	**1**								
∑PAHs	0.67	0.45	0.63	0.74	0.82	0.58	0.94	0.92	0.61	0.85	0.80	0.86	0.63	0.52	0.71	0.59	1							
TOC	0.26	0.31	0.26	0.22	0.40	0.36	0.17	0.28	0.29	0.22	0.36	0.26	0.27	0.03	0.26	0.25	0.30	**1**						
BC	**0.51**	0.33	**0.46**	**0.62**	**0.55**	0.32	**0.53**	**0.53**	0.38	**0.43**	**0.43**	**0.44**	0.23	0.10	0.35	**0.57**	**0.56**	**0.71**	**1**					
TN	0.15	0.05	0.18	0.21	0.13	0.08	-0.04	0.03	0.21	0.11	0.17	0.07	0.14	-0.09	0.01	-0.09	0.08	**0.77**	**0.52**	**1**				
pH	-0.38	-0.20	**-0.50**	**-0.49**	-0.45	-0.35	-0.36	-0.46	-0.35	**-0.51**	-0.39	-0.48	-0.21	-0.06	-0.15	-0.11	-0.50	-0.44	-0.47	-0.41	**1**			
Clay	0.29	0.32	**0.47**	0.41	**0.47**	0.37	**0.53**	**0.53**	0.37	**0.50**	**0.49**	**0.58**	0.43	**0.47**	**0.47**	0.44	**0.50**	0.44	**0.65**	0.35	-0.37	**1**		
Sand	-0.14	-0.21	-0.01	0.13	-0.15	-0.27	0.05	-0.10	-0.10	0.01	-0.05	-0.02	0.05	0.21	0.00	0.02	-0.08	**-0.77**	-0.34	-0.47	0.37	-0.12	**1**	
Silt	0.13	0.18	-0.04	-0.20	0.14	0.18	-0.05	0.06	0.09	-0.10	0.04	-0.08	-0.06	-0.24	-0.05	-0.13	0.05	**0.68**	0.27	0.40	-0.24	-0.08	**-0.93**	**1**

Values in bold are significant at the 0.01 level (2-tailed).

Significant correlations were found between multiple PAH compounds indicating that the correlated compounds might originate from some shared pollution sources in the study area. In addition, the significant positive correlations between some of the LMW PAHs and HMW PAHs might also be an indication of the prevalence of both petrogenic and pyrolytic sources.

The concentrations of sedimentary TOC (%) and BC (%) were recorded as 0.06–1.28 and 0.06–0.53 in winter and 0.15–0.69 and 0.06–0.28 in summer, respectively (Table A-9, Appendix A). No significant variation was observed in sediment properties between the two seasons (t-test, $p > 0.05$). However, the correlation results revealed a significant positive association between the ∑PAHs and BC ($r = 0.56$, $p < 0.01$), whereas the correlations between ∑PAHs and TOC were positive but not significant ($r = 0.30$, $p > 0.05$). Similar observations were also reported in other studies and the possible reason may be due to an irregular and heterogeneous distribution of PAHs in the coastal or estuarine sediments (Oen et al., 2006; Liu et al., 2007; Hsieh et al., 2010).In terms of individual PAHs, none of them showed significant association with TOC ($p > 0.05$). On the contrary, Nap, Ace, Flu, Phe, Flt, Pyr, Chr, BbF, BkF, and IP were positively ($r = 0.43$–0.71) and strongly ($p < 0.01$) correlated with the BC content. Overall, the correlation between both the individual and ∑PAH to the BC contents was stronger than the correlation to the TOC contents, indicating that PAH sorption in the studied sediments was BC-dominated. In addition, the PAHs well correlated with BC might be derived from pyrolytic origin (Li et al., 2016). Furthermore, sediment texture is considered to be one of the main factors affecting the accumulation profile of PAHs in sediment. In the present study, sediment quality characteristics ranged from silt loam or clay to sandy loam (Table A-9, Appendix A), but the texture varied seasonally. However, significant correlation between ∑PAHs and clay particles (r = 0.50, p < 0.001) implies the role of the fine-grained particles that are enriched with soot carbon (e.g. BC) (Maruya et al., 1996) in the transportation and redistribution of PAHs in the coastal area of Bangladesh.

6.3.2.5 Ecological risk assessment of sedimentary PAHs

6.3.2.5.1 Sediment potential toxicity based on carcinogenic PAHs (C-PAHs)

IARC (1987) has identified Chr, BaA, BkF, BbF, BaP, DahA, and IP as possible carcinogens. The total concentration of carcinogenic PAHs (∑C-PAHs) were 84.5–3638.2 ng/g dw (mean: 1381.2 ng/g dw) and 76.9–4908.8 ng/g dw (mean: 1393.7 ng/g dw), accounting 8–57% and 11–48% to the ∑PAHs in winter and summer, respectively (**Table 6-5**).

The potential toxicity of sediment was evaluated using the total toxic BaP equivalent (TEQ^{carc}) (Qiao et al., 2006; Chen and Chen, 2011, Chen et al., 2013). The TEQ^{carc} for all C-PAHs was calculated using the following equation:

$$\mathrm{TEQ}^{carc} = (\mathrm{C_i} \times \mathrm{TEF_i^{carc}}) \qquad (6.4)$$

where C_i is the C-PAH concentration (ng/g dw) and TEF_i^{carc} (toxic equivalency factors) is the toxic factor of carcinogenic PAHs relative to BaP. Among all known potentially carcinogenic PAHs, BaP is the only PAH for which toxicological data are sufficient to derive a carcinogenic potency factor (Peters et al., 1999). According to the USEPA (1993), TEFs for BaA, CH, BbF, BkF, BaP, DBA, and IP are 0.1, 0.001, 0.1, 0.01, 1, 1, and 0.1, respectively. In this study, the total TEQ^{carc} values of sediment samples varied from 15.2 to 717.7 ng TEQ/g dw, with a mean value of 239.9 ng TEQ/g dw in winter and 27.8 to 835.5 ng TEQ/g dw, with a mean value of 228.4 ng TEQ/g dw in summer. Regardless of seasons, the higher total TEQ^{carc} values were found at sites CT1, CT3, CX3, SN2 and SN3 near the port area and industrial zones (**Figure 6-10**). It suggests that the shipping and fishing activities in the port area along with other industrial activities play an important role in the leaching of C-PAHs into the environment. The TEQ^{carc} found in the Bangladeshi coastal sediments is comparable to that in sediments from Meiliang Bay, China (Qiao et al., 2006), Barents Sea, Russia (Savinov et al., 2003), but lower than in sediment from Naples harbor (Italy) (Sprovieri et al., 2007), and Kaohsiung Harbor, Taiwan (Chen and Chen, 2011, Chen et al., 2013).

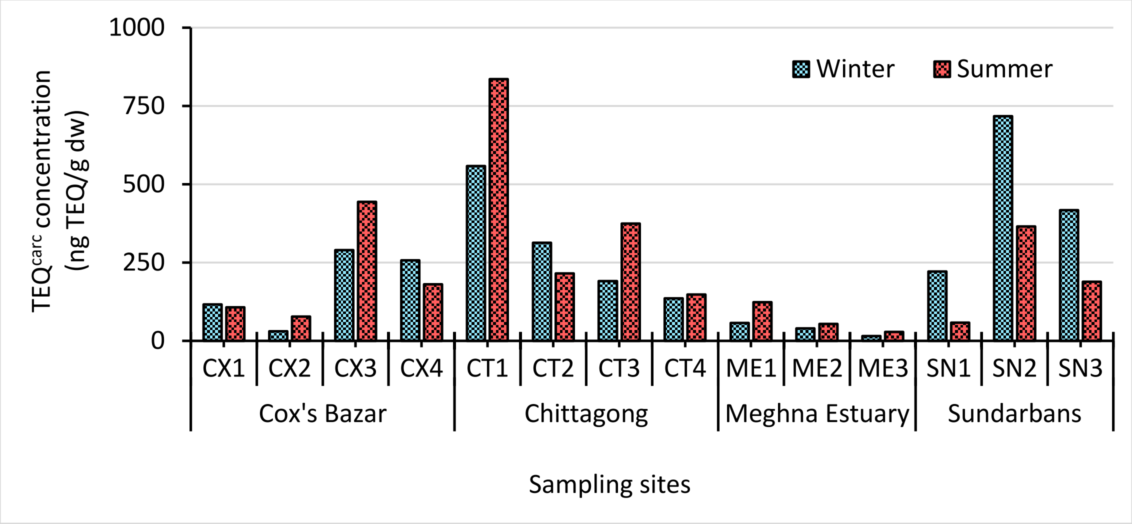

Figure 6-10. Distribution of TEQ^{carc} values (ng TEQ/g dw) in the surface sediments of the Bangladeshi coastal area.

6.3.2.5.2 Assessment of sediment quality using biological thresholds

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

Many approaches have been developed to evaluate the ecotoxicological aspect of sediment-bound PAH contamination. One such evaluation method is the sediment quality guidelines (SQG). A widely used sediment toxicity screening guideline of the US National Oceanic and Atmospheric Administration provides two target values to estimate potential biological effects: effects range low (ERL) and effects range median (ERM) (Long et al., 1995). The guideline was developed by comparing various sediment toxicity responses of marine organisms or communities with observed PAH concentrations in sediment samples. These two values delineate three concentration ranges for each particular chemical. When the concentration is below the ERL, then a biological effect may be rare. If the concentration is equal to or greater than the ERL but below the ERM, it indicates that a biological effect will occur occasionally. Concentrations at or above the ERM indicate that a negative biological effect will occur frequently. A rough comparison of the measured concentrations of PAHs with the ERM and ERL values are shown in **Table 6-5**. Among the sediment samples analyzed, none of them exceeded the ERM of ∑PAHs. The ∑PAHs are between ERL and ERM in 50% of samples in both seasons, and below ERL in the other samples (50%). In most of the samples except ME2 and ME3 in winter and CX2 and ME3 in summer, the concentrations of the majority of the individual PAHs were above ERL but below ERM, which indicate that biological effects would occur occasionally. Moreover, some PAHs were in at least one sample that exceeded ERM in sites CT3 and SN2 in winter and CX3, CT1, CT2, CT3, CT4 and SN2 in summer (e.g., site CX3: Phe, site CT1: Ace and Pyr, site CT2: Pyr, site CT3: Pyr and Chr, site CT4: Pyr and site SN2: Ace and DahA), which indicates that biological effect would occur frequently at these sites.

In addition, there are still no reported values of SQG for high molecular weight PAHs such as BbF, BkF, BghiP and IP, which may also contribute to toxicity in the sediments. Therefore, we evaluated the toxicological implications of the above four PAHs in sediment at the interim sediment quality guideline (ISQG) range recommended by the Canadian sediment guidelines for the protection of aquatic life in our research (Long et al., 1995; Behnisch et al., 2003). The low effect limits (LELs) of BbF, BkF, BghiP and IP are 200, 240, 200, and 170 ng/g dw, respectively, with serious effect limits (SELs) of 320000, 1340000, 320000, and 32000 ng/g dw. No PAH in any samples exceeded the SEL. The concentrations of BbF, BkF, BghiP and IP exceed the LEL values in 7–43% of samples irrespective to season. These findings indicate that PAHs in the sediment from coastal area of Bangladesh will cause adverse biological effects mostly occasionally but for some sites the effect would occur more frequently.

6.3.3 PAHs in seafood

6.3.3.1 Concentration of PAHs in seafood

The concentrations of PAHs in commonly consumed seafood (5 finfish and 2 shellfish species) from the coastal area of Bangladesh are summarized in **Table 6-9** and **Figure 6-11**. The detailed results are presented in Table C-8 and C-9 (Appendix C). Although at varying detection frequencies all of the 16 target PAHs were detectable, suggesting the ubiquitous

Table 6-9. Concentrations (ng/g ww) and detection frequency (%) of PAHs in seafood (finfish and shellfish) collected from the coastal area of Bangladesh in the two seasons.

Sample	Season		Nap	Acel	Ace	Flu	Phe	Ant	Flt	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	BghiP	IP	∑C-PAH[c]	∑PAHs[d]
Finfish	Winter	Minimum	83.9	<LOD[a]	2.6	6.8	45.6	2.9	5.3	10.9	<LOD	5.7	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	10.1	184.5
		Maximum	1540.8	89.5	163.8	266.5	745.9	86.2	129.0	160.6	49.7	169.9	47.8	20.6	4.0	13.0	204.2	64.7	214.3	2806.6
		Mean	525.1	25.1	31.4	57.5	242.0	31.7	58.7	74.4	18.7	57.6	5.5	2.1	0.6	2.8	28.0	15.7	104.0	1178.1
		Median	337.8	17.8	14.9	36.6	148.2	20.7	57.8	70.4	14.4	50.7	0.6	0.1	0.1	0.4	1.3	6.7	102.2	947.9
		D.F.[b]	100	94	100	100	100	100	100	100	94	100	69	50	75	75	69	81	100	100
	Summer	Minimum	98.6	2.4	1.3	30.8	129.4	3.3	12.4	20.7	1.0	0.6	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	15.0	350.2
		Maximum	1938.1	140.3	116.7	419.8	1133.7	149.4	459.9	302.2	153.0	320.9	37.0	27.2	3.7	29.6	45.1	40.0	167.6	4216.8
		Mean	821.2	44.2	43.8	109.3	398.1	52.9	151.7	127.5	41.1	72.6	3.6	4.7	0.7	3.2	8.0	7.8	68.7	1890.2
		Median	598.1	42.8	36.2	81.0	257.9	30.4	119.4	124.0	22.7	49.4	0.4	0.8	0.3	0.4	1.6	2.4	23.9	1615.7
		D.F.	100	100	100	100	100	100	100	100	100	100	69	75	81	69	81	75	100	100
Shellfish	Winter	Minimum	48.6	<LOD	15.2	9.8	62.8	6.7	24.4	28.8	1.4	1.0	<LOD	0.8	0.1	<LOD	<LOD	<LOD	3.2	254.0
		Maximum	281.0	17.1	210.8	200.7	761.1	209.0	293.0	264.7	23.2	66.3	34.9	21.2	5.1	30.0	163.9	57.4	468.9	2098.3
		Mean	135.0	4.7	69.0	71.6	247.3	61.7	102.7	115.6	7.9	16.4	12.7	7.6	1.8	4.8	35.3	17.4	133.6	911.7
		Median	103.5	1.6	50.2	27.1	108.0	34.0	81.7	96.3	5.0	6.5	7.9	3.6	1.3	1.0	2.7	4.1	106.0	524.0
		D.F.	100	75	100	100	100	100	100	100	100	100	88	100	100	75	63	63	100	100
	Summer	Minimum	22.7	0.5	2.1	2.7	28.6	2.2	8.1	9.2	1.7	3.2	<LOD	<LOD	<LOD	<LOD	<LOD	0.7	7.4	117.9
		Maximum	225.1	38.6	69.1	193.0	453.1	201.0	173.2	270.9	80.0	95.4	34.1	50.3	7.6	32.8	218.0	81.4	272.9	1694.3
		Mean	94.8	9.6	23.1	53.7	153.1	44.2	70.9	87.2	29.4	40.1	11.0	14.6	2.1	8.5	32.3	17.7	123.3	692.2
		Median	64.2	4.5	14.8	28.1	68.0	17.4	61.7	31.4	17.8	40.7	4.4	6.3	0.4	2.5	4.2	1.9	146.7	477.3
		D.F.	100	100	100	100	100	100	100	100	100	100	63	88	88	75	88	100	100	100

[a] Limit of detection; [b] Detection frequency (%); Please refer to Table C-2 (Appendix C) for the LOD values of investigated PAHs; [c] Sum of seven carcinogenic PAHs (BaA, Chr, BbF, BkF, BaP, DahA and IP); [d] Sum of 16 USEPA priority PAHs; The concentrations of congeners with <LOD were assumed to be zero while calculating the total PAHs.

occurrence and continuous accumulation of these compounds in the Bangladeshi seafood. The most frequently detected compounds were Nap, Ace, Flu, Phe, Ant, Flt, Pyr, and Chr, which had detection frequencies of 100% in both seasons. The other, less frequently detected compounds were Acel, BaA, BbF, BkF, BaP, DahA, BghiP, and IP, which had detection frequencies of 50–100% depending on the season and source. In general, low and medium molecular weight PAHs have higher detection rates than the high molecular weight PAHs (**Table 6-9**). This can be attributed to high molecular weight PAHs making the bioconcentration more difficult (Ni and Guo, 2013; Moon et al., 2010). The total concentrations of the PAHs (∑PAHs, sum of 16 USEPA priority PAHs) in the seafood samples ranged from 184.5 to 2806.6 ng/g ww in winter, and from 117.9 to 4216.8 ng/g ww in summer. The sample frequencies by class of ∑PAHs levels were: 54% (>1000 ng/g ww), 17% (500–100 ng/g ww), 29% (<500 ng/g ww). The sum of seven carcinogenic PAHs (∑C-PAHs) were 10.1–214.3 ng/g ww in winter, and 3.23–468.9 ng/g ww in summer, accounting <1–33% of the ∑PAHs concentrations.

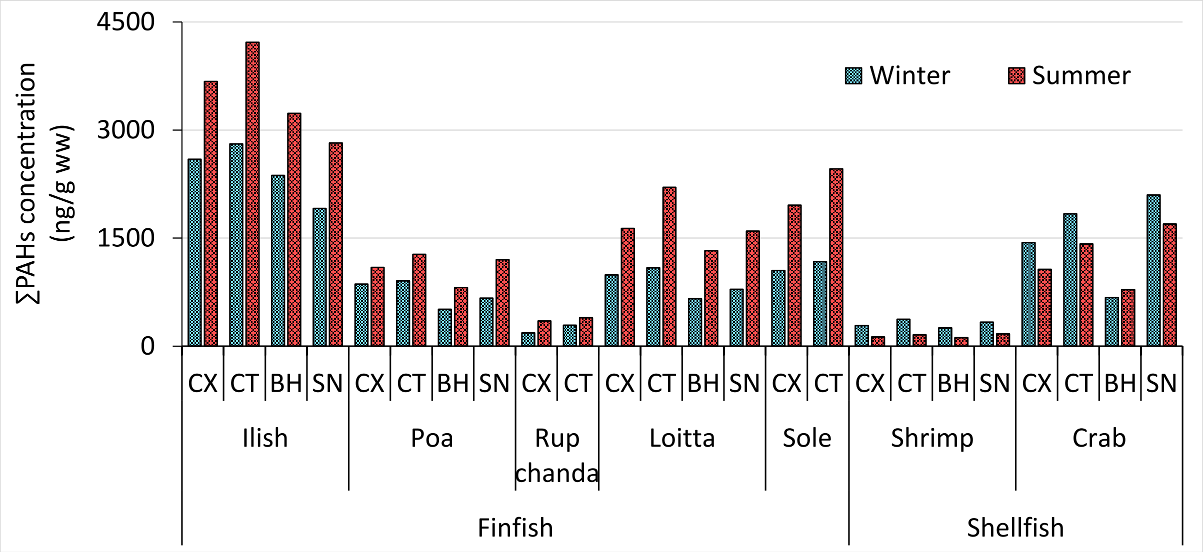

Figure 6-11. Concentrations of ∑PAHs in the Bangladeshi seafood (finfish and shellfish) in winter and summer. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans.

The levels of PAHs in the Bangladeshi seafood were compared with that of from other areas around the world and presented in **Table 6-10**. To some degree, the results of comparison could at least reflect the status of PAH contamination in the Bangladeshi seafood, although the numbers and types of monitored PAHs, the sampling time and methods, and the investigated species among these studies might be different from each other. The comparison results revealed that the ∑PAHs concentrations for both seasons in this study were comparable or higher than those observed in finfish and/or shellfish from the Daya Bay and Guangdong Province, South China (Sun et al., 2016; Ni and Guo, 2013), Mumbai Harbour, India (Dhananjayan and Muralidharan, 2012), Atlantic Ocean (Ramalhosa et al., 2012), Athabasca and Slave Rivers, Canada (Ohiozebau et al., 2016), Adriatic Sea, Italy (Perugini et al., 2007), Niger Delta, Nigeria (Effiong et al., 2016), Busan, South Korea (Moon et al., 2010), Hong Kong (Cheung et al., 2007) and Ariake Sea, Japan (Nakata et al., 2003), but lower than that from Haimen Bay, China (Shi et al., 2016). In general, the comparisons with other studies indicated that the seafood in this area were highly contaminated with PAHs. Furthermore, the mean concentrations of sum of four indicator PAHs (BaA, Chr, BbF, and BaP) in % of samples exceeded the 12.0 ng/g ww permissible limit set by the European Commission (EC, 2011). Therefore, the present level of PAHs measured in the Bangladeshi seafood is obviously a matter of concern in terms of both ecological and human health.

Table 6-10. Comparison of total PAH concentrations (ng/g ww) in seafood around the world.

Locations	Species	Sampling year	*N*[a]	∑PAHs	References
Daya Bay, South China	Finfish and shellfish	2012	16	110–520	Sun et al., 2016
Guangdong Province, South China	Shellfish	2005	16	<0.125–32	Ni and Guo, 2013
Haimen Bay, China	Finfish	2010	16	388.5–5640	Shi et al., 2016
Mumbai Harbour, India	Finfish	2006–08	15	17.43–70.44	Dhananjayan and Muralidharan, 2012
Atlantic Ocean	Finfish	2007–09	16	1.8–19.9	Ramalhosa et al., 2012
Athabasca and Slave Rivers, Canada	Finfish	2011–12	16	11–120	Ohiozebau et al., 2016
Adriatic Sea, Italy	Finfish and shellfish	2004	13	14.74–63.33	Perugini et al., 2007
Hong Kong	Finfish and shellfish	2004	16	15.5–118.0	Cheung et al., 2007
Niger Delta, Nigeria	Finfish	2013	16	20–1734	Effiong et al., 2016
Busan, South Korea	Finfish and shellfish	2005–07	16	4.73–87.8	Moon et al., 2010
Ariake Sea, Japan	Finfish and shellfish	2001	16	<0.04–18	Nakata et al., 2003
Huelva, Spain	Finfish and shellfish	2001	16	8.22–71.4	Bordajandi et al., 2004
Coastal area of Bangladesh	Finfish and shellfish	2015	16	184.5–2806.6 (W)[b] 117.9–4216.8 (S)[b]	This study

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

6.3.3.2 Influences of seasons and species on PAH accumulation in seafood

The levels of ∑PAHs in finfish were significantly higher in summer (350.2–4216.8 ng/g ww, mean: 1178.1 ng/g ww) than in winter (184.5–2806.6 ng/g ww, mean: 1890.2 ng/g ww) ($p < 0.05$). On the contrary, the concentrations of ∑PAHs in shellfish were slightly higher in winter (5.16–37.7 ng/g ww, mean: 911.7 ng/g ww) compared to summer (3.82–30.06 ng/g ww, mean: 692.2 ng/g ww), although this difference was not statistically significant ($p > 0.05$). (**Table 6-9** and **Figure 6-11**). These variations might be attributed to the seasonal discrepancies in their physiological activities, feeding behavior as well as the degree of contamination of their habitats and these will be explained in details in the following discussions. As a whole, the loads of ∑PAHs in the Bangladeshi seafood after combining finfish and shellfish data together did not differed significantly between winter and summer ($p > 0.05$), elucidating that the present study area might be influenced by almost similar sources in the two seasons and that are mainly from local inputs. In addition, regarding individual PAHs, for both the finfish and shellfish species, the majority of the monitored PAHs did not show clear seasonal variation, excepting the concentrations of Flt and Pyr in finfish which differed significantly between seasons ($p < 0.05$) with relatively higher concentrations in summer samples (**Table 6-9**).

Significant interspecies differences were found with regard to ∑PAHs concentrations in the seafood ($p < 0.05$). In particular, the highest levels of ∑PAHs were measured in Ilish (1911.0 –2806.6 and 2820.2–4216.8 ng/g ww in winter and summer, respectively) among the finfish species and Crab (673.9–2098.3 and 784.7–1694.3 ng/g ww in winter and summer, respectively) within the shellfish species, probably due to the elevated lipid content in these species (Table A-10 and A-11, Appendix A). PAHs accumulate in adipose tissue and thus tend to increase with lipid content (Ramalhosa et al., 2012; Sun et al., 2016). Our analysis revealed a positive and strong correlation between the levels of PAH and the lipid content in the studied seafood samples ($r = 0.78$; $p < 0.01$). In finfish, irrespective to season, the concentrations of ∑PAHs showed the following trend: Ilish > Sole > Loitta > Poa > Rupchanda. This pattern correlates well with lipid yields obtained from the individual seafood species. Besides the species-specific lipid content, also biological (trophic level, feeding behavior, reproductive status and metabolism including uptake and elimination) and environmental parameters (depth and habitat location) might influence the pollutant body burden. Comparatively higher PAHs levels in Ilish could be explained by the following phenomena: [1] It is a high trophic carnivorous species that tends to concentrate contaminants to a higher degree than other species (Das and Das, 2004; Miao et al., 2000); [2] It is an anadromous species which migrates from the sea to the rivers for spawning. Consequently, this species is habitated to different ecosystems (marine, estuarine, brackish and freshwater) and exposed to various degree of contamination; [3] Ilish has relatively a larger body surface area with a very thin layer of skin and a larger gill surface area that facilitate the process of accumulation of the contaminants like PAHs from water column into the fish body. Previous studies reported that bioconcentration from water via the skin and gills, and ingestion of contaminated food and/or sediments are potential routes to accumulate PAHs in fish (Van der Oost et al., 2003; Cheung et al., 2007; Shi et al., 2016). In addition, there were elevated concentrations of PAHs in the Sole tissue (1051.7 –1172.8 and 1957.5–2460.0 ng/g ww in winter and summer, respectively), which was perhaps relative to their living and feeding habits. PAHs were prone to sink in the sediments where the benthic fish such as Sole often predate benthic organisms or eat some organic particles, and so tend to accumulate these organic compounds like PAHs.

Within shellfish species, ∑PAHs levels in Shrimp were consistently low and the low extractable lipids of Shrimp (2.1 ± 0.4%) were probably related to these observations. This can also be partially explained by their pelagic nature and feeding pattern. Shrimp live slightly above the seabed, resulting in less intense contact with the sediment compared to the other exclusively benthic species, Crab. Shrimp primarily feed on mysids and amphipods (Oh et al., 2001), that occupy a low trophic level. The other shellfish species (Crab) contained relatively higher PAH levels and that can be attributed to their relatively high lipid content and their living and feeding habit as well. In particular, Crab is a typical benthic organism, also known as a scavenger that tends to feed partially on detritus or decaying organic material (such as carcasses of dead fish and other organisms), which can bear relatively high pollutant loads (Everaarts et al., 1998; Ip et al., 2005). Organisms living in close relationship with sediment such as crabs, are more likely to have greater exposures to PAH in a polluted environment than those living in pelagic environments (Ohiozebau et al., 2016). Benthic organisms can accumulated PAH via several routes, such as direct contact with sediment, respiration of interstitial water and incidental ingestion of sediments. Moreover, crabs also possess gills with a relatively larger surface area which could facilitate the accumulation of PAHs into their bodies since gills can continuously transfer the pollutants from both water and suspended particles onto its surface that are subsequently distributed throughout the whole body via blood.

Another important factor that can play a key role in determining the contaminant burdens between the species is the depth at which organisms live. In this study, Ilish, Sole and Crab being organisms inhabiting deeper waters showed elevated concentrations of PAHs. Several studies reported higher concentrations of certain POPs in seafood from deeper waters (de Brito et al., 2002; Storelli et al., 2009). In general, the concentrations of ∑PAHs in the finfish (average of 1178.1 and 1890.2 ng/g ww in winter and summer, respectively) were significantly higher than that in the shellfish (average of 911.7 and 692.2 ng/g ww in winter and summer, respectively) ($p < 0.05$). This indicated that the finfish were more contaminated with PAHs than the shellfish and that were due to higher potential of PAH bioaccumulation in finfish compared to the shellfish species (Perugini et al., 2007; Sun et al., 2016). Interestingly, the concentrations of ∑C-PAHs were higher in shellfish (average of 133.6 and 123.3 ng/g ww in winter and summer, respectively) than the finfish samples (average of 104.0 and 68.7 ng/g ww in winter and summer, respectively). This might be caused by comparatively higher metabolic rate of these PAHs in finfish than the shellfish species.

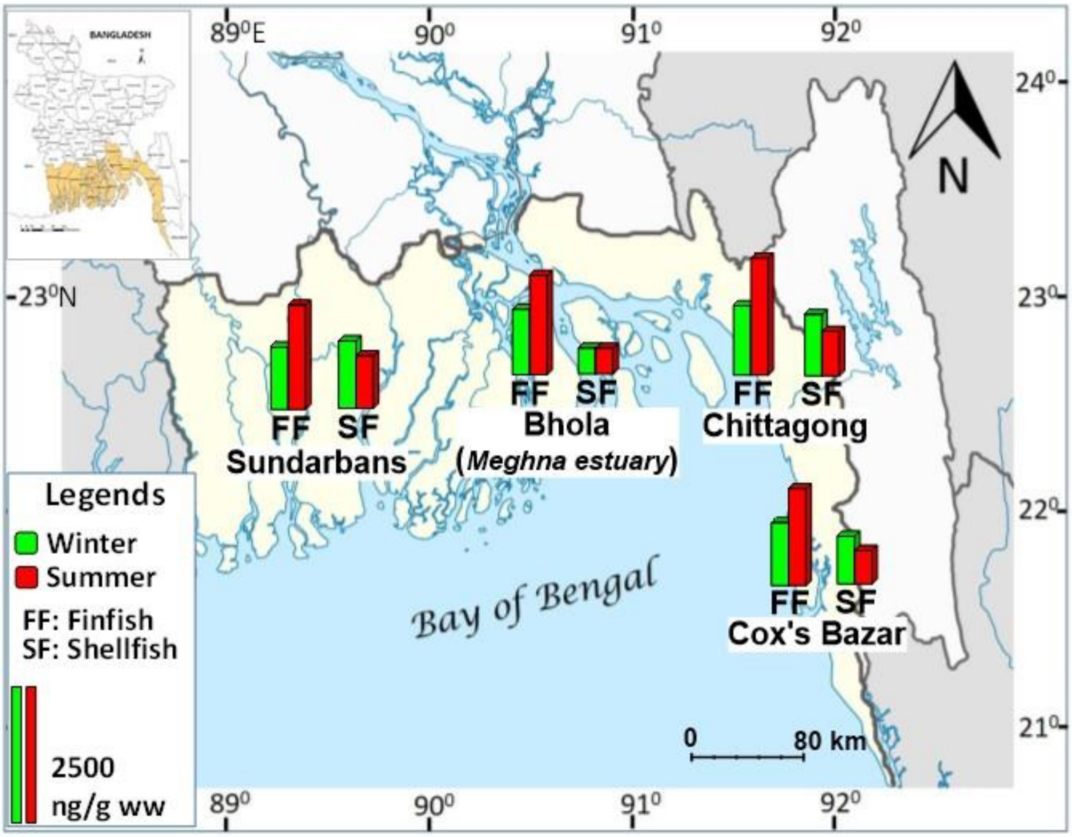

Figure 6-12. Spatial distribution of average ∑PAHs in seafood (finfish and shellfish) from the coastal area of Bangladesh. Colored area in the inset map represents the coastal area of Bangladesh.

6.3.3.3 Spatial distribution

The spatial distributions of PAHs were investigated in seafood collected from the coastal area of Bangladesh and the variations in the average of ∑PAHs are presented in **Figure 6-12**. Geographically, no significant differences in ∑PAHs concentrations among the four coastal areas were observed ($p > 0.05$). However, regarding finfish, the highest mean concentration of ∑PAHs were found in samples from Chittagong in both seasons (1253.2 ng/g ww in winter, 2109.8 ng/g ww in summer). On the other hand, shellfish samples from Chittagong also exhibited the maximum mean levels of ∑PAHs (1105.8 ng/g ww in winter, 788.0 ng/g ww in summer). In general, the mean concentration of ∑PAHs seemed to have the following trends: Chittagong > Cox's Bazar > Bhola > Sundarbans, and Chittagong > Sundarbans > Cox's Bazar > Bhola in finfish and shellfish, respectively (**Figure 6-12**). Overall, the seafood from Chittagong, Sundarbans, and Cox's Bazar area was more contaminated with PAHs than that from Bhola (Meghna estuary), and because of greater economic development, thus associating these compounds to urbanization and industrialization.

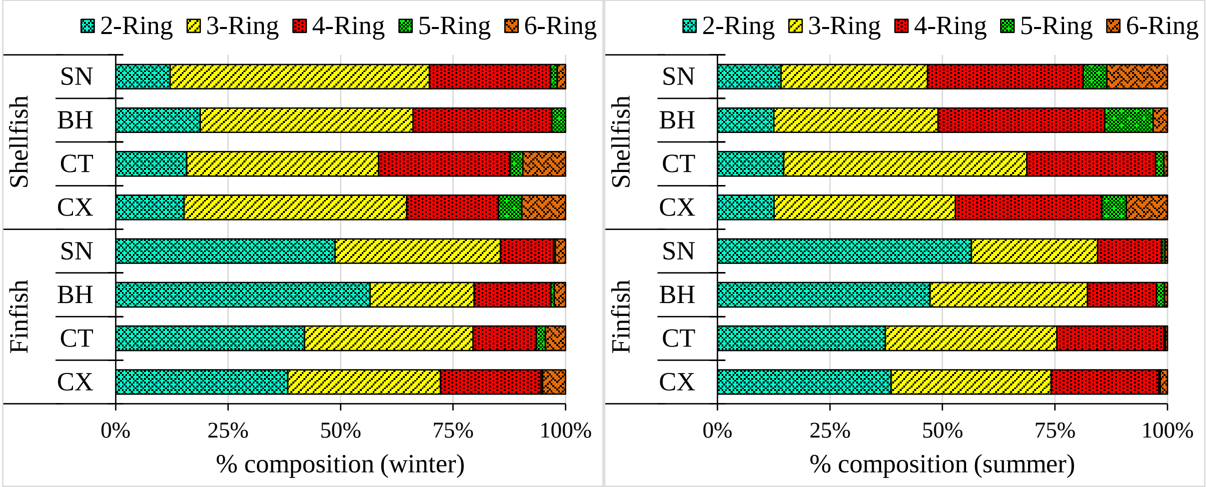

Figure 6-13. Compositional profiles of PAH compounds in the Bangladeshi seafood (finfish and shellfish) in winter and summer. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans.

6.3.3.4 PAH composition in seafood and source identification

The composition patterns and relative abundance of PAHs by the number of aromatic rings in the Bangladeshi seafood (finfish and shellfish) are illustrated in **Figure 6-13**. No significant difference was observed in the PAHs profile between the two seasons ($p > 0.05$). However, the profile significantly differed between finfish and shellfish ($p < 0.05$), although the LMW PAHs (2- and 3-ring) dominated the profile for both cases, which collectively constituted 49%–86% (mean: 69%) of the total PAHs (**Figure 6-13**). The HMW PAHs (4-, 5-, and 6-ring) contributed less 14%–53% (mean: 31%). These PAH profiles are similar to those reported for marine organisms in other countries (Nakata et al., 2003; Liang et al., 2007; Perugini et al., 2007; Cheung et al., 2007; Ni and Guo, 2013; Ohiozebau et al., 2016; Sun et al., 2016). In particular, Nap (2-ring) in finfish and Phe (3-ring) in shellfish species was the most abundant, contributing 18%–65% and 13%–36% to the ∑PAHs, respectively, and that are primarily due to their lesser affinity for particles and greater water solubility (Ohiozebau et al., 2016; Sun et al., 2016). Therefore, these non-metabolized PAHs can undergo bioaccumulation through direct exchange between the water column and the gills (Thomann and Komlos, 1999; Liang et al., 2007). However, the abundance of these particular PAHs in finfish and shellfish was also reported in several studies (Cheung et al., 2007; Moon et al., 2010; Shi et al., 2016). Interestingly, accumulation of HMW PAHs was higher in benthic organisms including Sole, Poa, Shrimp and Crab (15%–54%), suggesting sediments may be a more important PAH source for benthic organisms than pelagic fish in this area (Loutfy et al. 2007; Sun et al., 2016).

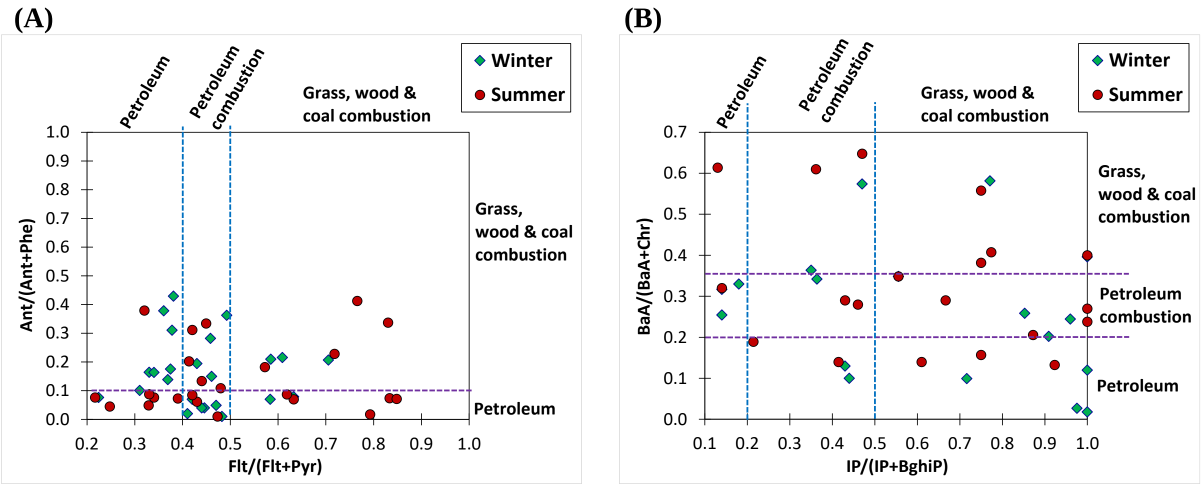

Figure 6-14. PAH cross-plots for the ratios of (A) Ant/(Ant+Phe) vs. Flt/(Flt+Pyr), (B) BaA/(BaA+Chr) vs. IP/(IP+BghiP).

Sources of PAHs are often inferred by some special molecular indices based on the ratios of selected PAHs Yunker et al., 2002; Katsoyiannis and Breivik, 2014). Among these molecular ratios, Ant/(Ant + Phe), Flt/(Flt + Pyr), BaA/(BaA + Chr) and IP/(IP + BghiP) are more widely used (Yunker et al., 2002; Tobiszewski and Namiesnik, 2012). Although the use of PAH diagnostic ratios for source characterization has been criticized (Katsoyiannis et al., 2007; Galarneau, 2008), it has been used widely and proven to effectively differentiate PAH sources (Jiang et al., 2009; Martins et al., 2010; Xue et al., 2013) and consequently, in the present study it is utilized as an indicative tool of PAHs source and distribution.

The criteria for PAH ratios as indicators of PAH sources are listed in **Table 6-7** and the plotted isomeric ratios of: Ant/(Ant + Phe) vs. Flt/(Flt + Pyr), and BaA/(BaA + Chr) vs. IP/(IP + BghiP) used in this study are shown in **Figure 6-14**. The values of Ant/(Ant + Phe) were 0.01–0.41, 42% of which were higher than 0.1; the ratios of Flt/(Flt + Pyr) were less than 0.4 in 31%, 0.4–0.5 in 40% and higher than 0.5 in 29% of samples; the values of BaA/(BaA + Chr) were less than 0.2 in 25%, 0.2–0.35 in 31% and higher than 0.35 in 44% of samples; the ratios of IP/(IP + BghiP) were less than 0.2 in 29%, 0.2–0.5 in 23% and higher than 0.5 in 48% of samples (**Figure 6-14**). The above data demonstrated that the PAHs in seafood from the Bangladeshi coastal area originated from both the petrogenic and pyrogenic sources including crude petroleum (e.g. gasoline/diesel), petroleum combustion, and combustion of grass, wood and coal. Overall, the identified ‘mixed’ type sources of PAHs contamination in the coastal area of Bangladesh may include shipping oil spills, fuel combustion of fishing boats and ship engines, industrial wastewater discharges, land runoff, riverine inputs, port activities, waste incineration, city sewage drainage discharges and industrial combustion that are transported to this area by direct dry and wet deposition from the atmosphere and rainwater runoff. However, the identification of PAH sources still remains an approximation, and must be done with caution, depending on the metabolization of individual PAHs that are affected by species and trophic states. Furthermore, the fingerprint of PAHs may be altered by biological (e.g., bacterial activity), chemical (e.g., oxidation and reduction), and/or physical (e.g., air mass mixing and sediment resuspension) processes during transport and after deposition into the environment (Soclo et al., 2000; Barhoumi et al., 2016).

6.3.3.5 Assessment of potential human health risks

Human exposure to dietary PAHs is of great concern due to their documented toxic effects including carcinogenicity and mutagenicity (Mumtaz et al., 1996; Cheung et al., 2007; Moon et al., 2010; Xia et al., 2010; Ni and Guo, 2013), however, reports concerning dietary health risk assessment for PAHs are still scarce. Since seafood contributes a major portion in the diet for the Bangladeshi coastal population, the consumption of seafood contaminated with

PAHs can be a potential risk for the consumers. Presently, there is no specific criteria for PAHs in seafood in Bangladesh. Therefore, to understand the magnitude of dietary exposure of PAHs by seafood consumption, the maximum admissible level of BaP set by the European Union (EU, 2005), and the recommended potency equivalent concentration (PEC) of total PAHs relative to BaP recommended by USEPA (USEPA, 2000) were evaluated in this study. BaP is the only PAH which has been well characterized toxicologically and is the most potent carcinogen among all individual PAHs (Agarwal et al., 2009).

Since February 2005, the European Union has recommended the maximum admissible levels of 2 ng/g and 5 ng/g ww for BaP in fish and crustaceans, respectively. In this study, fish and crustaceans are termed as finfish and shellfish, respectively. BaP was detected in 75–100% of the analyzed samples depending on the species, with concentrations of <LOD–4.05 ng/g ww in finfish species and <LOD–7.6 ng/g ww in shellfish species, respectively (**Table 6-9** and Table C-8 and C-9, Appendix C). Within finfish, the concentrations of BaP in Ilish from Cox' Bazar (2.4 ng/g ww in winter and 2.6 ng/g ww in summer) and Chittagong (4.0 ng/g ww in winter and 3.7 ng/g ww in summer) exceeded the maximum admissible level set by the European Union. Within shellfish, the elevated levels of BaP were measured in Crab from Bhola (winter, 5.1 ng/g ww), Chittagong (summer, 5.1 ng/g ww) and Sundarbans (summer, 7.6 ng/g ww) that exceeded the maximum admissible level.

The PEC could be a better index for the potential toxicity than PAHs concentration (Ding et al., 2012, 2013; Shi et al., 2016). Therefore, PEC of total PAHs for each seafood species was calculated according to the following equation:

$$\mathrm{PEC}=\sum_{i=1}^{n}\mathrm{TEF}_i\times\mathrm{C}_i \qquad (6.5)$$

where TEF is the toxic equivalent factor of individual PAHs and C is the concentration of each PAH in the sample. The calculated TEF is 0.001 for Nap, Acy, Ace, Fl, Phe, Flu and Pyr, 0.01 for Ant, Chr and BghiP, 0.1 for BaA, BbF, BkF and IP, 1 for BaP and DBahA (USEPA, 1993, 2000; Nisbet and LaGoy, 1992).

The PEC of total PAHs (ng/g ww) in the Bangladeshi seafood ranged from 1.02 (Rupchanda from Chittagong) to 49.6 (Crab from Cox's Bazar) in winter and 0.71 (Shrimp from Cox's Bazar) to 67.8 (Ilish from Bhola) in summer (**Figure 6-15**). The PEC values in the present study were higher than those in seafood from the Adriatic Sea, Italy (0.03–2.8 ng/g ww, Perugini et al., 2007), Daya Bay, China (0.57–10 ng/g ww, Sun et al., 2016), Istanbul Strait and Marmara Sea, Turkey (1.6–41 ng/g ww, Karacik et al., 2009)and those in freshwater and marine fish from Hong Kong markets (0.02–0.37 ng/g ww, Cheung et al., 2007), but lower than those in edible fish muscle from the largest freshwater lake, Poyang Lake, in China (79.46–1334 ng/g ww, Shi et al., 2016).

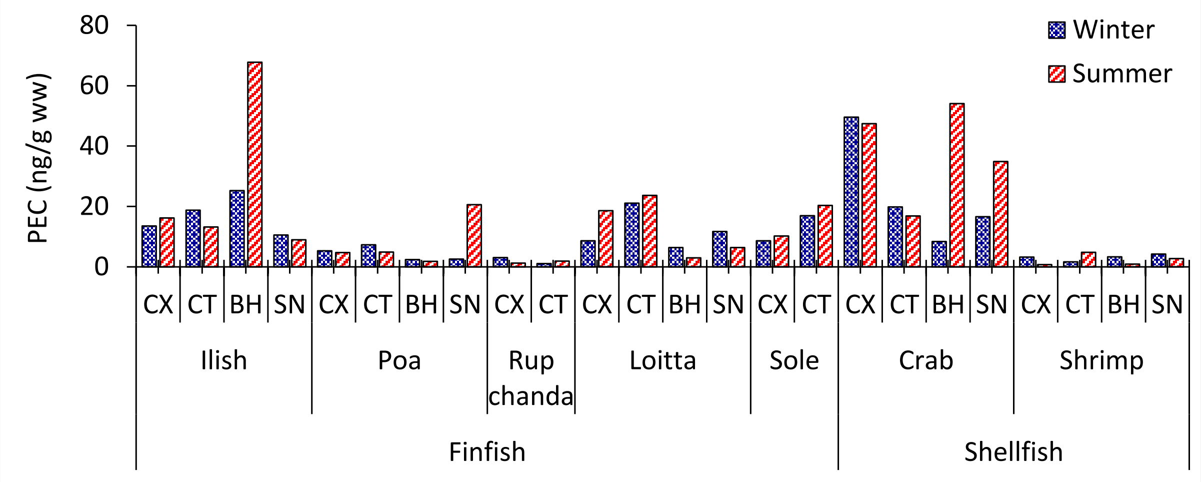

Figure 6-15. The calculated potency equivalent concentration (PEC, ng/g ww) in the Bangladeshi seafood.

The USEPA has suggested a screening value (SV) of the PEC of total PAHs for use in fish advisories which is defined as the concentration of the chemical contaminant in edible tissue that is of potential public health concern (USEPA, 2000). In this study, the calculated PEC values for all of the studied seafood species were higher than the recommended PEC of total PAHs (0.67 ng/g ww) for human consumption, indicating that consuming these PAH contaminated seafood will obviously induce adverse health effects on the Bangladeshi coastal residents including other consumers as well.

The cancer risks posed by dietary exposure to PAHs were evaluated using the following equation (USEPA, 1992):

$$\mathrm{ILCR}=\frac{\mathrm{PEC}\times\mathrm{CR}\times\mathrm{EF}\times\mathrm{ED}\times\mathrm{SF}\times\mathrm{CF}}{\mathrm{BW}\times\mathrm{AT}} \qquad (6.6)$$

where ILCR is the incremental lifetime cancer risk; CR is the consumption rate (g/d); EF is the exposure frequency (365 day/year); ED is the exposure duration (year, adults = 53 years, Children = 12 years); SF is the oral cancer slope factor (7.3 (mg/kg-d)$^{-1}$); BW is the average body weight (60 kg for adults and 25 kg for children); AT is the average lifespan for carcinogens (25,550 days); and CF is the conversion factor (10^{-6} mg/ng). A public screening criteria for carcinogens which is set at a carcinogenic risk level of 1.0E−06 was used for assessment. One in a million chance of additional human cancer over a 70 year lifetime (ILCR = 1.0E−06) is the level of risk considered acceptable or inconsequential, while the additional lifetime cancer risk of one in ten thousand or greater (ILCR = 1.0E−04) is considered serious and there is high priority for paying attention to such health problems (USEPA, 1994; FDA, 2010). The daily consumption data of seafood for the adults (≥18 years) and children (6–17 years) in the coastal area of Bangladesh were obtained from a questionnaire based dietary survey during our sampling campaign. The seafood consumption data for the adults and children in the coastal area of Bangladesh are given in Table C-10 (Appendix C).

Table 6-11. Incremental lifetime cancer risk (ILCR) for adults and children in the coastal area of Bangladesh.

Coastal area	ILCR	
	Adults	Children
Cox's Bazar	3.4E-04	9.0E-05
Chittagong	2.4E-04	6.8E-05
Bhola (Meghna Estuary)	2.7E-04	7.7E-05
Sundarbans	2.1E-04	5.6E-05

The ILCR values resulting from dietary exposure to 16 USEPA PAHs via consumption of these seafood ranged from 2.1E−04 to 3.4E−04 for adults and 5.6E−05 to 9.0E−05 for children, respectively (**Table 6-11**). The results indicated that the ILCR values for children were higher than the maximum acceptable risk level, whereas the values for adults were higher than the priority risk level, indicating a potential carcinogenic risk to coastal residents in Bangladesh. Furthermore, people could be exposed to PAHs through other routes (dietary and non-dietary routes) as well and the real cancer risk for local people would be greater than that estimated in this study (Ding et al., 2012). However, it is an urgent need to take appropriate measures to control the health hazards due to dietary PAH exposure in Bangladesh.

6.4 Conclusion

The concentrations of 16 USEPA priority PAHs were measured in the surface water, sediment, and seafood (5 finfish and 2 shellfish species) from the coastal areas of Bangladesh.

The samples were collected in winter (January-February) and summer (August-September), 2015. All of the 16 PAHs were detected in the three matrices. The levels of PAHs ranged from moderate to a relatively high PAH pollution compared to other coastal areas worldwide. In water, sediment and seafood samples, the most abundant PAHs by ring structures were 2- to 3- ring, 4- to 5- ring, and 2- to 3- ring, respectively., suggesting low to moderate molecular weight PAHs were prevalent in this area. The concentrations of ∑PAHs in the three medium were slightly higher in summer than those in winter, but the seasonal variations were not statistically significant in any cases, thus suggesting inputs of PAHs from almost similar sources in the two seasons. Some selected molecular ratios demonstrated that the PAHs in the Bangladeshi coastal area originated from both the petrogenic and pyrogenic sources including crude petroleum (e.g. gasoline/diesel), petroleum combustion, and combustion of grass, wood and coal. Spatial distribution revealed that the Chittagong, Sundarbans and Cox's Bazar areas were more contaminated with PAHs than the Meghna Estuary and because of greater development, thus associating these compounds to urbanization and industrialization. The PAHs concentrations in water, sediment and tissue exceeded the guidance levels in a high percentage of samples and thus, could adversely affect the ecological environment and human health through biomagnification. Besides, the dietary intakes of total PAHs through the analyzed seafood species for the adults and children of the Bangladeshi coastal area exceeded the USEPA proposed health based guidelines, suggesting a significant health risk in terms of toxicological effects of PAHs. Since the toxic effects might be caused to the ecosystem, some control measures or remedial actions should be undertaken to decrease the PAH contamination. Furthermore, long term continuous monitoring should be conducted to develop effective management strategies as well as the existing legal policies should be utilized to control the PAH pollution in the coastal area of Bangladesh.

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Chapter 7

GENERAL DISCUSSION, CONCLUSIONS AND RECOMMENDATIONS

General Discussion, Conclusions and Recommendations

7.1 Discussion

This study was initiated to assess certain POPs including PCBs, PFAAs, and PAHs in the surface water, sediment, and seafood (5 finfish and 2 shellfish species) from the coastal areas of Bangladesh. This is the first comprehensive report of these contaminants with a wide-scale monitoring campaign along the coastline of the Bay of Bengal in Bangladesh. The study helped us to figure out broadly the present situation, existing problems and challenges in terms of POPs pollution in the Bangladeshi coastal area. Despite the lack of sufficient evidence, we tried to find out some relevant information regarding the major source factors implicated in the emission of these pollutants in Bangladesh. The influence of seasons on the distribution of these pollutants in this complex tropical coastal ecosystem was carefully examined. A preliminary ecological risk assessment was carried out to determine the hazards of these contaminants to the ecosystem integrity in the Bangladeshi coastal area. Furthermore, we also assessed the health risk for the Bangladeshi coastal residents that might be posed from the dietary exposure to these toxic chemicals through seafood consumption.

7.1.1 PCBs in surface water and sediment

The concentrations of PCBs with levels of concern were measured in water and sediments from the Bangladeshi coastal areas. Comparing globally, the present data were at the middle range and that the study area was recognized as moderately polluted in terms of PCB pollution. The PCB congener and homolog profiles in environmental matrices can often provide valuable information on the environmental source, transport and fate processes of PCBs. In this study, a full profile of PCBs (209 congeners belonging to mono- to deca-CB homologs) was assessed. However, the dominant congeners identified in the present study corresponds to diverse homolog groups (tri- to hepta-CBs), suggesting multiple sources of PCBs in this area. The presence of different congeners corresponding to different homologs is an indication of various sources of PCBs in marine or coastal environments (Sahu et al., 2009). In addition, the majority of the most abundant congeners in sediment samples are enrich in Aroclor 1254 and 1260 or equivalents. However, the prevailing abundance of these Aroclor congeners in recently deposited sediments is of interest because its production and/or application was already banned decades ago, suggesting recent inputs and/or ongoing usage of PCB compounds in Bangladesh. Based on the data in hand, no clear seasonal variation was reported in the levels of PCBs either in water or sediment. However, slightly higher concentrations were noticeable in summer samples for most of the sampling sites. These variations might be due to the precipitation and surface runoff from heavily contaminated sites resulted from heavy rain and floods, and intense shipping and fishing activities particularly in summer. To some extent, emission from seasonally operated industries and/or factories might also be a responsible factor for the seasonal fluctuations of environmental PCBs. The PCB concentrations varied widely depending on the locations. The highest concentrations of PCBs in each medium were recorded from Chittagong ship breaking area (**Figure 3-2** and **3-4**). Dismantling of old ships is a significant source of PCB emission into the environment (Neşer et al., 2012; Siddiquee et al., 2012; Nøst et al., 2015). It was estimated that each old ship contained 250 kg of PCBs, inclusive of transformer oil (DoE, 2007). In Bangladesh, about 50–90 ships are dismantled annually. Therefore, ship breaking activities act as a prominent local emission source of PCBs in Bangladesh. The outdated PCB-containing equipment, (old transformers, capacitors, etc.) along with non-recycled e-waste mostly originated from ship breaking industries is simply deposited in landfills in this area. Therefore, leakage from the PCB-containing in-service equipment and leachates from landfill sites might be considered as potential sources of significant PCB inputs into the Bangladeshi coastal area. Elevated levels of PCBs were also recorded from sites of shipping port and fishing harbor (e.g. sites CT1, CT2, CX3, and SN3). The use of paints and grease repellents for ship and boat protection could contribute to the relatively high levels of PCBs in port area (Rajendan et al., 2005, Wurl and Obbard, 2005). Moreover, burning of waste at waste dump sites is common in Bangladesh, and these combustion processes can release many POPs including PCBs. There are several waste dump and landfill sites in Chittagong and Cox’s Bazar which also could be contributing to the elevated levels of PCBs at these sites. Discharging of untreated or semi treated wastewater into this area from numerous multipurpose industries including municipal sewage might also be contributing to the PCB pollution. Furthermore, the Meghna Estuary is an exclusively unindustrialized area. Therefore, lower but detectable levels of PCBs in this area (ME1–ME3) suggest that non-point sources such as surface runoff due to heavy rain and flooding, runoff from upstream inland rivers and tributaries, atmospheric wet and dry deposition, etc. may also be responsible factors for PCB pollution although to a small extent.

7.1.2 PFAAs in surface water and sediment

All of the fifteen target PFAAs were detectable in surface waters and sediments from the coastal areas of Bangladesh. The total concentrations of the PFAAs (sum of 15 PFAAs, ∑PFAAs) in the water phase ranged from 10.6 to 45.2 ng/L in winter, and from 11.5 to 46.8 ng/L in summer, whereas in sediments the ranges were 2.48 to 8.15 ng/g dw and 1.07 to 3.81 ng/g dw in winter and summer, respectively. PFOA in water (3.17–27.8 ng/L) and PFOS in sediment samples (0.60–1.14 ng/g dw) were found to be the most abundant PFAAs, and these concentrations were comparable to or less than most other reported values, particularly those recorded from the coastal areas of China, Japan, Korea and Spain. The majority of the monitored PFAAs did not show clear seasonal variation in either medium. However, the concentration of PFHpA in water was higher in summer, and that was probably from atmospheric deposition due to heavy rainfall in this season (Simcik and Dorweiler, 2005). On the contrary, the concentrations of PFBA, PFPeA, PFOA, PFUnDA, PFBS, PFHxS, and PFOS in sediments were significantly higher in winter than those measured in summer samples. However, the seasonal variations of some particular PFAAs (PFHpA, PFUnDA, and PFBS in water and PFBA, PFPeA, PFOA, PFUnDA, PFBS, PFHxS, and PFOS in sediment) might be due to their specific seasonal usage or applications in certain industries (e.g., the textile, paper, and leather industries) or the seasonal use of consumer products, although the exact reasons for this phenomenon are still unknown. Overall, in the case of sediment samples, the mean concentration of ∑PFAAs in winter (4.89 ng/g dw) was over two times higher than in summer (2.02 ng/g dw). One possible reason behind this phenomenon can be explained as: in summer, most of the coastal area of Bangladesh (> 90%) experiences heavy flooding and massive soil erosion occurs along the river banks due to heavy rainfall. Consequently, a huge amount of fresh soil (which may be less contaminated by PFAAs) is deposited as sediment in the aquatic environment by floodwaters and eroded soil in the estuarine and coastal areas. In winter, no such environmental disturbances occur, so the winter sediment samples showed significantly higher concentrations of PFAAs in the study area.

Interestingly, different spatial trends were noticeable in the levels of ∑PFAAs in water and sediment samples (**Figure 4-4**) which might suggest a site-specific PFAAs sources in the Bangladeshi coastal area. In particular, water samples at location CX3 (Bakkhali Estuary) showed the highest levels of PFAAs for both seasons (45.2 and 46.8 ng/L in winter and summer, respectively), followed by CT1 located in the Chittagong port area (22.1 and

40.1 ng/L in winter and summer, respectively). Unlike the water samples, sediments from CT4 (ship breaking area) showed the highest concentration of total PFAAs (8.15 ng/g dw) in winter whereas CX3 (Bakkhali Estuary) showed the highest value (3.81 ng/g dw) in summer. The Bakkhali Estuary (CX3) receives huge discharges of untreated municipal and industrial effluents from the city, and the water is affected by activities such as intensive boating and fishing. Corsolini et al. (2012) found that the wastewater effluent is a point source of PFAA contamination in the Ganges River and the adjacent Sundarbans mangrove wetlands in India. The main municipal garbage dump (mostly consumer products) of Cox's Bazar city is also located near CX3. Therefore, a significant amount of PFAAs might be emitted from this dumping site into the adjacent water body. Discharging of waste generated from ship breaking and e-waste recycling activities in Chittagong area (CT3 and CT4) might be the significant PFAA pollution sources in this area. Shipping and boat maintenance in the ports (CT1) may also be responsible for the release of PFAAs into the surrounding waters (Paul et al., 2009). After scrutinizing the levels and distribution of PFAAs, industrial and municipal wastewater effluents, ship breaking and port activities were identified as some of the potential sources of the PFAA contamination in this region. Regardless of time, the concentrations of PFAAs were relatively lower both in water and sediment from the Meghna Estuary which is an exclusively unindustrialized estuarine area. In general, the concentrations of ∑PFAAs in surface water and sediment at the Cox's Bazar (CX1–CX4) and Chittagong sites (CT1–CT4) were higher than those in the Meghna Estuary (ME1–ME3) and Sundarbans sites (SN1–SN3) because of greater development, including industrialization and urbanization activities.

The correlations among individual PFAAs between water and sediment revealed that some of them in water samples (PFPeA, PFHxA, PFDA) had strong positive correlations/associations with some of PFAAs in sediment samples (PFPeA, PFHxA, PFDoDA), which suggests that these compounds probably had a common pollution source in the study area. Since this the very first study in the study area, therefore, to draw a final conclusion, the specific sources of PFAAs in the Bangladeshi coastal environment should be examined because there is currently no information on the usage, production, import, and volumes of PFAAs in Bangladesh. However, because concentrations of PFAAs in sediments were greater than those in water, sediments might be a useful integrating medium for pollution monitoring.

7.1.3 PAHs in surface water and sediment

The total concentration of PAHs (∑PAHs) in water, and sediment were 855.4–9653.7 ng/L, and 349.8–11058.8 ng/g dw in winter, and 679.4–12639.3 ng/L, and 199.9–17089.1 ng/g dw in summer, respectively. The levels of ∑PAHs were comparable or relatively high in comparison with other coastal areas around the world. Two to five ring PAHs dominated their abundances in the coastal area of Bangladesh. In particular, Nap, Flu and Phe in water and Flt, Pyr, Chr and Phe in sediment were the most abundant PAH compounds, comprising up to 36–89% (mean: 68%) and 30–90% (mean: 65%) of ∑PAHs by sites, respectively.

In water samples, all of the sampling sites showed slightly higher concentrations of ∑PAHs in summer than that in winter (**Figure 6-4**), although the differences were not statistically significant ($p > 0.05$). However, seasonal variations to an extent were noticeable in sediment samples depending on the locations. For example, CX1, CX3, CX4, CT1, CT2, CT3, CT4, SN1 and SN2 showed slightly higher concentrations in summer, whereas the rest of the locations exhibited the opposite trends (**Figure 6-8**). In summer (wet period), PAHs previously buried in the surface soil of heavily contaminated sites and accumulated in dry weather were flushed into the estuary and/or river through surface runoff due to the floods and heavy rains. Furthermore, the concentrations of PAHs in the gas phase increase in summer or in general in tropical regions (Abdel-Shafy et al., 2016) that could be redeposited through the atmospheric wet and dry deposition. Additionally, increased shipping and fishing activities during summer in the study area might also be another crucial factor for the seasonal variations. On the contrary, elevated levels of PAHs in winter samples were probably due to either co-evaporation of PAHs with water or intense sedimentation from less mixing effects due to comparatively weaker tidal movement or wave action and lower inflow from upstream rivers in the dry season as well.

Levels of PAHs in surface water and sediment differed significantly between the four coastal regions ($p < 0.05$), indicating the PAH contamination mainly influenced by the local/regional source inputs in the study areas. In general, the concentrations of ∑PAHs in surface water and sediment at the Chittagong, Cox’s Bazar and Sundarbans sites were higher in comparison to Meghna Estuary sites, which indicates that the industrialized urban regions are potential sources of PAHs and that the economic development seems to be associated with the amount of PAH emissions. Likewise, the pattern is thus well correlated with the degree of energy consumption between areas. However, the distribution pattern of ∑PAHs between sites were more or less similar in winter and summer, suggesting almost similar emission sources in the two seasons. In particular, water and sediments from at location CT1 showed the highest levels of PAHs for both seasons followed by CT3. Other two sites from Chittagong area, CT2 and CT4 also exhibited elevated PAH levels compared to other areas. CT1 and CT2 are located within the Chittagong port area which is the largest seaport in Bangladesh and heavy traffic of tankers and cargo ships is common nearby. These sites are predominantly influenced by the port and fishing harbor activities. 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 (Dudhagara et al., 2016). CT3 and CT4 are located very close to Chittagong ship breaking yard. Ship breaking activities along the coast of Chittagong may contribute to the PAH pollution in the adjacent areas, as dismantling ships produces various types of inorganic and organic pollutants, including PAHs (Neşer et al., 2012; Siddiquee et al., 2012; Dudhagara et al., 2016). Moreover, the discharge of untreated or semi-treated effluents from numerous multipurpose industries including oil refinery factories may pose a significant contribution to the PAH contamination into this area. Furthermore, residential sewage as well as vehicular emission and resulted street runoff from this densely populated city may partly contribute to the observed elevated levels of PAHs. Previous studies have identified that the ship breaking activities, leakage of diesel fuel, crude oil used in ship engines and coal from ship and heavy transport vehicles and oil seepage from industrial activities lead to high concentrations of total PAHs in the coastal sediments (Dhananjayan et al., 2012; Dudhagara et al., 2016).

Molecular ratios suggested mixed sources of PAHs in the Bangladeshi coastal area with a slight imposition of pyrolytic over the petrogenic inputs closely related to shipping and fishing activities, and industrial and municipal sewage discharge. Interestingly, it was seen from **Figure 6-7** that the ratios were more scattered in summer season compared to winter, indicating a considerable input of PAH might be occurred through atmospheric wet and dry deposition and/or land runoff during times of heavy rainfall, and that are most probably originated from non-point defuse sources, such as vehicular exhaust from cities, high intensity of fishing by motorized boats and vessels, emission mainly as black smoke from seasonally operated factories and industries (e.g. brick kiln) from nearshore or far shore areas, etc.

7.1.4 Effects of sedimentary TOC and BC on POPs

TOC is crucial for their sorption, a key process in trapping and transporting POPs in sediments (Hung et al., 2010; Hong et al., 2012; Ali et al., 2014). Within total organic moieties, BC with the highest sorption capacity has been shown to large extent affect the distribution of POPs in sediments (Accardi-Dey and Gschwend, 2002). POPs may be extensively adsorbed onto condensed carbonaceous geo-sorbents such as BC owing to its condense and aromatic structure (Semple et al., 2013). It has been reported that adsorption of various POPs by BC can be approximately two orders of magnitude higher than by sedimentary TOC (Hung et al., 2010; Ali et al., 2014 and references therein). In this study, correlation analysis was applied to investigate the influence of sedimentary TOC and BC on the distribution of POPs. The concentrations of sedimentary TOC (%) and BC (%) were recorded as 0.06–1.28 and 0.06–0.53 in winter and 0.15–0.69 and 0.06–0.28 in summer, respectively (Table A-9, Appendix A).

The correlation results revealed a significant positive association between the ∑PCBs and BC (winter: $r = 0.68$, $p < 0.001$; summer: $r = 0.71$, $p < 0.001$), whereas the correlations between ∑PCBs and TOC were not significant in either seasons (winter: $r = 0.39$, $p > 0.05$; summer: $r = 0.41$, $p > 0.05$). Interestingly, irrespective to seasons, all homolog groups of PCBs (di- to octa-CBs) showed significant positive correlations with BC ($r = 0.55$–0.67; $p < 0.001$), while in case of TOC, the correlations were positive but not significant ($r = 0.27$–0.39; $p > 0.05$), suggesting BC plays a major role in the distribution of PCBs, and that the PCB in the Bangladeshi coastal area mainly originated from local emissions or point sources, because Cui et al. (2016) reported that if the sources of sedimentary PCBs are controlled by atmospheric emission or long-range transport (non-point/diffuse sources), then the amounts of PCBs should be well correlated with the sedimentary TOC.

In the case of PFAAs, the concentrations of PFHxA, PFNA, PFDA, PFDoDA, PFBS and PFOS were found to be correlated with TOC and BC ($p < 0.01$), which is supported by previous findings reported by Pan et al. (2014). Interestingly, no such correlations were found in the summer samples may be because newly deposited coastal sediments (due to river bank erosion and/or runoff from heavy flooding and rainfall) contain lower amounts of organic compounds.

However, the correlation results revealed a significant positive association between the ∑PAHs and BC ($r = 0.56$, $p < 0.01$), whereas the correlations between ∑PAHs and TOC were positive but not significant ($r = 0.30$, $p > 0.05$). Similar observations were also reported in other studies and the possible reason may be due to an irregular and heterogeneous distribution of PAHs in the coastal or estuarine sediments (Oen et al., 2006; Liu et al., 2007; Hsieh et al., 2010). In terms of individual PAHs, none of them showed significant association with TOC ($p > 0.05$). On the contrary, Nap, Ace, Flu, Phe, Flt, Pyr, Chr, BbF, BkF, and IP were positively ($r = 0.43–0.71$) and strongly ($p < 0.01$) correlated with the BC content. Overall, the correlation between both the individual and ∑PAHs to the BC contents was stronger than the correlation to the TOC contents, indicating that PAH sorption in the studied sediments was BC-dominated. In addition, the individual PAHs that are well correlated with BC might be originated from pyrolytic sources (Li et al., 2016).

7.1.5 Ecotoxicological concern

Water-borne POPs can affect surface-dwelling and pelagic organisms whereas benthic and bentho-pelagic organisms can be affected by the sediment-bound POPs. Therefore, considering the toxicity and bioaccumulation properties of the investigated POPs (PCBs, PFAAs and PAHs), as well as diverse ecological significance of the coastal area of Bangladesh, it is of great importance to evaluate the potential risk of these pollutants in this area. Since no environmental quality guideline for POPs has been established yet in Bangladesh, we compared our results with those proposed by other nations and/or international organizations. The detailed results of the ecological risk assessment for the ascertained POPs have been discussed in the corresponding chapters (Chapter 3, 4, and 6).

Briefly, the concentrations of ∑PCBs (32.17–199.35 ng/L) in the surface water were higher than the criterion continuous concentration (CCC) (USEPA, 2010) and the Chinese national environmental quality standards for surface water (GB 3838-2002), suggesting the quality of surface water of the Bangladeshi coastal area is heavily polluted with PCBs and thus their levels could adversely affect the ecological and human health. The concentrations of PFAAs in water samples were much lower than the corresponding AA-QSs of the Italian EQSs. However, 93% of the surface water samples exceeded the EC guideline for PFOS, suggesting a potential long-term risk to the pelagic organisms in the coastal area of Bangladesh. In addition, risk quotient (RQ) calculated from the ratio of PAH levels in water to their corresponding quality values (QV) revealed that the aquatic ecosystem risk posed by the water-borne PAHs in the coastal area of Bangladesh is extremely high.

The concentrations of ∑PCBs were higher than the "effect range low" (ERL, 22.7 ng/g), the "interim sediment quality guideline" (ISQG, 21.5 ng/g) in about 35% and 22% of stations in winter and summer, respectively, indicating the potential adverse effects on benthic and bentho-pelagic organisms may occasionally occur. The ∑PCBs concentrations higher than ERL/ISQG were observed at sites located in the vicinity of Chittagong ship breaking (CT3 and CT4) and port area (CT1 and CT2), and Bakkhali estuary near a fishing harbor in Cox's Bazar (CX3). Furthermore, the ratio of the measured environmental concentration (MEC) to the predicted no-effect concentration (PNEC), termed the hazard quotient (HQ), was used to evaluate the ecological risks posed by PFOA and PFOS to the sediment-dwelling benthic and bentho-pelagic fish and other aquatic animals of the study area. An HQ value below 1 indicates no potential risk. In the present study, all the HQ values for sedimentary PFOA and PFOS were less than 1 (**Table 4-4**), indicating little or insignificant risks to the benthic and bentho-pelagic organisms. In addition, in most of the samples except ME2 and ME3 in winter and CX2 and ME3 in summer, the concentrations of the majority of the individual PAHs were above ERL but below ERM, which indicate that biological effects would occur occasionally. Moreover, some PAHs were in at least one sample that exceeded ERM in sites CT3 and SN2 in winter and CX3, CT1, CT2, CT3, CT4 and SN2 in summer (e.g., site CX3: Phe, site CT1: Ace and Pyr, site CT2: Pyr, site CT3: Pyr and Chr, site CT4: Pyr and site SN2: Ace and DahA), which indicates that biological effect would occur frequently at these sites. Overall, the water and sediment of the Bangladeshi coastal area are contaminated with these POPs with levels that could negatively affect the ecosystem integrity in this area. It is, therefore, suggested that intensive and long term survey of the environmental quality should be conducted to develop effective management strategies and that should be utilized effectively to control the contamination of POPs in Bangladesh, particularly in the coastal area.

7.1.6 Seafood contamination with POPs and human health risk implications

POPs in the Bangladeshi seafood deserve our attention because of not only their potential ecological impacts but also the public health concerns for seafood safety. In this study, 3 classes of the most concerned POPs (PCBs, PFAAs, and PAHs) were measured in 48 seafood samples (5 finfish and 2 shellfish species) that are most commonly consumed by the Bangladeshi coastal people. The samples were collected in winter and summer, 2015. The majority of the monitored POPs were detected frequently, suggesting the ubiquitous occurrence and continuous accumulation of these compounds in the Bangladeshi seafood. In general, irrespective to season and species, the total concentrations of PCBs, PFAAs, and PAHs were 3.82–86.18, 0.32–14.58, and 117.9–4216.8 ng/g ww, respectively. Comparison with the data available in literatures revealed that the present level of POPs in the Bangladeshi seafood were comparable or in some cases higher than that measured in seafood from other countries.

The concentrations of ∑PCBs and ∑PAHs in finfish were slightly higher in summer, whereas in shellfish these were higher in winter. Besides, an elevated levels of ∑PFAAs were measured in summer for both the finfish and shellfish, although these differences were not statistically significant in either cases ($p > 0.05$). These variations might be attributed to the seasonal discrepancies in their physiological activities, feeding behavior as well as the degree of contamination of their habitats. Besides, bioaccumulation patterns of contaminants in biota also depend both on uptake and elimination rates (Voorspoels et al., 2004). Significant interspecies differences were found in the seafood body burden of POPs. Within the monitored seafood species, Ilish among the finfish group and Crab among the shellfish group exhibited the highest levels of POPs in all cases and that was probably due to the higher lipid content of these species. Most of the POPs accumulate in adipose tissue and thus tend to increase with lipid content. Positive and strong correlations were depicted between the levels of POPs and the lipid contents in the studied seafood samples. However, Ilish is a migratory carnivorous species and that is thus habitated to different ecosystems (marine, estuarine, brackish and freshwater) and exposed to various degree of contamination. Conversely, Crab is a non-migratory typical benthic organisms residing above or in the sediment which is regarded as the final sink of POPs in the coastal environment. Therefore, Crab should be considered as a bio-indicator species to elucidate the impact of local emission sources to the level POPs contamination in the coastal area of Bangladesh. Another important factor that can play a key role in determining the contaminant burdens between the species is the depth at which organisms live. In general, higher concentrations of POPs have been detected in seafood from deeper waters (de Brito et al., 2002; Storelli et al., 2009). This agrees well with our results, both Ilish, Sole and Crab being organisms inhabiting deeper marine or coastal areas. Overall, the levels of the examined POPs were higher in finfish species than those measured in shellfish species which was in consistent with the trends reported in seafood from other coastal areas (Howell et al., 2008; Shen et al., 2009).

Health benefit and risk associated with seafood consumption in an ongoing debate globally. Seafood is a major dietary source of protein and healthy lipids. In particular, the long chain omega-3 fatty acids have beneficial roles in human health. Despite the benefit of a seafood diet, a concern of frequent seafood consumption is the potential risk from exposure to toxic chemicals including POPs (Smith and Gangolli, 2002; Storelli et al., 2003; Domingo et al., 2012). Since seafood contributes a major portion in the diet for the Bangladeshi coastal population, the consumption of contaminated seafood can be a potential risk for the consumers. Therefore, it is an urgent need to assess the potential health risk that might be posed from the dietary exposure of POPs through seafood consumption. To assess potential impacts on human health resulting from the consumption of contaminated seafood an exposure assessment based upon a defined tolerable daily intake (TDI) is needed. Dietary intake (e.g. estimated daily intake, EDI) is an imperative approach for the exposure assessment of POPs to human. The detailed results have been presented and discussed in the corresponding chapters of this thesis.

Briefly, [1] the estimated exposure to PCBs ($EDI_{\sum PCBs}$) were 42.06–49.46 and 48.62–61.82 ng/kg bw/day for the adults and children, respectively, and that were 2–3 times higher than the WHO proposed health based guidance value for total PCB (TDI = 20 ng/kg bw/day, WHO, 2003). Hence, the risk from dietary intake of PCBs through seafood consumption appears to be high for the Bangladeshi seafood consumers. [2] The EDI of PFOA and PFOS through seafood consumption were much lower than the EFSA recommended TDIs for PFOA and PFOS at 1500 ng/kg/day and 150 ng/kg/day, respectively (EFSA, 2008). Furthermore, assuming the toxicological concern for the other PFAAs (e.g. PFCAs: PFBA, PFPeA, PFHxA, PFHpA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, and PFTeDA; PFSAs: PFBS, PFHxS, and PFDS) similar to PFOA or PFOS, the EDIs of total PFAAs through seafood consumption by the Bangladeshi coastal residents (3.42–9.67 ng/kg bw/day for adults and 4.10–12.82 ng/kg bw/day for children) are still far below the EFSA recommended TDIs. Therefore, this study elucidated that the coastal residents of Bangladesh were not significantly exposed to PFAAs via the consumption of seafood. [3] The potency equivalent concentration (PEC) could be a better index for the potential toxicity related to PAHs exposure (Ding et al., 2013; Shi et al., 2016). In this study, the calculated PEC values for all of the studied seafood species (0.71–67.8 ng/g ww) were higher than the USEPA recommended PEC of total PAHs (0.67 ng/g ww, USEPA, 2000) for human consumption, indicating that consuming these PAH contaminated seafood will obviously induce adverse health effects on the Bangladeshi coastal residents. In addition, the incremental lifetime cancer risk (ILCR) values for children were higher than the maximum acceptable risk level, whereas the values for adults were higher than the priority risk level, indicating a potential carcinogenic risk to coastal residents in Bangladesh. Therefore, the potential health risk for the coastal people from exposure to the dietary POPs, particularly PCBs and PAHs through seafood consumption at the present levels should not be ignored.

7.2 Conclusions

This thesis examined the levels of PCBs, PFAAs, and PAHs in the surface waters, sediments and commonly consumed seafood in the coastal areas of Bangladesh. The samples were collected from four coastal regions (Cox's Bazar, Chittagong, Bhola, and Sundarbans) in winter and summer, 2015. A wide-scale monitoring of these pollutants were undertaken by multidimensional approaches to decipher the source factors and origins. Seasonal and spatial distributions of these pollutants were depicted in this study. A preliminary ecological and human health risk assessment was also carried out.

The compounds detected in the highest amounts were PAHs in each of the medium examined in this study. However, POPs levels were at the middle of reported global range. The water, sediment, and seafood (finfish and shellfish) were noted to be severely polluted where the levels of POPs were higher than the national and international environmental quality guidelines, suggesting potential threats to the aquatic organisms and human health as well through biomagnification. There was no specific seasonal pattern of POPs in the Bangladeshi coastal areas. Spatial distribution revealed that the Chittagong, Cox's Bazar and Sundarbans areas were more contaminated with POPs than the Meghna Estuary (Bhola) and because of greater development, thus associating these compounds to urbanization and industrialization. The results depicted that the ship breaking and port activities, open dumping and burning of waste (mostly consumer products including e-waste), discharges of untreated industrial and municipal effluents were the major source factors of environmental burden of POPs.

The profile data revealed that the prominent sources of PCBs in the Bangladeshi coastal areas were derived as related to PCB technical mixtures, pigments/dyes, and combustion. Selected molecular ratios demonstrated that the PAHs in the Bangladeshi coastal area originated from both the petrogenic and pyrogenic sources including crude petroleum (e.g. gasoline/diesel), petroleum combustion, and combustion of grass, wood and coal. In addition, a few dominant PCB congeners have been identified from this study that could serve as markers of PCB contamination in Bangladesh. While the monitoring of all PCBs is vital and thus recommended, such a set of indicator congeners may be useful for selective monitoring in case of reasonable constraints on full congener approach.

Besides, the dietary intakes of POPs through the analyzed seafood species for the adults and children of the Bangladeshi coastal area exceeded the health based guidelines, suggesting a potential health risk to the Bangladeshi seafood consumers. Therefore, the present public health risk issues should not be ignored. We should concentrate our views to solve this problem with integrated approaches on an urgent basis as well.

7.3 Recommendations

One of the highlights in this study with respect to the levels of POPs in the environmental media is the recurrence of relatively elevated levels of contaminations, particularly PCBs at Chittagong ship breaking area, irrespective to season. This area should be taken in to special consideration for the future research. An extensive monitoring of POPs is recommended in this area. In particular, human blood, urine or breast milk should be examined to figure out the scenario of POPs exposure for the workers in the ship breaking sectors as well as for the local residents.

Many contaminants of concern in sediments are legacy contaminants (e.g., PCBs) released to the environment decades ago, and they can be buried beneath layers of surface sediments. Further analysis of core sediments is thus recommended to elucidate the historical inputs of these contaminants in this area.

Cooking of seafood at high temperature may reduce the POPs levels. Some of the POPs might be transformed to toxic or non-toxic forms in cooking processes. Since Bangladeshi people consume cooked seafood, it is thus recommended to examine the effects of cooking on the levels of POPs and the resulting exposure scenario. In addition, dietary exposure of POPs through consumption of other non-piscine foodstuffs such as rice, vegetables, meat, milk, eggs, etc. should be evaluated in the further health risk assessments.

Only three classes of POPs – PCBs, PFAAs, and PAHs – were examined in this study. We recommend expanding this study by considering other POPs including OCPs, PCDD/Fs, PBDEs, PCNs, etc.

The present work in Bangladesh was the first attempt for the intensive monitoring of certain POPs in the coastal ecosystems. It will be important to expand such monitoring on POPs concentrations in the inland freshwater ecosystems of the country.

In recognition of the environmental problems of POPs contamination the study identified, a precautionary approach to mitigate the problems based on risk management is advanced through the following recommendations:

(I) Sensitization on the need to refrain from dumping and burning of industrial and municipal solid waste including e-waste in the coastal areas. Special attention should be paid to the ship breaking and port activities. Dismantling of old ships, shipping and fishing operations should be done in an effective way to ensure a minimize emission of POPs from this activities.

(II) Dredged sediments that are previously contaminated with POPs should be used after proper treatment by the effective methods that are capable of removing a reasonable amount of POPs.

(III) Promotion of knowledge dissemination and comprehensive mass awareness campaign on POPs and resulted human health effects.

(IV) Effective measures should be taken to promote introduction of new technologies/alternatives to minimize and eliminate POPs.

(V) A feasible technology should be adopted in Bangladesh to destroy the stockpiles of certain POPs, such as PCBs. Another option is to export them to other countries where destruction facilities exist. Also, strategies for remediation of the most seriously contaminated sites should be developed.

(VI) It is recommended to conduct more researches on the assessment of POPs with the aim of the developing strategy and introducing mechanisms to reduce and/or eliminate POPs discharges, spills and wastes.

(VII) Proper monitoring and improved management are crucial to handle the industrial, municipal and domestic waste disposal activities. For this reason, the enforcement of existing laws and regulations should be ensured. Additionally, it is suggested to formulate the capacity building approaches through training, awareness and research for the proper handling of the pollutants. In this case, developing countries like Bangladesh, should adopt ecofriendly and economically feasible technology from developed countries for the treatment of industrial and other waste to control the POPs pollution.

(VIII) Finally, the government should take effective measures in relation to the proper scientific assessment by the environmental scientists and researchers regarding the monitoring and the impact of human activities on POPs pollution in the Bangladeshi environments. Moreover, strong political commitment, contingency planning, proper policy formulation, inter-sectoral (Government-NGO-Corporate and Private sectors) partnership and above all public participation are necessary to overcome the existing challenges.

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Appendix

SUPPLEMENTARY INFORMATION

APPENDIX A

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 km^2 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 A-1. 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
	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 A-2. Local, English and Scientific names, biometric data, habitat and pretreatment of finfish and shellfish collected from the coastal area of Bangladesh.

Seafood	Local name	English name	Scientific name	Length (cm)	Weight (g)	Fat (%)	Habitat	Pretreatment[a]
Finfish	Ilish	Hilsa shad	*Tenualosa ilisha*	35.77±6.5	656.5±158.2	17.6±3.3	Marine; Brackish; Pelagic-Neritic; Anadromous	1,2,3,4
	Rupchanda	Silver pomfret	*Pampus argentius*	20.42±3.7	395.5±21.4	2.9±0.3	Marine; Benthopelagic; Oceanodromous	1,2,3,4,6
	Loitta	Bombay-duck	*Harpadon nehereus*	24.2±3.6	86.4±35.5	2.4±0.6	Marine; Brackish; Benthopelagic; Oceanodromous	1,2,3,4,6
	Sole	Long tongue sole	*Cynoglossus lingua*	25.6±5.3	90.7±3.7	3.2±0.2	Marine; Freshwater; Brackish; Demersal; Amphidromous	1,2,3,4,6
	Poa	Pama croaker	*Otolithoides pama*	22.11±3.3	185.9±10.5	3.0±0.5	Marine; Freshwater; Brackish; Benthopelagic; Amphidromous	1,2,3,6
Shellfish	Shrimp, Chingri	Indian shrimp	*Penaeus indicus*	15.4±2.3	34.6±3.2	2.1±0.4	Marine, Estuarine, Mud or Sandy Mud	1,5,7
	Crab, Kakrha	Mud crab	*Scylla serrata*	14.65±2.6	250.6±9.4	3.9±0.4	Bottom muddy feeder; Benthic, Mangrove zone, Estuarine	1,5,7

[a] 1: Wash; 2: scales removal; 3: viscera removal; 4: bone removal; 5: head/cephalothorax removal; 6: gills removal; 7: shell removal;

Table A-3. Instrumental characteristics and parameters used for the analytical determination of PCBs.

GC analysis conditions	
GC	Agilent 7890A GC
Column	HT8-PCB (60 m×0.25 mm I.D, Thickness 0.25 μm)
Oven Temperature	120 ℃ (1 min)−20 ℃/min−180 ℃ (0 min) −2 ℃/min−210 ℃ (0 min) −5 ℃/min−310 ℃ (3 min)
Inlet Temperature	280 ℃
Carrier gas	Helium, 1.0 mL/min (constant flow)
Injection	1 μL, Splitless
MS/MS analysis conditions	
MS/MS	7000C
Collision gas	Nitrogen, 1.5 mL/min
Quenching gas	Helium, 2.25 mL/min
Ionization method	EI 70 eV
Transfer line temperature	300 ℃
Ion source temperature	280 ℃
Quadrupole temperature	150 ℃
Tuning	Auto tune
Mode of operation	Multiple reactions monitoring (MRM), Gain 10

MRM conditions

Compounds	MS1 (m/z)	MS2 (m/z)	CE (eV)	Dwell Time (msec)
Mono-CB	190.0	155.0	30	10
Mono-CB	188.0	153.0	30	10
^{13}C Mono-CB	200.1	165.1	30	10
^{13}C Mono-CB	202.1	165.1	30	10
Di-CB	224.0	152.1	30	10
Di-CB	222.0	152.1	30	10
^{13}C Di-CB	234.1	164.1	30	10
^{13}C Di-CB	236.1	164.1	30	10
Tri-CB	256.0	186.0	30	10
Tri-CB	258.0	185.9	30	10
Tri-CB	260.0	188.0	30	10
^{13}C Tri-CB	268.0	198.0	30	10
^{13}C Tri-CB	270.0	200.0	30	10
Tetra-CB	289.9	219.7	30	10
Tetra-CB	291.9	222.0	30	10
Tetra-CB	293.9	222.0	30	10
^{13}C Tetra-CB	303.9	234.0	30	10
^{13}C Tetra-CB	305.9	236.0	30	10
Penta-CB	323.9	253.7	30	10
Penta-CB	325.9	255.9	30	10
Penta-CB	327.9	255.8	30	10
^{13}C Penta-CB	337.9	267.9	30	10
^{13}C Penta-CB	339.9	269.9	30	10
Hexa-CB	359.8	289.9	30	10
Hexa-CB	357.9	287.8	30	10
Hexa-CB	361.8	289.7	30	10
^{13}C Hexa-CB	371.8	301.9	30	10
^{13}C Hexa-CB	373.8	303.9	30	10
Hepta-CB	393.8	323.9	30	10
Hepta-CB	395.8	325.9	30	10
Hepta-CB	397.8	362.5	30	10
^{13}C Hepta-CB	405.8	335.9	30	10
^{13}C Hepta-CB	407.8	337.9	30	10

Octa-CB	429.8	360.0	30	10
Octa-CB	427.8	358.0	30	10
^{13}C Octa-CB	439.8	369.9	30	10
^{13}C Octa-CB	441.8	369.9	30	10
Nona-CB	463.7	393.8	40	10
Nona-CB	461.7	391.8	30	10
^{13}C Nona-CB	473.8	403.8	40	10
^{13}C Nona-CB	475.8	405.8	30	10
Deca-CB	499.7	429.7	40	10
Deca-CB	497.7	427.7	30	10
^{13}C Deca-CB	507.7	437.8	40	10
^{13}C Deca-CB	509.7	439.8	30	10

Table A-4. Recoveries*, LODs and linearity of calibration curves for the analytical determination of PCBs.

	% Recoveries [mean (RSD)]				LOD[a]			
PCBs	Water 10 ng/L (n=3)	Sediment 10 ng/g dw (n=3)	Seafood 10 ng/g ww (n=3)	PCB Homologs	Water (ng/L)	Sediment (ng/g dw)	Seafood (ng/g ww)	Linearity (r^2)[b]
PCB3	77 (13)	62 (9)	57 (15)	Mono-CB	0.03-0.07	0.002-0.003	0.05-0.06	0.9997
PCB8 (8+5)[c]	118 (7)	115 (6)	112 (8)	Di-CB	0.021-0.033	0.012-0.01	0.012-0.014	0.9996
PCB28	88 (4)	79 (4)	93(7)	Tri-CB	0.043-0.07	0.003-0.004	0.01-0.013	0.9998
PCB52	103 (6)	98 (5)	101 (2)	Tetra-CB	0.03-0.11	0.001-0.004	0.013-0.017	0.9989
PCB101	94 (4)	103 (11)	91 (4)	Penta-CB	0.053-0.132	0.003-0.005	0.01-0.011	0.9996
PCB118	84 (5)	92 (3)	113 (9)	Hexa-CB	0.022-0.061	0.004-0.01	0.006-0.008	0.9999
PCB138	116 (14)	94 (5)	101 (4)	Hepta-CB	0.04-0.13	0.003-0.006	0.03-0.04	0.9993
PCB153	107 (8)	108 (6)	104 (4)	Octa-CB	0.034-0.052	0.011-0.014	0.01-0.014	0.9992
PCB180	93 (5)	116 (12)	99 (5)	Nona-CB	0.04-0.07	0.012-0.01	0.013-0.016	0.9994
PCB194	83 (4)	89 (5)	110 (8)	Deca-CB	0.06	0.004	0.012	0.9991
PCB206	93 (2)	85 (6)	95 (2)					
PCB209	71 (7)	82 (5)	67 (7)					

[a] Limit of detection;

[b] Calibration curves (0.1–100 µg/L for each compound);

[c] PCB8 was identified and quantified as a co-elution of PCB8+5;

[*] The recovery of spiked PCBs was calculated using the following equation:

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

where $C_{\text{sample + spiked}}$ is the concentration of PCBs in a spiked sample, C_{sample} is the concentration of PCBs in the sample (same as above without spiking target compounds, C_{spiked} is the concentration of the spiked target PCBs.

Table A-5. Concentration of PCB congeners in the surface water samples (ng/L) in winter 2015.

PCB IUPAC No.[a]	Coastal sites													
	CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	0.20	0.37	0.22	0.24	0.44	<LOD	0.52	0.89	<LOD	0.27	0.20	0.86	0.35	0.40
4	<LOD	0.20	0.80	0.70	<LOD	0.54	<LOD	<LOD	0.68	<LOD	0.62	<LOD	0.78	0.76
9	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	0.50	<LOD	0.69	<LOD	0.92	0.24	0.27	0.61	0.34	<LOD	0.32	1.23	<LOD	<LOD
11	0.64	0.37	1.13	0.42	1.93	1.12	1.90	2.27	0.60	<LOD	<LOD	0.84	0.39	1.47
6	<LOD	0.27	0.20	0.58	0.90	<LOD	<LOD	0.87	<LOD	0.37	0.31	0.63	0.50	0.47
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	1.40	1.10	1.60	0.80	2.32	1.97	2.14	3.87	1.80	1.04	1.20	2.10	2.17	1.64
13+12	0.59	0.40	<LOD	0.44	1.10	1.14	0.76	<LOD	0.78	0.41	<LOD	<LOD	0.58	0.80
15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.86	<LOD	<LOD	0.20	<LOD	<LOD	0.20
19	0.60	<LOD	<LOD	0.26	<LOD	<LOD	<LOD	<LOD	0.28	0.26	0.46	<LOD	0.49	0.75
30	<LOD	0.26	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.06	<LOD	0.51
18	2.01	1.01	1.89	1.35	2.75	3.13	2.98	3.13	2.24	0.90	1.18	2.31	1.95	2.16
17	0.58	0.61	1.79	1.08	1.83	1.71	1.15	2.25	0.84	0.56	0.73	1.00	1.31	1.12
24	0.36	<LOD	0.80	0.42	1.36	1.11	0.44	1.07	0.50	0.38	0.17	0.41	0.27	0.86
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
32	0.64	0.28	0.54	0.45	1.56	0.86	0.57	1.16	0.38	0.45	0.55	1.13	0.21	0.73
16	1.90	0.67	1.80	1.31	<LOD	1.37	1.47	2.60	0.94	0.64	1.10	1.69	1.36	1.91
23	<LOD	<LOD	<LOD	<LOD	0.72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.21	<LOD	<LOD	<LOD	<LOD	<LOD
29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	0.57	<LOD	0.88	0.42	1.28	1.13	1.43	1.06	0.55	<LOD	0.18	0.64	0.31	0.90
25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
31	0.80	0.79	1.66	2.10	1.43	1.62	2.53	1.89	1.30	0.59	0.63	1.41	1.11	1.30
28	1.00	1.27	2.40	1.06	3.10	2.89	2.28	1.46	1.26	0.74	1.09	2.21	2.33	1.79
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	1.26	1.11	1.99	2.79	2.05	1.97	1.47	1.77	1.83	1.27	1.84	2.03	1.95	2.56
22	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
36	0.60	<LOD	1.17	0.95	1.37	1.07	0.46	0.81	<LOD	0.29	0.20	<LOD	0.21	0.25
39	<LOD	0.38	0.38	0.45	0.77	1.36	1.36	1.02	0.21	<LOD	<LOD	0.68	0.43	0.60
38	<LOD	<LOD	0.65	<LOD	0.61	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	0.73	<LOD	<LOD	0.71	<LOD	1.13	0.52	1.28	<LOD	0.36	0.32	0.59	0.29	0.21
37	0.31	0.49	0.96	1.03	1.07	1.06	1.48	0.66	0.66	0.36	0.12	1.43	0.58	0.86
54	0.71	<LOD	0.91	0.38	<LOD	<LOD	<LOD	<LOD	0.20	<LOD	0.24	0.38	0.24	0.27
50	<LOD	<LOD	<LOD	<LOD	0.76	<LOD	<LOD	<LOD	<LOD	0.22	0.20	<LOD	<LOD	0.20
53	0.26	<LOD	0.74	0.47	<LOD	0.45	0.90	0.95	0.23	<LOD	0.34	0.51	<LOD	0.32
51	<LOD	<LOD	<LOD	<LOD	<LOD	0.78	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
45	0.22	0.23	0.68	0.35	0.96	<LOD	1.12	0.54	0.21	0.21	0.21	0.62	0.78	0.27
52	1.27	0.50	1.71	1.73	2.45	2.46	4.73	2.55	0.31	0.24	0.29	3.64	0.43	1.35
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.10	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	2.15	1.36	2.93	2.40	2.53	2.03	3.62	3.91	2.10	1.32	1.11	2.70	2.18	2.42
65+75+47+48	0.50	0.86	2.34	1.70	1.93	1.69	3.13	4.08	0.67	0.98	0.39	2.10	1.27	1.18
62	<LOD	<LOD	<LOD	<LOD	0.56	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	<LOD	<LOD	<LOD	0.97	<LOD	<LOD	1.29	1.38	<LOD	<LOD	<LOD	1.39	<LOD	0.80
44	1.85	1.21	2.34	2.68	1.94	2.91	4.48	4.16	1.47	0.56	0.42	2.71	1.42	2.78
42	<LOD	<LOD	<LOD	<LOD	<LOD	1.56	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
64	1.36		0.82	2.29	1.53	1.96	2.34	3.46	1.17	0.23	0.20	1.96	0.33	0.36
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	0.98	0.61	1.45	0.25	<LOD	1.11	1.88	0.87	0.83	1.13	0.21	1.99	1.02	2.06
41	0.76		0.63	0.23	1.11	1.73	1.54	1.00	0.39	0.24	0.34	0.77	0.65	0.79
68	<LOD	<LOD	<LOD	<LOD	1.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
67+57	1.11	0.75	1.95	0.90	<LOD	<LOD	2.93	2.02	1.14	0.75	0.28	1.21	0.93	1.72
40	0.52	1.08	0.48	0.42	2.97	2.08	1.27	2.96	0.47	0.66	0.23	0.72	1.41	0.57
61+63+58	<LOD	<LOD	<LOD	<LOD	2.20	1.94	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

74	0.27	0.51	0.72	1.55	0.99	1.64	1.28	1.60	0.24	0.96	0.59	0.61	0.65	0.97
66	1.23	0.60	1.49	0.98	2.20	1.82	3.26	2.88	0.61	0.51	0.30	0.82	1.28	1.57
55+76+80	<LOD	<LOD	<LOD	<LOD	<LOD	1.22	<LOD	2.24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
70	1.60	1.15	2.76	1.83	3.89	3.68	3.58	3.45	1.76	0.87	0.82	2.20	1.64	2.37
60+56	0.18	0.69	1.89	1.31	0.94	<LOD	1.40	1.01	0.28	<LOD	0.30	1.35	1.18	0.67
81	0.20	0.36	0.62	0.86	0.93	0.78	0.95	0.87	0.21	0.21	0.21	0.21	0.46	0.22
77	0.20	0.48	1.37	0.80	0.95	0.67	0.94	1.13	0.23	1.29	0.27	0.24	0.36	0.23
79	<LOD	<LOD	0.75	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
78	0.24	<LOD	<LOD	0.21	<LOD	0.82	1.12	0.87	0.41	0.23	0.37	0.39	0.52	0.46
104	1.00	<LOD	0.28	0.33	0.28	0.20	0.34	0.70	<LOD	0.89	0.21	0.20	0.24	0.21
96	<LOD	0.35	<LOD	<LOD	0.34	0.87	1.48	1.34	0.24	<LOD	<LOD	0.25	<LOD	0.27
103	0.61	<LOD	0.68	0.24	0.27	<LOD	0.74	0.67	<LOD	0.35	0.23	0.23	0.22	<LOD
100	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
94	0.62	<LOD	0.25	<LOD	0.36	0.61	0.83	1.35	0.52	<LOD	<LOD	0.46	<LOD	0.20
102+93+98+95	<LOD	<LOD	2.58	1.20	1.13	2.44	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.43	<LOD
88	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.56	1.10	<LOD	<LOD	<LOD	0.72	<LOD	<LOD
91+121	0.48	0.45	0.26	1.14	0.89	1.03	1.79	3.19	0.65	0.20	0.21	1.92	0.28	0.44
92+84	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	1.23	0.52	0.82	0.86	<LOD	2.01	1.71	2.74	0.99	0.34	0.28	0.86	0.43	1.66
101	1.27	0.91	2.17	1.54	2.46	2.14	2.35	3.54	1.62	1.88	0.87	1.80	1.17	2.21
99	0.51	0.24	1.79	0.86	2.34	1.96	3.00	2.24	0.25	1.18	0.25	1.21	0.46	0.24
113	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.31	0.62	0.66	1.35	1.51	1.68	1.31	1.34	0.73	0.26	0.37	0.33	0.43	0.48
83+115	0.29	<LOD	0.79	<LOD	1.46	<LOD	<LOD	3.19	0.81	<LOD	<LOD	<LOD	<LOD	<LOD
86+117+97	<LOD	0.39	0.91	0.65	1.51	2.11	2.51	3.44	1.15	0.74	0.51	0.96	0.43	0.67
125+116+108	1.03	<LOD	1.72	<LOD	<LOD	<LOD	3.19	3.58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
87	0.41	0.23	1.12	1.61	1.79	1.03	1.95	2.69	0.66	0.55	0.28	0.21	0.59	0.50
111	0.96	0.27	0.23	<LOD	0.34	1.73	<LOD	<LOD	1.04	0.27	<LOD	0.30	<LOD	0.26
85	<LOD	<LOD	<LOD	<LOD	0.46	<LOD	<LOD	0.59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
120+110	1.62	1.20	2.11	1.79	2.82	2.20	3.13	3.83	1.62	0.75	0.99	2.54	2.30	1.92
82	0.25	<LOD	0.45	0.34	<LOD	0.89	0.59	1.81	1.34	1.26	<LOD	0.25	<LOD	<LOD
106+124	<LOD	<LOD	<LOD	<LOD	<LOD	0.57	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.76	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.79	0.53	0.39	0.87	0.39	0.95	1.43	1.83	1.17	0.84	0.26	0.79	0.23	0.21
118	0.99	1.72	1.72	0.99	1.99	1.96	3.34	2.84	0.38	0.87	0.47	1.34	1.86	1.57
114	0.26	0.21	0.34	0.54	0.24	0.65	0.80	1.42	0.38	0.23	0.34	0.21	0.42	0.22
122+127	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	0.87	1.23	2.03	1.17	0.87	1.86	0.73	2.54	0.76	1.15	0.32	0.42	1.19	0.55
126	<LOD	<LOD	0.23	<LOD	<LOD	<LOD	0.82	1.41	0.33	<LOD	<LOD	<LOD	<LOD	0.51
155	0.37	<LOD	<LOD	<LOD	0.44	0.28	0.96	0.98	0.24	<LOD	<LOD	<LOD	<LOD	<LOD
150	<LOD	<LOD	0.29	0.25	0.34	0.33	1.45	1.46	<LOD	0.28	0.21	0.21	0.24	0.21
152	<LOD	<LOD	0.34	<LOD	1.12	<LOD	0.94	0.59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
145	<LOD	<LOD	<LOD	<LOD	0.38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.27	<LOD	0.68	0.83	0.50	0.73	<LOD	1.24	0.30	<LOD	<LOD	<LOD	0.41	0.38
154	<LOD	<LOD	<LOD	<LOD	<LOD	0.68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
149+139	0.82	1.25	2.13	1.41	<LOD	2.41	3.82	2.87	0.68	1.29	<LOD	2.11	1.28	<LOD
135	<LOD	<LOD	0.35	<LOD	<LOD	<LOD	0.69	1.73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	0.40	0.31	0.25	0.36	1.88	0.47	1.06	0.94	0.26	0.24	0.20	0.45	0.32	0.25
140	<LOD	<LOD	<LOD	<LOD	0.55	0.31	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
143	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.47	0.47	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
134	<LOD	<LOD	0.32	<LOD	<LOD	0.70	<LOD	0.57	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
142+131+133	0.69	0.26	1.24	<LOD	2.72	0.78	0.96	0.91	0.53	<LOD	<LOD	1.32	<LOD	1.53
165	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	<LOD	<LOD	0.24	<LOD	<LOD	0.36	0.26	0.61	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
132+161	<LOD	<LOD	<LOD	<LOD	2.82	1.98	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
137	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
168	0.78	0.22	0.91	0.25	0.41	0.86	0.61	0.38	0.28	0.21	0.23	0.20	0.25	0.34
141	<LOD	<LOD	<LOD	<LOD	<LOD	0.46	0.52	0.34	<LOD	<LOD	<LOD	0.24	<LOD	<LOD
153	1.67	0.91	1.52	1.04	2.60	2.90	3.00	2.02	0.51	0.61	0.76	1.85	1.19	1.16
130	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
158	0.37	0.24	0.26	0.36	0.26	0.36	0.45	0.46	0.35	0.33	0.33	0.29	0.21	0.29
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	0.46	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
138	0.61	1.03	1.95	1.04	3.63	2.93	2.56	2.31	0.56	1.34	1.00	1.34	1.34	1.67

169	<LOD	<LOD	<LOD	<LOD	0.22	<LOD	<LOD	0.43	<LOD	<LOD	<LOD	0.26	<LOD	<LOD
157	0.29	0.28	0.22	0.22	0.33	0.93	0.91	0.39	0.44	0.27	0.24	0.45	0.31	0.25
128	1.56	0.66	1.76	0.51	0.64	1.49	1.78	1.61	0.50	1.05	0.76	0.47	0.17	1.71
159+162	0.42	<LOD	1.28	<LOD	<LOD	<LOD	0.27	0.90	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
167	0.69	0.83	0.28	0.23	0.43	0.56	0.34	0.64	0.39	0.20	0.22	0.23	0.22	0.20
156	0.40	0.30	0.29	0.47	0.21	0.71	0.29	1.45	0.23	0.29	0.21	0.23	0.28	0.26
166	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
129	<LOD	<LOD	<LOD	<LOD	<LOD	0.40	0.39	0.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
188	0.48	<LOD	<LOD	<LOD	0.46	0.47	0.37	0.29	0.23	<LOD	<LOD	<LOD	<LOD	<LOD
184	<LOD	0.21	0.42	0.65	0.56	0.50	<LOD	0.27	<LOD	0.21	0.26	0.40	<LOD	<LOD
179	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
176	<LOD	<LOD	<LOD	<LOD	0.33	<LOD	<LOD	0.43	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
187	0.74	0.37	1.74	0.30	0.37	0.70	1.23	0.89	0.25	0.81	0.21	1.39	0.46	0.51
178	0.32	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.43	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
175	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
182+186	1.13	1.18	0.64	0.61	<LOD	1.21	1.16	1.02	0.69	<LOD	<LOD	0.24	<LOD	0.60
183	0.34	0.52	0.31	0.53	0.35	1.01	0.52	0.57	0.30	0.51	0.20	0.53	0.49	0.20
174	1.21	0.22	1.31	0.70	1.40	1.25	2.23	2.29	0.55	0.87	0.22	0.24	1.20	0.63
185	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
181	<LOD	<LOD	<LOD	<LOD	0.40	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
171	0.66	<LOD	<LOD	<LOD	<LOD	0.67	0.77	<LOD	0.93	1.15	0.78	1.63	1.23	1.11
177	0.30	0.38	1.13	0.61	0.76	1.36	1.61	0.21	0.23	0.55	0.25	0.71	0.42	0.31
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.39	0.27	0.22	0.24	0.35	0.52	0.69	0.16	0.27	0.25	0.26	0.50	0.23	0.20
180	0.95	0.95	1.97	0.98	1.58	1.41	1.82	2.27	1.12	1.07	0.20	1.23	0.94	0.98
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	0.65	0.79	0.72	1.21	1.77	1.36	2.16	1.98	0.94	0.22	0.35	1.09	0.77	1.12
191	<LOD	<LOD	<LOD	<LOD	<LOD	0.78	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
193	<LOD	0.29	0.27	<LOD	<LOD	<LOD	<LOD	0.14	<LOD	<LOD	<LOD	0.50	<LOD	<LOD
190	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
172	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.69	0.54	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	0.29	0.33	0.44	0.71	1.15	1.42	0.45	0.38	0.47	0.30	0.22	0.69	0.35	0.25
195	0.84	0.92	0.72	0.26	0.67	0.89	0.75	0.65	0.23	0.37	0.31	0.26	0.21	0.22
200	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
198+199	<LOD	<LOD	0.25	0.77	0.29	0.66	0.23	0.47	<LOD	0.25	0.37	<LOD	<LOD	0.48
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	0.48	<LOD	1.58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
194	1.17	<LOD	1.06	<LOD	<LOD	<LOD	0.85	0.59	0.98	<LOD	<LOD	0.65	1.14	0.91
205	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑PCBs[b]	62.62	41.55	95.16	69.87	108.8	119.4	137.7	160.7	55.37	43.93	32.17	84.57	58.63	71.72

[a] The order of PCB congeners are based on the order of chromatographic separation on an HT8-PCB column;

[b] The concentrations of congeners with <LOD were assumed to be zero.

Table A-6. Concentration of PCB congeners in the surface water samples (ng/L) in summer 2015.

PCB IUPAC No.	Coastal sites													
	CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	0.32	0.23	0.34	<LOD	<LOD	0.22	0.34	<LOD	0.26	0.29	<LOD	0.29	0.30	0.38
4	0.76	<LOD	1.79	0.68	1.71	1.10	0.47	0.46	0.80	0.59	<LOD	0.62	<LOD	1.85
9	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	<LOD	0.50	0.44	<LOD	0.58	0.64	0.60	0.71	0.67	<LOD	0.26	<LOD	<LOD	0.94
11	0.51	0.36	2.04	0.39	2.58	1.60	1.42	2.94	0.27	0.23	0.25	0.40	0.62	1.41
6	0.66	0.49	0.26	0.24	0.65	0.60	0.52	0.56	<LOD	<LOD	<LOD	<LOD	<LOD	0.65
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	2.12	1.19	3.13	2.10	3.59	3.14	4.15	5.23	1.43	1.34	1.22	2.97	2.16	2.36
13+12	0.79	0.55	<LOD	<LOD	1.52	0.77	1.04	1.09	0.72	<LOD	0.44	<LOD	0.42	0.81
15	<LOD	<LOD	0.56	1.14	1.80	<LOD	<LOD	<LOD	<LOD	0.75	<LOD	1.41	0.63	1.13
19	<LOD	0.21	0.23	<LOD	0.88	0.73	<LOD	0.91	0.23	0.81	0.49	0.39	0.26	1.15
30	0.23	<LOD	<LOD	0.21	0.58	0.38	0.72	0.69	0.38	0.52	0.36	0.62	0.32	1.01
18	2.02	0.96	3.68	3.49	5.16	3.35	5.05	5.71	2.69	2.25	2.94	2.26	2.86	3.17
17	0.86	0.50	1.17	1.35	3.64	3.50	3.36	2.41	1.97	1.22	1.36	1.61	1.59	2.45
24	0.31	<LOD	0.51	0.23	0.96	<LOD	0.74	<LOD	<LOD	<LOD	<LOD	1.29	0.20	1.17
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
32	0.28	0.31	0.65	0.30	<LOD	0.94	1.26	<LOD	0.45	<LOD	0.68	0.45	0.32	<LOD
16	0.33	0.45	1.35	1.25	3.34	2.27	4.13	1.98	0.96	1.57	0.55	1.29	1.26	1.50
23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.60	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	0.46	0.23	0.50	0.41	<LOD	0.49	1.29	0.99	0.33	<LOD	0.59	0.53	0.44	1.97
25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
31	1.36	0.47	2.12	1.26	2.84	2.67	3.32	2.12	1.43	0.76	0.97	2.44	1.59	1.24
28	1.25	1.80	2.07	2.07	4.34	1.71	4.23	4.89	2.63	2.10	1.68	2.91	2.49	1.80
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	2.14	1.26	3.49	2.71	3.71	2.38	3.01	3.72	0.81	2.76	1.14	2.72	2.12	2.91
22	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
36	0.98	0.35	0.94	0.38	<LOD	<LOD	0.84	<LOD	<LOD	0.90	0.25	0.39	0.56	0.39
39	1.23	0.25	0.69	0.46	0.56	0.91	0.99	0.63	0.31	<LOD	<LOD	<LOD	0.98	0.40
38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	0.19	<LOD	0.65	0.47	<LOD	<LOD	0.77	0.84	<LOD	0.51	0.32	0.23	0.31	0.46
37	0.51	0.62	1.29	0.89	2.48	1.62	1.38	2.77	1.08	1.04	0.42	0.58	0.74	1.62
54	0.20	<LOD	0.25	0.20	0.45	0.50	0.50	<LOD	0.21	0.22	0.34	0.29	0.27	0.21
50	<LOD	0.20	<LOD	0.29	<LOD	<LOD	0.66	0.83	0.22	<LOD	0.50	0.63	<LOD	0.29
53	0.21	0.23	0.48	0.31	0.87	0.51	0.55	0.43	0.28	0.30	<LOD	0.46	0.36	0.37
51	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
45	0.25		0.75	0.22	0.36	0.81	0.35	0.66	0.44	0.23	<LOD	0.34	0.21	0.38
52	1.27	0.30	3.35	3.06	2.66	1.75	4.13	6.23	<LOD	2.11	1.29	1.29	1.74	3.19
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	2.33	1.51	3.60	2.19	5.20	4.52	3.05	5.50	3.47	1.85	1.63	2.79	2.42	2.96
65+75+47+48	1.41	0.93	1.80	0.84	1.36	1.15	0.92	1.70	0.48	0.66	0.68	0.57	0.56	1.38
62	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.64	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	0.22	0.25	0.38	<LOD	0.42	<LOD	0.58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
44	2.05	0.51	3.21	0.24	3.55	3.96	3.77	5.62	2.46	1.93	2.52	2.32	1.37	1.65
42	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
64	0.46	<LOD	0.50	0.31	0.35	0.38	1.28	4.14	0.27	0.59	<LOD	<LOD	0.24	0.94
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	1.45	0.90	0.24	1.84	1.47	1.85	0.72	2.16	0.71	0.83	0.69	1.70	0.68	1.46
41	1.15	0.72	0.21	2.40	1.19	2.84	1.28	1.28	0.29	<LOD	1.22	<LOD	0.37	1.63
68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
67+57	0.82	0.87	1.73	1.45	1.21	1.89	1.81	1.63	0.66	0.48	0.39	0.92	0.67	1.12
40	0.41	0.22	0.72	0.33	1.42	2.75	0.54	1.24	1.30	0.42	0.26	0.52	0.86	0.36
61+63+58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

74	1.03	0.49	1.15	1.73	1.39	1.77	1.06	2.81	0.61	0.26	0.45	1.26	0.79	3.25
66	0.51	0.22	2.88	0.79	1.63	2.24	2.93	5.30	2.08	0.64	1.65	1.54	1.94	2.50
55+76+80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
70	1.85	1.85	3.56	3.42	5.14	3.74	5.95	5.44	3.00	1.91	2.15	2.35	2.90	2.54
60+56	0.63	1.28	2.02	0.42	1.09	0.72	1.48	<LOD	0.57	0.61	<LOD	0.60	0.59	0.53
81	1.80	1.05	1.43	0.55	2.46	1.86	3.11	1.54	0.73	0.54	0.97	0.71	2.16	2.28
77	0.33	1.41	0.88	0.96	1.42	0.84	2.18	1.50	1.15	0.37	0.54	0.66	0.75	1.32
79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.32	<LOD	<LOD	<LOD	<LOD	<LOD
78	0.21	0.26	1.04	0.26	1.70	1.37	1.14	0.49	0.42	0.27	0.20	0.32	0.44	0.23
104	0.24	0.22	0.57	<LOD	0.67	0.20	0.34	<LOD	0.22	<LOD	<LOD	0.20	0.22	<LOD
96	0.22	<LOD	<LOD	0.20	<LOD	<LOD	<LOD	0.44	<LOD	0.22	0.21	0.22	0.27	<LOD
103	<LOD	0.24	0.34	0.39	0.47	0.21	0.67	<LOD	0.23	<LOD	0.22	0.30	<LOD	0.22
100	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
94	<LOD	0.39	0.71	<LOD	0.38	0.41	0.58	0.58	0.30	<LOD	<LOD	<LOD	<LOD	0.31
102+93+98+95	<LOD	0.93	2.23	0.93	3.31	0.98	4.08	5.77	0.70	1.13	0.43	0.93	0.85	2.92
88	<LOD	<LOD	<LOD	<LOD	<LOD	0.33	0.41	0.81	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
91+121	0.92	1.08	2.00	0.96	1.93	1.17	0.55	1.35	0.27	0.27	<LOD	0.86	<LOD	1.16
92+84	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	0.87	0.84	1.40	1.22	1.76	1.43	1.36	5.32	0.28	<LOD	<LOD	0.92	0.76	0.87
101	2.24	1.26	3.10	2.69	4.06	2.22	5.36	6.17	1.53	0.93	1.50	2.75	1.88	2.00
99	1.16	0.21	1.94	0.76	0.39	0.95	1.08	3.15	0.48	0.78	0.51	0.50	0.56	2.00
113	<LOD	<LOD	0.79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.73	0.61	2.31	1.24	0.89	0.80	1.93	1.56	0.60	0.25	0.30	0.52	0.61	1.39
83+115	<LOD	0.38	<LOD	<LOD	0.74	<LOD	1.32	<LOD	<LOD	<LOD	<LOD	0.33	<LOD	<LOD
86+117+97	0.84	0.46	2.56	1.14	0.78	0.78	1.62	1.21	0.37	0.40	0.29	0.76	0.37	0.23
125+116+108	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.11	1.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
87	0.96	<LOD	2.30	0.78	1.41	1.43	1.64	0.89	0.28	0.26	0.52	<LOD	0.43	1.67
111	0.29	0.46	1.79	<LOD	0.56	0.81	3.45	0.53	1.30	0.60	<LOD	0.80	<LOD	1.42
85	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
120+110	1.84	1.17	3.59	3.30	4.95	2.62	6.13	6.78	2.22	1.36	2.36	2.91	1.85	2.93
82	0.29	0.92	0.51	<LOD	0.38	0.89	0.83	1.07	<LOD	<LOD	<LOD	1.36	<LOD	1.44
106+124	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.22	0.27	0.32	0.88	1.16	0.72	1.16	1.17	0.47	0.23	0.52	0.82	0.29	0.24
118	1.24	0.89	2.83	2.46	2.79	2.05	4.68	4.92	1.54	1.12	1.04	2.68	2.12	2.14
114	0.37	0.23	0.97	0.34	0.83	0.97	3.19	3.31	0.22	0.53	0.70	0.27	1.17	0.34
122+127	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	0.75	0.70	2.11	2.06	2.16	1.64	2.53	6.02	0.32	1.03	0.37	1.59	1.43	1.16
126	0.31	<LOD	0.53	<LOD	0.33	<LOD	0.81	2.37	0.35	<LOD	<LOD	0.46	<LOD	<LOD
155	<LOD	<LOD	<LOD	<LOD	0.22	<LOD	0.29	<LOD	0.22	<LOD	<LOD	<LOD	<LOD	0.22
150	0.20	0.23	0.22	0.21	0.31	0.22	0.20	0.46	0.42	0.21	0.21	0.30	0.22	<LOD
152	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.52	<LOD	<LOD	<LOD	0.23	<LOD	0.26
145	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.88	0.64	0.51	0.88	0.76	0.42	0.91	0.86	<LOD	<LOD	<LOD	0.76	<LOD	<LOD
154	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
149+139	0.90	0.56	2.53	1.78	<LOD	1.67	2.77	3.84	0.96	<LOD	0.80	2.72	1.51	1.12
135	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.41	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	0.21	0.22	0.81	1.11	1.38	0.85	1.36	2.35	0.99	0.48	0.24	0.34	0.27	0.23
140	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
143	<LOD	<LOD	<LOD	<LOD	0.22	<LOD	<LOD	0.61	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
134	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
142+131+133	<LOD	<LOD	0.87	<LOD	0.54	0.41	0.79	1.73	0.68	<LOD	<LOD	0.41	0.88	0.86
165	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	<LOD	<LOD	<LOD	<LOD	0.25	0.22	<LOD	0.60	<LOD	<LOD	<LOD	0.34	<LOD	<LOD
132+161	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
137	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
168	0.22	0.26	0.29	0.50	0.80	0.62	1.34	1.29	0.39	0.54	0.32	0.24	0.23	0.24
141	<LOD	<LOD	<LOD	<LOD	<LOD	0.42	0.82	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.18
153	1.16	0.78	1.85	1.88	3.19	2.23	3.29	4.52	1.45	1.30	0.76	2.38	1.35	1.73
130	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
158	0.66	0.33	0.22	0.56	0.30	0.65	1.41	1.10	0.58	0.66	0.32	0.57	0.24	0.30
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
138	1.37	1.03	2.56	1.99	2.83	2.66	2.99	3.88	1.60	1.49	1.79	3.03	1.93	2.54

169	0.44	<LOD	<LOD	0.35	<LOD	0.46	0.35	0.68	0.34	0.27	<LOD	0.40	<LOD	0.71
157	0.31	0.23	0.26	0.88	0.20	0.35	1.04	0.85	0.32	0.22	0.30	0.26	0.27	0.31
128	1.13	0.99	1.00	0.70	1.32	0.90	1.89	2.59	0.71	1.30	0.85	0.67	1.15	1.76
159+162	<LOD	<LOD	0.47	<LOD	0.49	0.20	0.63	0.86	<LOD	<LOD	<LOD	<LOD	<LOD	0.70
167	0.73	0.24	0.23	0.39	0.69	0.42	0.72	1.11	0.43	0.27	0.27	0.28	0.43	0.77
156	0.54	0.26	0.22	0.37	0.67	0.35	1.58	1.20	0.38	0.33	0.29	0.35	0.51	0.87
166	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
129	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.61	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.22
188	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.28	<LOD	<LOD
184	0.24	0.20	0.20	<LOD	0.20	0.20	0.21	0.38	0.21	<LOD	<LOD	<LOD	0.22	0.23
179	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
176	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.53	<LOD	<LOD	<LOD	0.24	<LOD	<LOD
187	0.72	0.35	0.28	0.25	0.48	0.71	0.48	0.78	0.45	0.24	0.20	0.44	0.29	0.99
178	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
175	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.40	0.25	<LOD	<LOD	<LOD	<LOD	0.26
182+186	1.28	0.67	0.95	0.84	0.74	0.76	0.66	1.41	<LOD	0.44	<LOD	0.73	0.59	0.96
183	0.22	0.28	0.29	0.52	0.58	0.84	0.23	0.57	0.46	0.55	0.49	0.86	0.42	0.74
174	0.76	0.22	0.32	1.13	0.73	1.43	2.81	1.64	0.31	0.39	0.82	1.85	0.66	1.81
185	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.28
181	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.35	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
171	1.21	0.91	2.29	1.56	3.58	1.91	3.16	2.52	1.83	1.34	0.95	2.17	0.98	2.42
177	0.41	0.23	0.65	1.29	1.59	0.37	1.80	2.19	0.48	0.63	0.40	0.41	0.29	1.36
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.26	0.32	0.26	0.44	0.22	0.58	0.18	1.13	0.24	0.58	0.28	0.23	0.42	0.36
180	0.77	0.79	2.01	1.04	1.45	2.00	4.99	5.15	1.39	0.95	0.73	2.57	1.37	1.89
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.32	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	0.82	0.61	2.97	0.93	1.40	1.79	3.69	3.93	1.18	1.19	1.08	2.05	1.19	1.88
191	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
193	<LOD	<LOD	<LOD	<LOD	0.29	0.75	0.23	0.58	<LOD	<LOD	<LOD	<LOD	0.24	0.36
190	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
172	0.27	<LOD	0.38	<LOD	0.37	<LOD	0.29	<LOD	<LOD	<LOD	<LOD	<LOD	0.23	0.28
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	0.25	0.24	0.47	0.20	0.20	0.23	0.68	0.71	0.58	0.33	0.30	0.24	0.21	0.22
195	<LOD	0.28	0.37	0.29	0.36	0.69	0.29	0.77	0.32	<LOD	<LOD	0.81	<LOD	0.58
200	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
198+199	<LOD	<LOD	0.49	<LOD	0.79	<LOD	0.48	1.88	0.24	<LOD	<LOD	<LOD	0.32	1.27
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
194	0.88	0.39	0.78	0.55	0.84	<LOD	1.76	2.92	0.65	0.58	<LOD	1.11	0.38	<LOD
205	<LOD	<LOD	0.23	<LOD	<LOD	<LOD	0.91	0.61	<LOD	<LOD	<LOD	<LOD	<LOD	0.78
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑PCBs	66.26	46.45	116.0	81.80	134.2	110.4	172.8	199.4	66.88	55.24	49.77	89.10	69.12	112.4

Table A-7. Concentration of PCB congeners in the surface sediment samples (ng/g dw) in winter 2015.

PCB IUPAC No.	Coastal sites													
	CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	0.03	<LOD	0.04	<LOD	0.06	0.03	<LOD	0.12	0.10	0.02	0.04	<LOD	0.02	0.04
4	<LOD	<LOD	<LOD	0.30	<LOD	0.14	0.16	<LOD	0.09	<LOD	<LOD	0.10	<LOD	0.15
9	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	<LOD	<LOD	0.04	<LOD	0.05	<LOD	0.70	0.33	<LOD	0.02	0.02	0.05	0.02	<LOD
11	0.04	<LOD	0.40	0.45	<LOD	0.10	0.28	0.72	<LOD	<LOD	<LOD	<LOD	<LOD	0.10
6	0.06	0.10	<LOD	<LOD	0.13	0.06	0.16	0.23	0.10	0.03	0.02	0.08	0.03	0.02
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	0.12	0.13	0.74	0.62	0.13	0.29	1.15	1.49	0.17	0.06	0.11	0.21	0.11	0.31
13+12	<LOD	<LOD	<LOD	<LOD	0.17	0.13	0.40	0.93	0.11	0.05	<LOD	0.09	<LOD	0.06
15	0.07	0.03	0.44	0.19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.05	<LOD
19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD
30	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	0.04	<LOD	0.07	<LOD	<LOD
18	0.40	0.48	0.89	0.93	0.54	0.59	1.87	1.95	0.20	0.18	0.21	0.55	0.21	0.50
17	<LOD	<LOD	0.69	0.66	0.16	0.31	1.61	0.64	<LOD	<LOD	0.07	0.50	0.11	0.42
24	<LOD	0.14	<LOD	0.07	0.18	0.10	0.49	0.12	0.09	0.06	0.07	<LOD	<LOD	0.07
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
32	0.17	0.16	<LOD	<LOD	0.22	0.20	0.51	0.15	0.14	0.07	0.05	0.10	<LOD	0.12
16	<LOD	0.23	0.19	0.08	0.41	<LOD	<LOD	1.83	0.19	0.03	0.17	0.14	0.10	0.22
23	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	0.16	<LOD	<LOD	<LOD	<LOD	0.45	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	0.16	0.17	<LOD	<LOD	0.21	0.19	0.63	0.08	0.06	0.08	0.06	0.06	<LOD	0.06
25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
31	0.33	0.30	0.31	0.61	0.30	0.43	1.02	1.94	0.16	0.07	<LOD	0.44	0.10	0.24
28	0.44	0.39	0.81	0.65	0.20	0.44	1.55	1.83	0.24	0.15	0.10	0.69	0.20	0.41
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	0.22	0.26	0.94	0.46	0.30	0.62	1.39	2.19	0.23	0.11	0.10	0.62	0.16	0.18
22	<LOD	<LOD	0.10	<LOD	0.32	0.09	<LOD	<LOD	<LOD	<LOD	0.11	<LOD	<LOD	0.04
36	0.21	<LOD	0.13	0.06	<LOD	0.15	0.08	0.42	0.09	0.12	0.13	<LOD	0.10	0.04
39	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	0.52	0.10	<LOD	0.10	<LOD	<LOD	<LOD	<LOD
38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	<LOD	0.15	<LOD	0.05	0.16	0.07	0.51	0.16	<LOD	0.05	0.02	<LOD	0.14	0.05
37	0.09	0.18	0.29	0.12	0.18	0.18	0.68	1.23	0.11	0.12	0.06	0.55	0.09	0.18
54	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
50	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
53	0.08	<LOD	<LOD	<LOD	0.07	0.04	0.20	0.35	<LOD	<LOD	<LOD	0.05	<LOD	0.06
51	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
45	0.07	<LOD	<LOD	0.23	0.14	0.05	0.13	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	0.08
52	0.37	0.43	0.61	0.62	0.84	0.24	2.02	2.81	0.11	0.11	0.09	0.11	0.16	0.20
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	0.31	0.32	0.82	0.50	0.77	0.41	2.61	2.88	<LOD	0.16	0.08	1.21	0.23	0.44
65+75+47+48	<LOD	0.04	<LOD	<LOD	0.26	<LOD	<LOD	1.08	0.19	0.02	<LOD	0.46	<LOD	0.07
62	<LOD	0.03	<LOD	<LOD	0.54	<LOD	0.19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	<LOD	<LOD	<LOD	<LOD	<LOD	0.29	<LOD	0.13	0.17	<LOD	0.08	<LOD	<LOD	0.09
44	0.39	0.53	0.80	0.53	0.90	0.49	2.04	0.82	0.32	0.15	0.11	0.81	0.22	0.39
42	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
64	0.33	0.15	0.02	0.39	0.88	0.27	0.36	0.59	0.12	0.16	0.06	<LOD	0.18	0.16
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	0.27	<LOD	0.22	<LOD	<LOD	0.18	<LOD	<LOD	<LOD	<LOD	<LOD	0.11	0.03	0.09
41	<LOD	0.08	<LOD	0.05	0.84	<LOD	0.52	0.58	0.18	0.13	0.10	0.34	0.06	0.08
68	0.09	<LOD	<LOD	<LOD	<LOD	<LOD	0.21	0.12	0.14	<LOD	<LOD	<LOD	0.04	<LOD
67+57	<LOD	<LOD	0.06	0.17	<LOD	<LOD	<LOD	1.21	<LOD	<LOD	0.04	<LOD	<LOD	<LOD
40	0.38	0.06	0.16	0.09	1.13	0.34	2.09	0.59	0.23	0.23	0.13	0.14	0.12	0.17
61+63+58	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	1.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

74	0.15	0.20	0.27	0.40	0.40	0.14	<LOD	2.41	0.16	0.03	0.05	0.30	0.15	0.03
66	0.22	0.41	0.62	0.51	0.82	0.36	1.93	2.50	0.22	0.26	0.14	0.44	0.15	0.27
55+76+80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.13
70	0.47	0.46	0.73	0.67	1.25	0.35	2.29	2.13	0.26	0.24	0.18	0.51	0.25	0.47
60+56	0.21	0.27	0.26	<LOD	0.75	0.20	0.25	0.97	<LOD	<LOD	0.04	<LOD	0.03	<LOD
81	0.09	0.07	0.09	0.10	<LOD	0.05	0.81	0.95	0.15	0.01	0.06	0.19	0.03	0.20
77	0.09	<LOD	0.12	0.10	0.30	0.07	0.88	0.47	0.16	0.04	0.04	0.13	<LOD	0.16
79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
78	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	1.12	0.17	<LOD	<LOD	<LOD	<LOD	0.02	<LOD
104	<LOD	<LOD	0.04	0.04	<LOD	0.03	0.08	0.25	0.05	0.05	0.05	0.05	0.16	0.08
96	<LOD	<LOD	0.06	0.13	0.48	0.05	0.49	0.41	0.05	0.05	0.05	0.13	0.07	0.07
103	<LOD	<LOD	0.14	0.08	0.15	0.07	0.33	0.22	0.08	<LOD	<LOD	<LOD	<LOD	0.14
100	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
94	0.27	<LOD	<LOD	0.11	0.36	0.10	0.60	<LOD	0.02	0.06	<LOD	<LOD	<LOD	0.20
102+93+98+95	0.35	0.34	1.16	0.26	0.54	0.68	<LOD	3.15	0.19	0.18	0.04	0.64	0.11	0.26
88	0.03	0.03	<LOD	<LOD	0.35	0.17	0.28	0.19	<LOD	0.05	<LOD	<LOD	<LOD	0.20
91+121	<LOD	<LOD	<LOD	0.23	0.86	0.21	0.83	0.98	0.13	0.12	<LOD	<LOD	<LOD	0.22
92+84	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	0.03	<LOD	0.12	0.21	0.11	0.47	<LOD	1.77	<LOD	<LOD	<LOD	0.16	<LOD	0.12
101	0.51	0.49	0.58	0.47	0.88	0.61	2.44	2.25	0.30	0.25	0.09	0.41	0.28	0.36
99	0.32	0.33	0.25	0.37	0.69	0.27	1.52	1.41	0.12	0.14	0.05	0.18	0.11	0.19
113	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.35	0.33	0.23	<LOD	<LOD	<LOD	0.83	0.56	0.13	0.21	0.07	0.18	<LOD	0.21
83+115	0.42	0.19	0.12	<LOD	0.75	0.31	0.52	0.42	0.19	<LOD	<LOD	<LOD	<LOD	<LOD
86+117+97	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.48	0.82	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
125+116+108	0.37	<LOD	<LOD	<LOD	0.16	0.19	<LOD	0.78	<LOD	<LOD	<LOD	<LOD	<LOD	0.33
87	0.36	0.38	0.06	0.15	0.68	0.32	0.93	1.13	0.12	0.11	0.02	0.18	0.18	0.38
111	0.10	<LOD	0.38	0.11	<LOD	0.09	<LOD	1.28	<LOD	0.02	<LOD	0.25	0.12	0.30
85	0.19	<LOD	0.33	0.16	0.67	0.22	0.27	<LOD	<LOD	0.09	<LOD	<LOD	0.07	0.02
120+110	0.79	0.56	1.00	0.52	1.13	0.91	3.17	3.31	0.27	0.21	0.10	0.92	0.31	0.45
82	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.55	0.68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
106+124	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.21	0.16	0.07	0.17	0.23	0.08	0.26	0.59	0.02	0.06	0.17	0.11	0.07	0.07
118	0.62	0.41	1.19	0.43	0.92	0.89	2.05	2.16	0.19	0.10	0.10	0.96	0.24	0.19
114	0.12	0.25	0.47	0.20	0.22	0.06	0.47	1.05	0.07	0.04	0.06	0.13	0.07	0.06
122+127	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	0.34	0.09	0.20	0.27	0.41	0.67	2.61	2.55	0.09	0.07	0.04	0.48	0.20	0.19
126	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02
155	<LOD	0.20	0.06	0.06	0.25	0.05	0.09	0.05	<LOD	0.05	0.03	<LOD	0.04	0.03
150	<LOD	<LOD	0.05	0.11	0.53	0.05	0.22	0.07	0.05	0.05	0.02	<LOD	0.12	0.04
152	<LOD	<LOD	0.06	<LOD	0.21	0.07	0.35	0.64	0.06	<LOD	<LOD	<LOD	<LOD	<LOD
145	<LOD	<LOD	0.05	<LOD	0.10	0.07	0.21	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.19	<LOD	<LOD	<LOD	0.38	<LOD	<LOD	<LOD	0.10	<LOD	<LOD	<LOD	<LOD	<LOD
154	<LOD	<LOD	<LOD	0.13	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.04	0.11	<LOD	<LOD
149+139	<LOD	0.26	0.53	<LOD	<LOD	0.86	1.08	1.27	0.22	0.08	0.14	0.63	0.18	0.25
135	0.08	0.05	0.07	0.06	0.13	0.13	0.05	0.16	<LOD	0.02	<LOD	<LOD	<LOD	0.03
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	0.12	0.23	0.14	0.07	0.22	0.25	0.52	0.20	0.11	0.03	0.06	0.07	0.10	0.03
140	0.26	0.09	0.08	0.07	<LOD	0.32	0.05	<LOD	0.08	0.06	0.05	0.05	<LOD	0.05
143	0.22	0.08	0.11	<LOD	<LOD	0.05	0.12	0.31	<LOD	0.04	0.04	<LOD	<LOD	0.06
134	<LOD	<LOD	<LOD	<LOD	0.32	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	0.08	<LOD	<LOD
142+131+133	0.19	<LOD	<LOD	<LOD	0.57	<LOD	0.25	0.82	0.13	0.08	0.07	<LOD	<LOD	0.08
165	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	0.12	<LOD	<LOD	<LOD	0.07	<LOD	<LOD
132+161	0.22	<LOD	<LOD	0.21	0.22	0.17	<LOD	<LOD	<LOD	0.08	0.13	0.18	<LOD	0.06
137	<LOD	<LOD	<LOD	<LOD	0.48	0.42	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05
168	0.09	0.10	0.17	0.13	<LOD	<LOD	0.17	0.55	0.09	0.03	0.06	0.10	0.07	0.09
141	0.33	0.23	0.23	0.08	0.24	0.66	0.98	0.61	<LOD	0.08	0.02	<LOD	<LOD	<LOD
153	0.42	0.37	0.57	0.35	0.51	1.07	1.65	2.23	0.16	0.11	0.13	0.49	0.20	0.17
130	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD
158	0.26	0.09	0.11	0.07	0.19	<LOD	0.51	0.57	0.06	0.07	0.02	0.11	0.05	0.03
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	0.09	<LOD	<LOD	<LOD
138	0.52	0.40	1.02	0.49	0.74	1.14	2.20	1.91	0.16	0.13	0.12	0.68	0.11	0.41

169	0.04	<LOD	<LOD	0.04	<LOD	0.05	0.03	<LOD	0.03	0.03	0.03	0.04	0.03	0.04
157	0.15	0.10	0.19	0.06	0.35	0.20	0.48	0.70	0.06	0.07	0.04	0.21	0.05	0.03
128	0.31	0.28	0.75	0.18	0.48	1.19	0.99	1.13	0.13	0.05	0.05	0.23	0.05	0.21
159+162	0.14	<LOD	<LOD	<LOD	0.47	0.23	0.98	0.65	<LOD	<LOD	<LOD	<LOD	0.12	0.11
167	0.09	0.20	0.14	0.13	0.29	0.12	1.48	0.50	0.07	0.06	0.03	0.08	0.04	0.06
156	0.26	0.08	0.20	0.05	0.29	0.10	0.54	0.53	0.15	0.02	0.02	0.07	0.07	0.08
166	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD
129	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.29	0.44	<LOD	<LOD	<LOD	<LOD	0.03	<LOD
188	<LOD	0.08	0.03	0.26	0.35	0.07	0.15	0.17	0.07	0.06	0.02	0.20	0.06	0.02
184	<LOD	0.03	0.10	0.22	0.41	0.07	0.09	0.17	0.09	0.08	0.03	0.09	0.07	0.13
179	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
176	0.05	0.05	0.08	<LOD	0.47	0.34	<LOD	0.37	0.08	0.07	0.04	0.11	0.03	0.09
187	0.19	0.24	0.49	0.34	0.47	0.51	0.89	0.87	0.07	0.10	0.02	0.34	0.24	0.16
178	0.06	0.10	0.03	0.07	0.11	0.06	0.09	0.28	<LOD	<LOD	<LOD	0.03	0.02	0.03
175	0.14	0.17	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	0.03	<LOD
182+186	<LOD	<LOD	<LOD	0.17	0.28	0.18	0.16	1.23	0.08	<LOD	0.03	0.19	0.05	0.08
183	0.32	0.06	0.10	0.11	0.14	0.05	0.65	0.54	0.07	0.12	0.03	0.24	0.12	0.05
174	0.33	0.30	0.27	0.29	0.24	<LOD	0.08	0.40	0.11	0.11	0.03	0.24	0.15	0.19
185	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
181	<LOD	<LOD	0.48	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
171	<LOD	<LOD	<LOD	<LOD	<LOD	0.57	0.98	1.30	<LOD	<LOD	<LOD	0.55	0.21	0.20
177	0.35	0.34	0.27	0.08	0.10	0.19	0.49	0.51	0.07	<LOD	0.04	0.23	0.03	0.02
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.07	0.04	0.03	0.03	0.08	0.06	0.79	1.15	0.07	0.12	0.02	0.12	0.02	0.04
180	0.56	0.54	0.39	0.47	0.80	0.85	1.41	1.97	0.22	0.20	0.08	0.51	0.11	0.15
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	0.53	0.48	0.49	0.43	0.56	0.80	1.63	2.17	0.20	0.19	0.09	0.43	0.16	0.21
191	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
193	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.24	<LOD	0.05	<LOD	0.03	0.03	0.03
190	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
172	<LOD	0.06	0.04	<LOD	0.23	<LOD	0.16	0.76	<LOD	<LOD	<LOD	0.05	0.03	0.04
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	0.11	0.19	0.32	0.15	0.55	0.15	0.26	1.69	0.07	0.18	0.09	0.22	0.11	0.06
195	0.04	0.17	0.26	0.11	0.30	0.41	0.68	0.31	0.03	<LOD	0.06	0.10	0.10	0.05
200	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
198+199	0.09	0.14	<LOD	<LOD	0.52	0.55	0.45	0.36	<LOD	0.08	<LOD	0.12	0.13	0.22
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
194	0.13	<LOD	0.05	0.05	<LOD	0.64	1.17	0.85	<LOD	0.12	<LOD	<LOD	<LOD	0.21
205	0.04	<LOD	<LOD	<LOD	<LOD	0.09	0.16	0.03	<LOD	<LOD	0.03	<LOD	<LOD	<LOD
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑PCBs	18.47	15.06	25.33	18.71	35.79	27.36	76.20	92.21	9.52	7.48	5.27	20.85	8.15	14.27

Table A-8. Concentration of PCB congeners in the surface sediment samples (ng/g dw) in summer 2015.

PCB IUPAC No.	Coastal sites													
	CX1	CX2	CX3	CX4	CT1	CT2	CT3	CT4	ME1	ME2	ME3	SN1	SN2	SN3
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	0.03	<LOD	0.06	<LOD	0.05	0.03	0.11	0.12	<LOD	<LOD	0.02	0.03	<LOD	0.05
4	0.11	0.04	0.05	0.03	0.14	<LOD	0.24	0.13	0.03	<LOD	0.04	0.03	0.03	0.08
9	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
11	<LOD	<LOD	0.17	0.12	0.18	0.08	0.71	0.23	0.05	<LOD	<LOD	0.09	<LOD	0.15
6	0.12	0.03	0.06	<LOD	0.09	<LOD	0.11	0.06	<LOD	<LOD	<LOD	0.04	<LOD	0.02
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	0.13	0.11	0.27	0.09	0.83	0.33	1.22	1.62	0.11	0.10	0.15	0.22	0.21	0.36
13+12	<LOD	<LOD	0.13	<LOD	<LOD	0.06	0.68	0.85	0.06	<LOD	0.02	<LOD	<LOD	<LOD
15	<LOD	<LOD	<LOD	0.09	0.20	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.07	0.05	0.09
19	<LOD	<LOD	0.03	0.02	0.04	0.03	0.17	0.11	0.02	0.02	<LOD	<LOD	0.02	<LOD
30	0.06	0.03	<LOD	0.05	0.05	0.04	0.14	0.26	0.03	0.03	0.03	0.09	<LOD	0.04
18	0.22	0.16	0.42	0.26	0.56	0.41	1.32	1.49	0.19	0.15	0.10	0.41	0.39	0.43
17	0.14	0.26	0.21	0.33	0.58	0.21	1.10	1.20	0.05	<LOD	0.04	0.13	0.05	0.37
24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.11	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
32	<LOD	<LOD	0.08	<LOD	0.39	<LOD	0.26	0.12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
16	0.10	0.06	0.08	0.08	0.17	0.08	1.13	1.20	0.05	0.03	0.02	0.06	0.06	0.11
23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	0.05	0.04	0.03	<LOD	0.05	<LOD	0.37	0.29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
31	0.10	0.06	0.05	0.10	0.15	0.11	0.92	0.86	0.12	0.08	0.03	<LOD	0.14	0.22
28	0.12	0.18	0.31	0.16	0.47	0.50	1.53	1.93	0.15	0.13	0.07	0.11	0.15	0.50
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	0.14	0.03	0.33	0.31	0.67	0.52	1.90	1.45	0.24	0.15	0.06	0.35	0.29	0.44
22	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
36	<LOD	<LOD	0.03	0.03	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
39	<LOD	0.03	0.07	0.02	0.03	0.03	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	0.03
38	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	<LOD	0.04	<LOD	0.03	0.07	0.04	0.09	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.05
37	0.06	0.06	0.06	0.05	0.31	0.11	0.23	0.59	0.07	0.08	0.04	0.18	0.07	0.11
54	0.03	<LOD	0.02	0.02	0.03	0.03	<LOD	<LOD	<LOD	<LOD	0.02	0.03	<LOD	0.03
50	<LOD	0.02	<LOD	0.05	0.07	0.03	0.14	0.09	0.02	0.02	<LOD	0.05	<LOD	<LOD
53	0.05	0.03	0.02	0.03	0.17	<LOD	0.07	0.19	<LOD	<LOD	<LOD	0.05	<LOD	0.06
51	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
45	<LOD	<LOD	0.03	<LOD	0.28	0.04	0.76	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
52	0.16	0.10	0.17	0.41	0.36	0.34	2.97	2.75	0.04	0.11	0.04	0.32	0.52	0.03
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	0.24	0.18	0.20	0.53	0.62	0.68	2.33	2.21	0.38	0.13	0.12	0.53	0.41	0.54
65+75+47+48	<LOD	0.08	0.06	0.08	0.28	<LOD	0.11	1.67	<LOD	<LOD	0.02	<LOD	0.04	0.14
62	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	0.12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
44	0.16	0.11	0.22	0.26	0.69	0.52	1.15	2.42	0.21	0.04	0.03	0.44	0.13	0.32
42	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
64	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.75	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	0.07	0.15	0.03	0.36	0.47	0.05	0.93	1.58	0.08	<LOD	<LOD	<LOD	<LOD	0.39
41	<LOD	<LOD	0.05	0.19	0.07	<LOD	<LOD	1.06	<LOD	<LOD	<LOD	0.14	<LOD	0.34
68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
67+57	<LOD	<LOD	0.04	0.36	<LOD	0.03	1.18	0.95	<LOD	<LOD	<LOD	<LOD	<LOD	0.13
40	0.14	0.03	0.08	0.13	0.08	0.03	0.15	0.29	0.03	0.02	0.02	0.03	0.16	0.04
61+63+58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

74	0.21	0.03	0.23	0.18	0.39	0.09	0.94	1.72	0.05	0.04	0.02	0.42	0.03	0.15
66	0.18	0.17	0.30	0.29	0.53	0.17	1.58	2.17	0.16	0.09	0.05	0.49	0.08	0.40
55+76+80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
70	0.03	0.38	0.50	0.29	0.58	0.41	2.25	2.35	0.17	0.12	0.10	0.66	0.14	0.48
60+56	<LOD	0.13	<LOD	<LOD	<LOD	0.04	0.68	0.18	<LOD	<LOD	<LOD	<LOD	<LOD	0.11
81	0.09	0.13	0.22	0.42	0.48	0.18	0.65	1.51	0.17	0.11	0.14	0.29	0.27	0.29
77	0.05	0.09	0.10	0.14	0.11	0.21	1.18	1.38	0.10	0.03	0.03	0.06	0.14	0.30
79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
78	0.08	<LOD	0.12	0.04	<LOD	<LOD	1.64	0.15	<LOD	<LOD	<LOD	0.02	<LOD	0.04
104	0.11	0.02	0.03	0.03	<LOD	0.03	0.12	0.56	0.02	0.03	<LOD	0.04	0.03	0.03
96	0.14	0.03	<LOD	0.06	0.08	0.03	0.14	0.20	0.06	<LOD	0.03	0.07	0.06	0.05
103	0.03	<LOD	0.04	0.02	0.11	0.06	0.25	0.76	0.05	<LOD	<LOD	0.03	0.02	0.02
100	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
94	<LOD	<LOD	0.02	<LOD	0.16	<LOD	<LOD	0.15	<LOD	<LOD	<LOD	<LOD	0.03	0.04
102+93+98+95	0.34	0.25	0.49	0.22	1.20	0.47	3.23	2.80	0.37	0.26	0.13	0.82	0.30	0.59
88	<LOD	0.03	<LOD	<LOD	0.74	<LOD	<LOD	0.18	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
91+121	<LOD	0.04	0.04	0.10	0.77	0.05	1.69	0.30	<LOD	0.07	<LOD	0.09	<LOD	0.06
92+84	0.19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	0.07	0.07	0.05	0.29	<LOD	0.36	2.81	<LOD	0.20	<LOD	0.03	0.28	0.32	0.36
101	0.60	0.31	0.69	0.88	1.49	0.94	4.79	3.84	0.14	0.33	0.28	0.76	0.58	0.63
99	0.09	0.21	0.13	0.62	0.74	0.81	2.66	2.59	0.19	0.05	0.06	0.14	0.13	0.06
113	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.07	0.09	0.06	0.08	0.58	0.09	0.64	0.50	0.04	0.02	0.02	0.08	0.10	0.06
83+115	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	1.77	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
86+117+97	0.08	0.03	0.14	0.11	0.22	0.17	2.45	0.45	<LOD	0.05	<LOD	0.14	0.04	0.05
125+116+108	<LOD	0.13	<LOD	0.21	0.63	<LOD	0.63	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	0.07
87	0.37	0.03	0.16	0.15	0.85	0.32	2.34	1.52	0.06	0.03	0.02	0.17	0.04	0.35
111	0.15	0.47	0.24	<LOD	0.76	0.60	0.82	1.18	<LOD	0.06	<LOD	<LOD	<LOD	0.20
85	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
120+110	0.56	0.35	0.88	0.89	1.18	0.96	4.31	2.87	0.40	0.38	0.14	1.15	0.33	0.78
82	<LOD	0.05	<LOD	<LOD	0.81	<LOD	0.77	0.50	<LOD	<LOD	<LOD	<LOD	<LOD	0.06
106+124	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.08	0.02	0.25	0.02	0.49	0.04	0.79	0.40	0.10	0.07	0.02	0.09	0.03	0.06
118	0.54	0.31	0.80	0.33	1.07	0.68	3.10	3.62	0.27	0.43	0.19	0.13	0.38	0.78
114	0.23	0.06	0.29	0.32	0.77	0.24	1.29	1.82	0.06	0.12	0.03	0.11	0.17	0.47
122+127	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	0.35	0.28	0.72	0.77	0.78	0.27	2.61	2.18	0.23	0.38	0.19	0.64	0.30	0.40
126	<LOD	<LOD	0.02	0.02	0.01	0.02	0.02	0.02	0.02	<LOD	0.02	0.02	<LOD	0.02
155	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	0.05	0.72	<LOD	0.03	<LOD	<LOD	<LOD	<LOD
150	0.03	0.04	0.05	<LOD	0.06	0.03	<LOD	0.13	0.03	<LOD	<LOD	<LOD	<LOD	0.03
152	<LOD	0.05	0.03	0.02	0.12	<LOD	0.05	0.35	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
145	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.11	0.09	0.07	0.03	0.25	0.15	0.36	1.03	0.04	<LOD	0.02	<LOD	0.20	<LOD
154	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
149+139	0.45	0.33	0.77	0.32	0.62	0.55	1.49	2.10	0.30	0.21	0.31	0.56	0.56	0.79
135	<LOD	0.02	0.04	<LOD	0.13	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	0.05	0.04	0.09	0.02	0.89	0.19	1.28	1.57	0.04	0.08	0.03	0.04	0.11	0.12
140	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
143	0.10	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
134	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
142+131+133	0.13	0.07	0.09	0.08	0.29	0.17	0.42	1.01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
165	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	0.04	<LOD	<LOD	<LOD	0.18	0.03	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
132+161	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
137	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
168	0.07	0.11	0.21	0.09	0.93	0.15	0.18	0.21	0.04	0.05	0.03	0.07	0.05	0.12
141	0.02	0.09	0.36		0.50	0.40	1.10	1.74	<LOD	<LOD	<LOD	0.32	<LOD	<LOD
153	0.49	0.36	0.72	0.48	1.14	0.82	2.16	3.83	0.33	0.20	0.10	0.27	0.36	0.60
130	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
158	0.04	0.08	0.15	0.05	0.25	0.09	0.81	1.33	0.05	0.04	0.02	0.26	0.13	0.10
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
138	0.59	0.42	0.90	0.34	0.97	0.67	3.31	4.47	0.49	0.24	0.23	0.40	0.31	0.70

169	0.07	0.06	0.07	0.07	<LOD	0.05	0.02	0.05	0.04	0.02	0.02	0.03	0.03	0.02
157	0.07	0.06	0.06	0.10	0.62	0.14	0.41	0.74	0.03	0.03	0.03	0.04	0.04	0.14
128	0.46	0.27	0.74	0.35	1.13	0.97	1.20	2.14	0.21	0.16	0.26	0.23	0.35	0.55
159+162	<LOD	<LOD	0.21	0.07	0.42	<LOD	<LOD	0.24	<LOD	<LOD	<LOD	<LOD	<LOD	0.17
167	0.09	0.11	0.15	0.14	0.83	0.18	0.57	1.32	0.02	0.07	0.04	0.05	0.05	0.14
156	0.19	0.09	0.26	0.08	0.62	0.18	1.22	0.67	0.02	0.12	0.05	0.04	0.06	0.53
166	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.09
129	<LOD	0.05	0.05	<LOD	0.06	0.03	0.29	0.27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
188	<LOD	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
184	0.04	0.03	0.03	0.03	<LOD	0.03	<LOD	0.52	<LOD	<LOD	<LOD	0.03	<LOD	0.10
179	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
176	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	0.03	0.16	<LOD	<LOD	<LOD	<LOD	<LOD	0.05
187	0.17	0.05	0.11	0.04	0.34	0.05	0.45	1.46	0.07	0.05	0.03	0.06	0.33	0.07
178	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
175	<LOD	<LOD	<LOD	<LOD	0.16	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	0.08
182+186	0.05	0.09	0.13	0.15	0.25	0.23	0.42	0.55	<LOD	<LOD	<LOD	0.03	<LOD	0.15
183	0.06	0.10	0.26	0.07	0.31	0.44	0.66	0.74	0.03	0.05	0.04	0.06	0.21	0.05
174	0.10	0.25	0.51	0.26	0.79	0.69	1.13	0.58	0.31	0.16	0.05	0.03	0.11	0.42
185	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.14	<LOD	<LOD	<LOD	<LOD	<LOD	0.03
181	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
171	0.25	0.21	0.79	0.24	0.68	0.67	1.32	1.20	0.38	0.14	0.33	0.82	0.25	0.68
177	0.04	0.11	0.10	0.03	0.13	0.25	0.63	0.49	0.04	0.09	0.03	0.13	0.06	0.12
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.11	0.06	0.07	0.20	0.29	0.16	0.23	1.13	0.04	0.06	0.05	0.07	0.17	0.09
180	0.56	0.34	0.81	0.37	1.03	0.87	2.87	3.85	0.26	0.28	0.21	0.50	0.50	0.65
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	0.23	0.37	0.93	0.36	0.69	0.78	1.96	1.06	0.34	0.14	0.24	0.63	0.63	0.80
191	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
193	<LOD	<LOD	0.21	<LOD	<LOD	0.06	0.12	0.11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
190	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
172	0.02	0.02	0.22	0.08	<LOD	0.08	0.20	0.56	<LOD	<LOD	<LOD	<LOD	<LOD	0.03
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	0.11	0.09	0.05	0.03	0.35	0.03	0.05	0.30	0.06	0.03	0.03	0.02	0.04	0.14
195	<LOD	0.19	0.12	<LOD	0.32	<LOD	0.23	0.41	0.02	<LOD	<LOD	<LOD	0.02	0.02
200	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
198+199	<LOD	<LOD	0.27	0.09	0.51	<LOD	0.13	0.94	<LOD	<LOD	0.02	<LOD	0.09	0.09
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
194	0.22	0.38	0.08	<LOD	0.60	0.07	0.37	0.71	0.03	0.07	0.07	0.16	0.13	<LOD
205	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	0.07	0.09	<LOD	<LOD	<LOD	0.05	<LOD	<LOD
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑PCBs	12.10	10.23	19.76	14.86	40.32	20.79	92.26	105.3	8.05	6.12	4.61	15.08	11.06	19.62

Table A-9. Physical and chemical characteristics of surface water and sediment collected from the coastal area of Bangladesh.

		Water				Sediment							
Season	Site	pH	Temper-ature (°C)	Salinity (‰)	TSS (mg/L)	pH	TOC (%)	BC (%)	TN (%)	Clay (%)	Silt (%)	Sand (%)	Texture*
Winter	CX1	7.3	19.5	17.3	465	7.1	1.28	0.21	0.3	10.6	18.1	71.2	Sandy Loam
	CX2	6.2	20	18.4	340	6.1	0.66	0.14	0.1	9.1	11.7	79.2	Loamy Fine Sand
	CX3	7.2	20.5	19.6	490	6.9	0.94	0.33	0.1	22.2	34.3	43.5	Loam
	CX4	7.1	20.5	18.6	435	6.5	1.27	0.15	0.2	25.5	27.1	47.4	Sandy Clay Loam
	CT1	6.6	19.5	21.3	280	6.5	0.98	0.39	0.1	13.3	21.4	65.3	Sandy Loam
	CT2	6.3	21	19.5	310	6.1	0.40	0.13	0.1	15.8	70.6	13.5	Silt Loam
	CT3	6.7	22	17.4	230	5.9	0.55	0.14	0.1	44.9	25.4	29.7	Clay
	CT4	6.5	21.5	18.6	260	6.3	1.05	0.53	0.1	13.9	19.7	66.4	Sandy Loam
	ME1	7.7	19	16.8	800	7.5	0.31	0.15	0.0	2.8	70.8	26.4	Silt Loam
	ME2	7.1	19.5	20.4	650	6.9	0.07	0.06	0.0	4.2	64.5	31.3	Silt Loam
	ME3	7.8	22	21.6	460	7.3	0.06	0.06	0.0	6.2	65.4	28.4	Silt Loam
	SN1	6.9	19	17.3	850	6.7	0.39	0.13	0.0	20.0	68.8	11.3	Silt Loam
	SN2	7.6	21	18.6	730	7.3	0.24	0.07	0.0	17.2	73.5	9.3	Silt Loam
	SN3	6.9	23	19.8	960	7.2	0.15	0.06	0.0	13.5	71.3	15.2	Silt Loam
Summer	CX1	7.2	24.5	22.5	500	7.3	0.69	0.11	0.2	13.4	16.4	70.2	Sandy Loam
	CX2	6.5	23.3	24.0	350	6.1	0.64	0.13	0.1	7.6	15.8	76.5	Sandy Loam
	CX3	6.8	24.1	12.5	480	6.7	0.49	0.12	0.0	23.2	32.3	44.5	Loam
	CX4	7.1	23.5	16.5	460	6.6	0.55	0.15	0.1	23.2	31.3	45.5	Loam
	CT1	6.5	22.3	13.5	350	6.3	0.49	0.19	0.0	5.9	27.4	66.6	Sandy Loam
	CT2	6.2	23.8	15.5	440	6.3	0.43	0.13	0.1	17.8	38.4	43.8	Loam
	CT3	6.3	23.6	17.0	250	6.1	0.62	0.28	0.1	44.1	28.4	27.6	Clay
	CT4	6.5	23.5	18.5	290	6.3	0.53	0.27	0.0	20.8	21.3	57.9	Sandy Clay Loam
	ME1	7.5	22	4.5	850	7.5	0.22	0.07	0.0	10.1	61.7	28.3	Silt Loam
	ME2	6.8	24.3	3.5	630	6.8	0.19	0.06	0.0	8.3	65.3	26.4	Silt Loam
	ME3	7.3	23.6	7.0	520	7.3	0.15	0.06	0.0	6.2	66.3	27.5	Silt Loam
	SN1	6.5	23.5	13.5	900	6.6	0.49	0.28	0.1	18.0	68.3	13.7	Silt Loam
	SN2	6.2	24.4	14.0	750	6.7	0.31	0.16	0.1	11.5	77.3	11.3	Silt Loam
	SN3	5.5	24.9	16.5	850	5.9	0.37	0.19	0.1	11.8	69.6	18.6	Silt Loam

*According to the United States Department of Agriculture (USDA) soil texture classification.

Table A-10. Concentration of PCB congeners in the seafood (finfish and shellfish) samples (ng/g ww) in winter 2015.

PCB IUPACNo.	Ilish				Rupchanda		Loitta				Sole		Poa				Shrimp				Crab			
	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	BH	SN	CX	CT	BH	SN
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	<LOD	0.02	<LOD	0.02	0.02	0.02	0.02	0.03	<LOD	0.02	0.03	0.02	0.02	0.03	0.02	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02
4	0.05	<LOD	0.02	<LOD	<LOD	<LOD	0.04	0.04	<LOD	0.03	<LOD	0.04	0.05	0.02	0.03	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.02	<LOD	<LOD
9	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	0.12	0.06	<LOD	0.04	0.04	0.03	<LOD	0.02	0.02	0.05	<LOD	0.03	<LOD	<LOD	0.02	0.03	0.03	<LOD	<LOD	0.11	0.20	<LOD	<LOD	0.08
11	0.20	0.17	0.12	0.17	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.11	0.04	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.23	0.13	0.03	0.20
6	0.09	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.15	<LOD	0.03	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.03	<LOD	<LOD
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	0.72	0.78	0.84	0.99	0.26	0.22	0.46	0.32	0.14	0.22	0.67	0.44	0.33	0.29	0.31	0.12	0.21	0.13	0.03	0.33	0.97	0.67	0.41	0.90
13+12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
15	0.14	0.02	0.18	0.11	0.06	0.04	0.03	<LOD	<LOD	0.10	0.08	<LOD	<LOD	<LOD	<LOD	0.04	0.02	0.03	<LOD	0.09	0.03	0.09	0.02	0.15
19	0.05	0.02	0.02	0.03	0.02	<LOD	0.03	0.02	0.02	0.02	0.02	0.02	0.02	<LOD	0.02	0.02	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD
30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
18	0.67	1.12	0.88	0.85	0.24	0.28	0.43	0.45	0.15	0.20	0.39	0.45	0.24	0.25	0.23	0.15	0.11	0.12	0.09	0.41	0.96	0.87	0.57	0.51
17	0.11	0.77	0.39	0.51	0.07	0.14	0.20	0.14	0.05	0.04	0.27	0.14	0.15	0.14	0.06	0.07	0.03	0.04	0.06	0.29	0.65	0.23	0.33	0.33
24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.06	0.02	<LOD	0.03
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
32	0.04	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.03	0.03	0.07	<LOD	<LOD
16	0.33	0.19	0.13	0.08	0.03	0.04	0.05	0.06	0.02	0.09	0.06	0.23	0.05	0.05	0.03	0.04	0.13	0.05	0.04	0.16	0.82	0.51	0.03	0.15
23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD
29	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	1.18	0.44	0.52	1.41	0.43	0.31	0.40	0.36	0.21	0.18	0.43	0.47	0.45	0.27	0.27	0.23	0.33	0.12	0.10	0.23	<LOD	0.02	0.91	0.72
25	0.08	0.03	0.04	0.05	0.02	0.07	0.04	0.02	<LOD	0.03	0.04	0.05	0.03	0.02	0.04	<LOD	0.02	0.03	0.02	0.04	<LOD	<LOD	<LOD	<LOD
31	0.72	0.23	0.30	0.21	0.15	0.13	0.29	0.17	0.07	0.05	0.11	0.13	0.25	0.13	0.05	0.08	0.27	0.08	0.08	0.23	0.88	0.59	0.04	0.58
28	0.84	0.89	1.17	1.10	0.17	0.22	0.34	0.26	0.13	0.16	0.43	0.50	0.34	0.27	0.23	0.18	0.05	0.13	0.12	0.28	0.48	0.97	0.40	0.46
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.11	<LOD	<LOD	0.05	<LOD	0.02	<LOD	0.04	<LOD	0.04	0.12	0.39	<LOD	0.09
22	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
36	0.04	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD
39	<LOD	0.03	0.03	0.04	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.02	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.05	<LOD	<LOD
38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	<LOD	0.03	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.02	0.02	0.03	0.03	<LOD	<LOD
37	0.18	0.07	0.09	0.08	0.03	<LOD	0.03	0.02	0.03	0.03	0.04	0.03	0.06	0.07	0.02	0.05	0.03	0.02	0.03	0.03	0.20	0.30	0.03	0.02
54	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD
50	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
53	<LOD	0.05	0.02	<LOD	0.02	0.02	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	0.03	0.02	0.02	<LOD	<LOD	0.06	0.02	<LOD
51	0.03	0.04	<LOD	0.03	<LOD	<LOD	<LOD	0.02	0.02	0.02	<LOD	0.02	0.03	0.02	0.03	0.02	0.02	<LOD	0.03	<LOD	0.03	<LOD	<LOD	<LOD
45	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.02	<LOD	0.09	<LOD	<LOD
52	2.14	2.13	1.54	1.82	0.14	0.16	0.72	0.76	0.22	0.36	0.62	0.79	0.65	0.58	0.40	0.22	0.53	0.43	0.32	0.50	1.33	0.99	0.66	1.12
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	1.30	1.86	2.15	1.24	0.46	0.28	0.60	0.57	0.18	0.31	0.44	0.70	0.38	0.67	0.33	0.27	0.21	0.15	0.10	0.22	0.63	1.08	0.24	0.73
65+75+47+48	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
62	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD
44	0.04	1.11	0.23	0.15	0.11	0.10	0.06	0.04	0.04	0.03	0.06	0.07	0.03	0.04	0.03	0.03	0.05	0.05	0.03	0.07	0.23	0.54	0.08	0.04
42	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD
64	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.02	<LOD	0.05	<LOD	<LOD
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	0.05	0.02	0.04	0.04	0.03	0.03	<LOD	0.02	0.03	0.03	0.04	0.03	0.04	0.03	0.04	<LOD	0.07	0.14	0.02	<LOD	0.23	0.19	0.14	<LOD
41	0.10	0.99	0.54	0.32	0.06	0.14	0.03	0.03	0.03	0.03	0.07	0.26	0.06	0.03	0.06	0.03	<LOD	0.02	0.11	0.09	0.46	0.34	0.06	0.27
68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
67+57	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.16	<LOD	<LOD
40	0.24	0.42	0.30	0.22	0.03	0.10	0.01	0.07	0.05	0.05	0.08	0.09	0.10	0.07	0.09	0.08	0.03	0.02	0.04	0.02	0.12	0.29	0.08	0.02
61+63+58	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
74	0.13	0.17	0.14	0.04	0.09	0.04	0.12	0.03	0.05	0.06	0.12	0.05	0.04	0.09	0.03	0.11	0.23	0.06	0.03	0.05	0.04	0.03	0.18	0.03
66	1.54	2.04	0.77	1.10	0.07	0.12	0.34	0.29	0.17	0.14	0.39	0.56	0.55	0.46	0.08	0.20	0.42	0.18	0.24	0.38	0.58	1.19	0.41	0.59
55+76+80	1.49	2.30	1.25	2.14	0.55	0.26	0.46	0.67	0.15	0.26	0.55	0.94	0.33	0.58	0.33	0.25	0.17	0.15	0.15	0.42	0.76	<LOD	<LOD	<LOD
70	0.13	1.35	0.17	0.15	0.08	0.03	0.12	0.07	0.04	0.04	0.17	0.42	0.20	0.08	0.12	0.06	0.11	0.04	0.05	0.06	0.49	<LOD	0.16	0.02
60+56	0.10	0.51	0.12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.17	<LOD	<LOD	0.03
81	0.30	0.29	0.19	0.08	0.13	0.10	0.05	0.04	0.05	0.03	0.05	0.27	0.08	0.40	0.09	0.12	0.03	0.04	0.06	0.20	0.55	0.48	0.23	0.21
77	0.12	0.21	0.12	0.04	0.05	0.02	0.02	0.02	<LOD	<LOD	<LOD	0.03	0.04	0.07	<LOD	<LOD	<LOD	0.02	<LOD	0.02	0.33	0.12	<LOD	0.29
79	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD
78	0.06	0.10	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.03	<LOD	0.03	<LOD	<LOD
104	0.14	0.06	<LOD	<LOD	<LOD	<LOD	0.02	0.03	<LOD	<LOD	0.04	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	1.01	<LOD	<LOD

96	3.09	3.16	2.31	2.65	<LOD	<LOD	0.63	0.94	0.36	0.37	1.03	0.98	0.49	0.74	0.44	0.33	0.21	0.33	0.20	0.37	1.02	<LOD	0.36	0.94
103	0.05	0.09	<LOD	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD
100	0.11	0.04	0.03	0.04	0.03	0.02	0.02	0.04	0.02	0.02	0.02	<LOD	0.02	0.03	0.02	0.02	0.02	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD
94	<LOD	0.17	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	0.03	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.05	<LOD	<LOD
102+93+98+95	3.50	3.79	3.53	3.24	0.86	0.65	0.91	1.21	0.41	0.33	1.09	1.27	0.57	0.95	0.49	0.38	0.48	0.38	0.14	0.42	0.98	1.34	0.56	1.26
88	0.03	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	0.04	0.11	<LOD	<LOD
91+121	0.10	0.09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.30	<LOD	<LOD
92+84	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	0.20	1.01	0.26	0.10	0.08	0.05	0.04	0.17	<LOD	0.03	0.09	0.15	0.07	0.13	0.08	<LOD	<LOD	<LOD	<LOD	0.09	0.09	1.05	0.12	0.03
101	2.35	2.33	2.87	2.37	0.46	0.29	0.42	1.05	0.16	0.39	1.20	1.14	0.65	0.69	0.31	0.21	0.25	0.27	0.09	0.35	0.34	0.80	0.02	0.37
99	1.32	1.93	1.48	1.27	0.28	0.18	0.30	0.62	0.09	0.12	0.56	1.01	0.44	0.36	0.21	0.23	0.21	0.11	0.06	0.10	0.94	0.75	0.69	0.91
113	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.22	0.13	0.13	0.10	0.03	0.05	0.05	0.06	0.05	0.03	0.06	0.12	0.06	0.02	0.04	0.04	0.02	0.03	0.02	<LOD	0.03	0.15	0.07	0.02
83+115	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04
86+117+97	0.33	0.22	0.07	0.13	0.05	<LOD	0.03	0.05	0.04	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.10	<LOD	<LOD
125+116+108	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD
87	0.80	1.09	0.10	0.16	0.09	0.04	0.04	0.06	0.08	0.04	0.08	0.09	0.08	0.09	0.06	0.03	0.05	0.02	0.05	0.04	0.15	0.48	0.37	0.12
111	0.27	0.18	0.04	0.03	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	0.04	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.12	0.41	0.02	<LOD
85	<LOD	<LOD	<LOD	<LOD	0.02	0.10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD
120+110	3.37	3.37	1.73	0.96	0.19	0.33	0.70	0.35	0.16	0.09	1.11	1.35	0.77	0.81	0.35	0.18	0.41	0.18	0.15	0.30	0.90	1.62	0.12	1.04
82	0.14	0.07	0.21	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.04	<LOD	0.02
106+124	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.68	0.18	0.35	0.34	0.06	0.06	0.04	0.09	0.07	0.04	0.11	0.07	0.18	0.13	0.05	0.04	0.13	0.09	0.08	0.11	0.58	0.36	0.28	0.39
118	3.10	1.84	1.50	2.61	0.41	0.43	0.98	1.01	0.31	0.34	0.98	1.31	0.67	0.72	0.59	0.35	0.32	0.23	0.13	0.37	1.32	1.53	0.50	0.84
114	0.68	0.22	0.28	0.07	0.04	0.13	0.12	0.13	0.07	0.03	0.08	0.59	0.16	0.06	0.10	0.12	0.14	0.14	0.05	0.17	0.42	0.21	0.17	0.59
122+127	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	2.29	2.29	2.40	1.47	0.09	0.21	0.49	0.76	0.19	0.30	0.78	1.14	0.42	0.48	0.46	0.29	0.43	0.31	0.15	0.31	0.59	0.35	0.47	0.46
126	0.06	0.14	0.02	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.09	0.14	<LOD	0.12
155	<LOD	0.02	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.02	<LOD	<LOD	0.02
150	0.04	0.07	<LOD	0.02	0.02	<LOD	0.03	0.03	0.02	<LOD	0.02	<LOD	0.02	0.03	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.03	0.03	<LOD	0.02
152	<LOD	0.10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.02	0.05	<LOD	<LOD	0.02	<LOD	<LOD	0.02	0.02	0.13	0.09	0.02	0.11
145	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05
154	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
149+139	3.30	3.13	2.62	2.61	0.44	0.33	0.79	0.99	0.27	0.31	1.16	1.44	0.97	1.19	0.85	0.30	0.44	0.20	0.27	0.43	0.97	1.61	1.48	1.29
135	<LOD	0.25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	0.04	2.35	1.10	1.46	0.36	0.19	0.47	0.61	0.02	0.18	0.69	1.15	0.55	0.77	0.42	0.28	0.32	0.11	0.03	0.11	0.92	0.74	0.18	1.01
140	<LOD	<LOD	<LOD	<LOD	0.09	<LOD	<LOD	0.03	<LOD	0.03	0.08	<LOD	0.03	0.08	<LOD	0.03	<LOD	<LOD	<LOD	0.19	0.15	<LOD	0.04	<LOD
143	0.14	0.13	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.06	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	0.03
134	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
142+131+133	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.13	<LOD	0.04
165	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	0.07	0.08	0.05	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	0.02	<LOD	0.11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.07	<LOD
132+161	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
137	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
168	1.22	0.31	0.11	0.07	0.03	0.05	0.08	0.08	0.05	0.03	0.09	0.10	0.23	0.12	0.02	0.05	0.03	0.02	0.02	0.05	0.20	0.25	0.20	0.35
141	0.41	0.10	0.43	0.10	<LOD	0.03	<LOD	0.03	<LOD	<LOD	0.18	0.17	0.04	<LOD	0.08	0.05	0.07	<LOD	0.05	0.28	0.45	0.26	0.37	0.08
153	4.61	4.88	4.21	3.72	1.02	0.66	1.17	1.32	0.52	0.40	1.83	1.79	1.20	1.50	1.28	0.47	0.25	0.26	0.11	0.45	1.13	1.44	0.76	0.93
130	0.06	0.02	0.03	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.08	0.03	0.04	0.03	0.03	<LOD	<LOD	<LOD	0.02	0.03	<LOD	<LOD	0.02
158	0.36	0.14	0.07	0.08	0.04	0.04	0.08	0.07	0.03	0.02	0.07	0.12	0.06	0.07	0.15	0.03	0.02	0.04	0.03	0.05	0.14	0.09	0.29	0.09
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
138	3.56	3.14	2.16	2.66	0.51	0.49	0.65	1.02	0.23	0.24	1.38	1.36	1.08	1.43	1.11	0.37	0.34	0.32	0.24	0.53	0.91	1.33	1.25	0.59
169	0.11	<LOD	<LOD	0.09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.06	0.09	<LOD	<LOD	0.33	0.27	0.11	<LOD
157	0.45	0.23	0.39	0.49	0.07	0.04	0.11	0.08	0.02	0.02	0.15	0.18	0.08	0.17	0.08	0.02	0.02	0.03	0.02	0.05	0.14	0.28	0.37	0.34
128	2.92	2.45	2.12	1.67	0.17	0.18	0.45	0.41	0.17	0.15	0.90	0.63	0.56	1.03	0.41	0.22	0.23	0.18	0.10	0.28	1.17	0.58	1.00	0.80
159+162	<LOD	0.05	<LOD	0.12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	0.03	<LOD	0.06	0.02
167	0.33	0.57	0.03	0.28	0.05	0.03	0.10	0.17	0.02	0.04	0.18	0.16	0.11	0.11	0.09	0.03	0.03	0.07	0.04	0.12	0.14	0.16	0.58	0.21
156	0.49	1.32	0.11	0.20	0.09	0.10	0.11	0.05	0.03	0.02	0.26	0.24	0.13	0.27	0.17	0.04	0.05	0.04	0.10	0.21	0.27	0.26	0.19	0.15
166	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD
129	<LOD	0.08	0.03	0.07	<LOD	<LOD	0.03	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.07	0.04	0.02
188	<LOD	0.04	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.08	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.02	<LOD	<LOD	<LOD
184	0.10	0.09	0.04	0.04	<LOD	0.03	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.02	0.02	0.02	<LOD	<LOD	<LOD	0.02
179	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04
176	0.25	0.15	0.05	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
187	3.64	3.64	2.35	2.79	0.37	0.33	0.64	0.94	0.28	0.34	0.95	1.29	0.93	1.27	0.47	0.24	0.27	0.27	0.24	0.23	1.46	1.15	1.37	1.26
178	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.17	<LOD	<LOD
175	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.09	0.06	<LOD	0.02	<LOD
182+186	<LOD	0.11	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	0.07	0.05	<LOD	<LOD	<LOD	0.05	0.18	<LOD	0.48	0.08	0.07
183	0.77	0.24	0.06	0.05	0.06	0.04	0.03	0.07	0.02	0.02	0.05	0.18	0.38	0.03	0.03	0.02	0.03	0.05	0.08	0.12	0.03	0.27	0.35	0.22
174	1.70	1.20	1.07	0.44	0.03	0.05	0.07	0.12	0.03	0.03	0.11	0.27	0.22	0.05	0.02	0.03	0.06	0.15	0.16	0.27	0.26	0.35	0.11	0.25
185	<LOD	0.08	0.06	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	0.03
181	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

171	2.51	1.49	0.19	0.19	0.10	0.08	0.06	0.17	0.05	0.02	0.07	0.03	0.31	0.16	0.03	0.04	0.07	0.13	0.12	0.06	0.31	0.29	0.53	0.26
177	1.40	2.55	1.49	1.16	0.12	0.16	0.11	0.46	0.07	0.08	0.56	0.20	0.38	0.29	0.07	0.07	0.06	0.19	0.05	0.14	0.12	0.85	0.14	0.80
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.15	0.55	0.23	0.12	0.07	0.03	0.06	0.10	0.02	0.04	0.15	0.12	0.29	0.06	0.06	0.09	0.03	0.10	0.02	0.09	0.23	0.48	0.26	0.12
180	3.67	3.90	3.42	3.26	0.77	0.44	0.84	1.16	0.32	0.52	1.41	1.50	0.79	1.19	0.76	0.33	0.45	0.35	0.19	0.45	2.18	1.81	1.16	1.60
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	2.91	3.61	2.26	2.69	0.65	0.36	0.67	0.74	0.22	0.36	1.30	1.25	0.62	0.78	0.35	0.30	0.33	0.27	0.13	0.33	1.42	1.24	0.84	1.25
191	<LOD	0.05	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.03	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	<LOD
193	0.16	0.09	0.04	0.24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.02	0.09	0.03	<LOD	0.04
190	0.08	0.14	0.06	0.08	0.03	<LOD	0.03	0.03	0.02	0.03	0.05	<LOD	<LOD	0.10	0.03	<LOD	<LOD	0.02	0.03	0.08	<LOD	<LOD	<LOD	<LOD
172	0.22	0.28	0.14	0.28	<LOD	0.03	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.04	0.17	0.13	<LOD	0.09
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	0.41	0.71	0.62	0.43	<LOD	0.12	0.30	0.20	0.09	0.09	0.18	0.27	0.26	0.17	0.11	0.09	0.09	0.03	<LOD	0.18	0.64	0.75	0.39	0.63
195	0.06	0.07	0.07	0.06	0.08	0.02	0.06	<LOD	<LOD	0.02	0.09	0.09	0.04	0.03	0.04	0.03	<LOD	<LOD	0.03	0.07	0.03	0.07	0.02	0.04
200	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	0.03
198+199	<LOD	0.20	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.24	<LOD	0.16
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
194	<LOD	<LOD	0.12	<LOD	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	0.12	<LOD	0.17	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.36	0.20	0.22	<LOD
205	0.10	0.05	0.06	0.11	<LOD	<LOD	0.10	<LOD	<LOD	0.03	<LOD	0.11	<LOD	0.09	<LOD	<LOD	<LOD	<LOD	0.02	0.04	0.02	<LOD	<LOD	<LOD
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Mono-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Di-CB	1.32	1.05	1.24	1.33	0.38	0.31	0.54	0.41	0.16	0.42	1.04	0.56	0.50	0.42	0.38	0.20	0.26	0.17	0.03	0.57	1.43	0.94	0.46	1.34
Tri-CB	4.25	3.83	3.62	4.35	1.18	1.22	1.81	1.51	0.68	0.80	1.94	2.12	1.63	1.28	0.94	0.85	1.03	0.63	0.55	1.83	4.26	4.08	2.32	2.90
Tetra-CB	7.75	13.60	7.62	7.37	1.87	1.43	2.53	2.64	1.03	1.34	2.62	4.22	2.56	3.11	1.61	1.37	1.90	1.34	1.26	2.12	5.96	5.72	2.26	3.34
Penta-CB	22.84	22.45	17.40	15.70	2.71	2.59	4.80	6.62	2.03	2.14	7.30	9.52	4.65	5.26	3.22	2.21	2.71	2.13	1.14	2.80	7.76	10.81	3.73	7.14
Hexa-CB	18.36	19.43	13.49	13.69	2.93	2.15	4.07	4.96	1.37	1.51	7.05	7.53	5.27	6.86	4.68	1.97	1.86	1.38	1.02	2.89	7.27	7.64	7.00	6.17
Hepta-CB	17.54	18.22	11.54	11.48	2.22	1.53	2.53	3.81	1.03	1.57	4.67	5.11	4.25	4.15	1.86	1.12	1.33	1.60	1.11	2.14	6.48	7.26	4.87	6.03
Octa-CB	0.57	1.03	0.88	0.60	0.08	0.14	0.46	0.31	0.09	0.13	0.39	0.47	0.46	0.29	0.15	0.12	0.12	0.05	0.05	0.28	1.12	1.26	0.63	0.86
Nona-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Deca-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑DL-PCBs[a]	8.76	7.85	5.62	5.79	1.04	1.16	2.09	2.51	0.78	0.87	2.73	4.11	2.16	2.47	1.68	1.12	1.24	1.14	0.64	1.66	5.00	4.64	3.15	3.72
∑iPCBs[b]	17.18	17.27	15.37	14.93	3.07	2.26	4.14	5.58	1.58	2.07	6.87	7.08	4.71	5.67	4.09	1.77	1.88	1.77	1.06	2.56	6.38	7.32	4.24	5.08
∑PCBs[c]	72.63	79.62	55.80	54.52	11.37	9.37	16.74	20.26	6.39	7.92	25.01	29.53	19.32	21.35	12.84	7.84	9.20	7.30	5.16	12.64	34.29	37.70	21.27	27.78

[a] Sum of 12 dioxin-like PCBs (4 non-ortho substituted PCBs: PCB77, 81, 126, 169; 8 mono-ortho substituted PCBs: PCB105, 114, 118, 123, 156, 157, 167, 189);

[b] Sum of 6 ICES indicator or marker PCBs (ICES6-PCBs: PCB28, 52, 101, 138, 153, and 180) (European Commission, 2011);

[c] Sum of 209 PCBs;

CX: Cox's Bazar, CT: Chittagong, BH: Bhola, SN: Sundarbans;

The concentrations of congeners with <LOD were assumed to be zero while calculating ∑PCBs.

Table A-11. Concentration of PCB congeners in the seafood (finfish and shellfish) samples (ng/g ww) in summer 2015.

PCB	Ilish				Rupchanda		Loitta				Sole		Poa				Shrimp				Crab			
IUPAC No.	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	BH	SN	CX	CT	BH	SN
1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
10	0.06	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	0.03	0.02	<LOD	0.01	<LOD	<LOD	<LOD	<LOD	0.02	0.02	0.03	<LOD	<LOD
4	<LOD	0.67	<LOD	0.04	0.02	0.02	<LOD	<LOD	0.02	<LOD	0.03	<LOD	<LOD	0.04	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	0.02
9	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	0.02	0.07	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
7	0.23	<LOD	0.05	0.42	<LOD	0.04	<LOD	<LOD	<LOD	0.03	0.13	0.06	0.04	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.05
11	0.47	0.47	0.45	0.55	0.07	<LOD	<LOD	0.07	<LOD	<LOD	0.09	0.08	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.23	0.18	0.06	0.07
6	0.11	0.04	<LOD	0.07	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8+5	1.20	1.17	0.92	1.15	0.29	0.29	0.37	0.26	0.22	0.21	0.95	0.33	0.20	0.32	0.40	0.14	0.07	0.10	0.02	0.11	0.96	0.64	0.31	0.66
13+12	0.85	<LOD	0.04	<LOD	0.09	<LOD	<LOD	<LOD	<LOD	0.05	0.09	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	0.11	<LOD	<LOD
15	<LOD	0.70	0.36	<LOD	0.03	0.08	0.16	<LOD	<LOD	<LOD	<LOD	0.02	0.03	0.02	0.03	<LOD	<LOD	0.02	<LOD	0.06	0.02	0.04	0.02	0.06
19	<LOD	<LOD	0.02	0.02	<LOD	<LOD	<LOD	<LOD	0.01	<LOD	<LOD	0.03	<LOD	0.02	0.02	0.02	<LOD	0.02	0.02	0.02	0.02	0.10	0.02	0.02
30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
18	0.75	0.91	1.13	1.21	0.23	0.28	0.49	0.81	0.39	0.12	0.69	0.78	0.46	0.46	0.38	0.11	0.05	0.08	0.05	0.13	0.97	0.75	0.39	0.49
17	0.30	0.14	0.37	0.50	0.13	0.23	0.38	0.21	0.06	0.08	0.94	<LOD	0.35	0.14	0.11	0.06	0.04	0.14	0.07	0.08	0.36	0.25	0.15	0.11
24	0.06	0.03	0.23	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD
27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD
32	0.12	0.07	0.08	0.05	<LOD	0.02	<LOD	<LOD	0.02	<LOD	0.04	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD
16	0.97	0.46	0.54	1.16	0.04	0.09	0.30	0.06	0.03	0.06	0.31	0.43	0.27	0.26	0.05	0.07	0.07	0.02	0.04	0.02	0.42	0.34	0.18	0.17
23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
34	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.02	<LOD	<LOD
29	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
26	0.04	0.39	<LOD	0.04	0.31	0.40	0.62	0.66	0.21	0.10	0.67	0.99	0.54	0.31	0.30	0.30	0.16	0.09	0.06	0.25	0.54	0.98	0.35	0.32
25	<LOD	<LOD	0.03	<LOD	0.02	0.03	<LOD	0.03	<LOD	<LOD	0.58	0.74	<LOD	<LOD	<LOD	<LOD	0.27	0.26	0.14	0.29	0.06	0.07	<LOD	0.02
31	1.11	0.78	0.78	0.61	0.05	0.18	0.26	0.25	0.03	0.09	0.92	0.43	0.11	0.40	0.06	0.05	0.07	0.02	<LOD	0.02	0.13	0.73	0.11	0.39
28	1.76	1.38	1.27	0.83	0.16	0.24	0.45	0.47	0.32	0.21	0.60	1.09	0.62	0.23	0.11	0.12	0.15	0.04	0.06	0.22	0.78	0.88	0.30	0.31
21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
20+33	0.65	0.31	0.30	0.51	0.07	0.13	<LOD	0.03	0.03	<LOD	<LOD	0.57	0.68	<LOD	<LOD	<LOD	0.09	<LOD	0.03	<LOD	<LOD	<LOD	0.04	0.03
22	<LOD	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.03	<LOD	<LOD	<LOD
36	0.27	0.03	0.44	<LOD	0.02	0.02	0.02	<LOD	<LOD	0.02	0.05	0.05	0.15	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	0.02
39	0.24	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.07	<LOD	<LOD
38	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
35	0.24	<LOD	0.07	0.02	0.03	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.01	<LOD	<LOD	0.02	0.04	<LOD	<LOD
37	0.71	0.11	0.50	0.05	0.12	0.04	0.05	0.42	0.02	0.10	0.24	0.64	0.22	0.04	0.02	0.03	0.02	<LOD	0.02	0.03	0.05	0.22	0.04	0.05
54	0.10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
50	<LOD	1.25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
53	1.48	0.03	0.05	<LOD	0.08	0.02	0.05	<LOD	0.02	<LOD	<LOD	0.02	0.02	<LOD	0.02	<LOD	0.02	<LOD	0.02	0.02	0.03	0.02	<LOD	<LOD
51	<LOD	<LOD	0.02	<LOD	0.02	0.03	<LOD	0.05	<LOD	<LOD	0.04	0.05	<LOD	0.04	0.05	<LOD	<LOD	0.02	0.02	0.02	0.02	0.11	0.02	0.02
45	0.16	0.10	0.06	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.07	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD
52	2.10	2.45	2.26	1.63	0.46	0.54	0.65	0.86	0.44	0.29	0.96	1.20	0.77	1.41	0.62	0.36	0.46	0.40	0.26	0.35	0.58	1.10	0.40	0.59
46+69	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
73	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
43+49	1.82	2.19	1.63	1.36	0.38	0.38	0.79	1.19	0.12	0.09	0.81	1.51	0.43	1.21	0.89	0.29	0.25	0.14	0.11	0.21	0.37	0.92	0.46	0.38
65+75+47+48	0.88	<LOD	<LOD	0.25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
62	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
59	<LOD	0.02	0.04	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD
44	0.67	0.82	0.54	1.74	0.03	0.05	0.36	0.52	0.06	0.17	0.15	0.79	0.02	0.13	0.52	0.03	0.24	0.22	0.07	0.02	0.23	0.07	0.05	0.12
42	<LOD	0.05	0.04	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD
64	<LOD	0.26	0.06	<LOD	0.04	0.04	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD
72	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
71	0.10	0.39	0.36	0.87	0.14	0.07	0.03	<LOD	<LOD	<LOD	0.02	<LOD	0.12	0.17	0.02	0.03	<LOD	<LOD	0.02	<LOD	0.29	0.09	<LOD	<LOD
41	0.93	1.58	0.98	1.47	0.17	0.02	0.49	0.60	0.14	0.07	0.22	0.12	0.27	1.11	0.26	0.23	0.08	0.17	0.08	0.13	0.04	0.17	0.06	0.03
68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
67+57	<LOD	0.25	0.29	<LOD	0.03	0.03	<LOD	0.02	0.02	<LOD	<LOD	0.07	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	0.04	<LOD	0.02	<LOD	<LOD
40	0.91	1.58	1.28	0.47	0.16	0.04	0.02	0.07	0.03	0.02	0.02	0.05	0.02	0.02	0.02	0.03	0.02	0.03	0.02	0.09	0.07	0.05	0.02	0.03
61+63+58	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
74	0.08	0.48	1.01	0.08	0.07	0.06	0.05	0.05	0.03	0.01	0.16	0.30	0.29	0.04	0.02	0.03	0.03	0.08	0.11	0.17	0.15	0.03	0.13	0.20
66	1.52	2.11	1.83	2.05	0.43	0.13	0.94	0.68	0.11	0.32	0.82	1.54	0.44	0.40	0.32	0.29	0.24	0.22	0.16	0.29	0.84	0.94	0.37	0.54
55+76+80	<LOD	<LOD	1.23	1.60	0.59	0.33	0.63	<LOD	0.04	0.39	0.98	0.75	0.69	1.47	1.01	0.37	0.19	0.06	0.08	0.04	0.87	0.87	0.23	0.22
70	1.15	0.90	1.02	0.07	0.08	0.05	0.03	0.05	0.04	0.33	0.95	0.44	0.05	0.44	0.07	0.05	0.06	0.03	0.09	0.25	0.52	0.65	0.09	0.14
60+56	<LOD	0.40	0.08	0.02	0.02	0.03	0.04	0.07	<LOD	0.02	0.09	0.15	0.01	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	0.02
81	1.05	1.24	0.99	0.73	0.04	0.11	0.31	0.24	0.07	0.09	0.13	0.11	0.20	0.16	0.17	0.09	0.18	0.16	0.03	0.11	0.33	0.08	0.05	0.04
77	0.32	0.23	0.18	0.09	<LOD	<LOD	0.15	0.09	<LOD	0.06	0.08	0.06	0.10	0.20	0.05	<LOD	<LOD	0.08	<LOD	<LOD	0.14	0.02	0.02	0.03
79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
78	0.27	0.12	0.39	0.02	0.02	0.02	0.02	0.02	0.02	<LOD	<LOD	0.03	0.03	0.03	0.02	<LOD	<LOD	0.02	<LOD	0.02	0.02	0.10	0.02	0.02
104	0.20	0.02	0.02	0.03	0.02	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.03	0.02	<LOD	<LOD	<LOD
96	0.63	2.50	0.09	0.05	0.52	0.32	0.58	0.88	0.21	<LOD	0.80	1.25	0.46	0.35	0.45	0.24	0.14	0.08	0.11	<LOD	0.89	0.79	0.35	0.64
103	0.12	0.04	0.25	0.14	<LOD	<LOD	0.03	0.03	<LOD	<LOD	0.05	<LOD	0.02	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD
100	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	0.05	0.02	0.02	0.02	0.02	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	0.03	0.03	0.02	0.02	0.02
94	1.65	0.66	0.03	<LOD	<LOD	0.03	0.02	0.03	<LOD	0.03	<LOD	0.05	0.21	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD
102+93+98+95	2.53	3.19	3.11	2.67	0.98	0.37	0.76	1.13	0.40	0.46	1.17	1.48	0.43	1.42	1.28	0.36	0.24	0.08	0.15	0.32	0.85	1.28	0.44	0.79
88	0.17	0.11	0.16	0.02	<LOD	<LOD	0.03	0.03	<LOD	0.02	<LOD	0.05	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

91+121	0.12	0.27	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
92+84	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
89+90	0.14	1.63	1.79	1.21	0.13	<LOD	0.11	0.56	0.08	0.06	0.26	0.14	0.07	0.14	0.22	0.08	0.02	<LOD	<LOD	<LOD	0.09	0.09	<LOD	0.14
101	3.18	2.31	2.96	2.33	0.66	0.29	0.41	0.92	0.19	0.56	0.96	1.39	0.26	1.22	0.40	0.43	0.25	0.26	0.03	0.24	0.34	0.94	0.31	0.46
99	2.75	2.55	1.67	1.97	0.84	0.10	0.34	0.42	0.14	0.48	1.42	1.06	0.98	0.61	0.26	0.21	0.04	0.13	0.09	0.15	0.50	0.63	0.17	0.12
113	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
112+119	0.24	0.15	0.28	0.02	0.03	0.02	0.02	0.02	0.05	0.03	0.02	0.07	0.07	0.05	0.03	0.06	0.02	0.03	0.02	0.05	0.03	0.04	0.02	0.03
83+115	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
86+117+97	0.29	0.28	0.06	<LOD	0.05	0.03	<LOD	0.04	0.03	<LOD	0.07	0.12	0.03	0.07	0.04	0.02	<LOD	<LOD	<LOD	<LOD	0.05	0.11	<LOD	<LOD
125+116+108	0.24	0.22	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
87	1.97	1.51	0.43	0.54	0.07	0.06	0.21	0.16	0.08	0.08	0.52	0.58	0.40	0.40	0.30	0.10	0.28	0.21	0.03	0.05	0.08	0.20	0.07	0.10
111	0.19	1.27	0.66	<LOD	<LOD	<LOD	0.03	0.03	0.02	<LOD	0.07	0.09	<LOD	0.09	0.03	<LOD	<LOD	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
85	<LOD	0.06	0.13	0.04	0.02	<LOD	<LOD	<LOD	<LOD	0.04	0.04	0.03	<LOD	<LOD	<LOD	0.02	0.36	0.03	0.17	0.30	0.05	0.09	0.02	0.02
120+110	2.48	2.11	2.75	1.92	0.55	0.22	0.92	0.95	0.16	0.53	1.45	1.39	0.78	0.80	0.83	0.38	<LOD	<LOD	<LOD	0.11	0.42	0.65	0.15	0.34
82	0.12	0.02	0.04	<LOD	<LOD	0.03	0.03	0.02	0.02	<LOD	0.08	0.04	<LOD	0.02	<LOD	<LOD	0.30	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
106+124	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
109+107	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
123	0.77	1.45	0.58	0.81	0.11	0.14	0.17	0.13	0.09	0.06	0.13	0.77	0.49	0.47	0.11	0.17	0.07	0.04	0.03	0.06	0.31	0.20	0.21	0.11
118	2.57	2.43	1.92	2.10	0.46	0.38	1.13	1.45	0.24	0.25	0.79	1.67	0.70	0.88	0.51	0.43	0.34	0.16	0.14	0.23	0.72	1.18	0.40	0.60
114	0.76	0.60	1.12	0.63	0.21	0.14	0.14	0.73	0.04	0.16	0.59	0.12	0.22	0.12	0.44	0.14	0.05	0.06	0.02	0.11	0.15	0.11	0.22	0.17
122+127	<LOD	<LOD	0.07	0.03	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
105	1.59	0.99	0.42	0.56	0.38	0.34	0.11	0.51	0.04	0.12	1.32	0.91	0.17	0.12	0.26	0.17	0.10	0.23	0.07	0.16	0.63	0.56	0.36	0.33
126	0.07	0.11	0.02	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	0.03	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.12	<LOD	<LOD
155	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.02	<LOD	<LOD	0.02	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.02	<LOD
150	0.07	0.03	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	0.03	0.03
152	0.66	0.07	0.10	0.02	0.06	0.05	0.03	0.04	0.02	<LOD	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD
145	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
136+148	0.23	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	0.09	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
154	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD
149+139	2.36	3.05	2.65	2.36	0.82	0.33	0.35	0.68	0.30	0.56	1.55	1.26	0.93	1.07	1.11	0.49	0.37	<LOD	<LOD	0.30	0.40	1.46	0.78	1.18
135	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
144+147	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
151	1.25	0.03	0.12	2.93	0.62	0.03	0.11	0.41	0.02	0.03	0.10	1.45	1.03	0.84	0.53	0.19	0.33	0.19	0.09	0.39	0.70	0.46	0.33	0.50
140	<LOD	0.24	0.16	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	0.06	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.48	0.14	<LOD	<LOD
143	0.22	0.03	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02
134	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
142+131+133	0.50	0.30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
165	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
146	0.22	0.14	0.03	<LOD	<LOD	<LOD	<LOD	0.12	<LOD	<LOD	0.02	<LOD	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	0.02	<LOD	0.06
132+161	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
137	<LOD	0.05	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
168	1.22	2.47	0.30	0.08	0.19	0.07	0.18	0.15	0.03	0.03	0.13	0.17	0.23	0.10	0.04	0.08	0.07	0.09	0.02	0.08	0.10	0.19	0.09	0.16
141	0.10	0.05	0.86	0.87	0.05	0.14	0.49	0.08	0.10	0.09	0.98	0.65	0.60	0.10	<LOD	0.13	0.11	<LOD	<LOD	0.06	0.22	0.23	0.22	0.17
153	2.77	3.91	3.36	2.44	1.34	0.64	0.97	0.86	0.53	0.40	1.87	1.75	1.37	1.49	1.36	0.55	0.13	0.18	0.10	0.32	0.74	1.08	0.58	0.86
130	<LOD	<LOD	0.02	0.04	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	0.05	0.05	0.03	0.04	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.07	0.05	0.03
158	0.45	0.16	0.22	0.06	0.16	0.10	0.15	0.14	0.02	0.03	0.55	0.14	0.02	0.07	0.03	0.09	0.02	0.02	0.04	0.08	0.06	0.04	0.11	0.13
160+164+163	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
138	2.47	3.75	2.04	2.56	1.15	0.48	0.50	1.20	0.26	0.74	1.36	1.54	1.14	1.10	0.77	0.18	0.11	0.21	0.22	0.38	1.05	1.17	0.93	1.25
169	0.05	0.03	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.12	0.09	<LOD	<LOD
157	0.34	1.39	1.78	1.08	<LOD	0.03	0.25	0.16	0.02	0.04	0.10	0.11	0.13	0.14	0.17	0.09	0.05	0.05	0.03	0.11	0.14	0.18	0.05	0.03
128	1.22	2.07	0.93	0.93	0.53	0.26	0.40	0.81	0.35	0.20	0.77	0.48	0.26	0.26	0.39	0.12	0.19	0.07	0.13	0.15	0.64	0.64	0.51	0.71
159+162	0.08	0.09	0.20	0.12	0.06	<LOD	0.02	<LOD	<LOD	0.02	0.05	0.06	0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD
167	1.52	2.24	0.84	1.21	0.10	0.14	0.14	0.15	0.08	0.08	0.08	0.05	0.09	0.08	0.16	0.07	0.08	0.03	0.04	0.23	0.14	0.16	0.26	0.11
156	1.16	1.90	1.28	0.79	0.12	0.10	0.09	0.12	0.06	0.12	0.16	0.19	0.32	0.31	0.09	0.09	0.07	0.05	0.02	0.10	0.30	0.14	0.17	0.14
166	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
129	0.11	0.05	0.05	0.34	<LOD	0.04	<LOD	<LOD	0.01	<LOD	0.02	0.03	0.08	0.03	0.02	<LOD	<LOD	<LOD	<LOD	0.06	<LOD	0.04	<LOD	<LOD
188	0.07	0.02	<LOD	0.11	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02
184	0.15	0.04	<LOD	<LOD	0.04	<LOD	0.06	<LOD	0.02	0.02	0.10	0.03	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.04	0.02	0.03
179	<LOD	<LOD	0.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
176	<LOD	<LOD	0.08	0.03	<LOD	0.02	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
187	2.16	2.81	3.18	2.26	0.13	0.10	0.59	0.75	0.32	0.49	0.73	1.28	0.88	0.92	0.48	0.39	0.21	0.26	0.15	0.29	0.72	0.79	0.04	1.24
178	<LOD	0.36	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD
175	<LOD	0.05	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	0.06
182+186	0.55	<LOD	<LOD	<LOD	0.07	0.05	0.11	0.05	0.03	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
183	0.92	0.28	0.54	0.40	0.15	0.03	0.12	0.09	0.05	0.05	0.10	0.13	0.34	0.09	0.05	0.02	0.02	0.02	0.03	0.02	0.05	0.24	0.03	0.19
174	1.51	0.94	0.50	0.49	0.26	0.12	0.05	0.19	0.02	0.04	0.22	0.32	0.39	0.10	0.16	0.04	0.06	0.07	0.04	0.07	0.46	0.43	0.14	0.63
185	<LOD	0.05	0.05	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.03	0.04
181	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
171	0.50	1.34	0.60	1.11	0.11	0.04	0.03	0.46	0.05	0.16	0.71	0.29	0.06	0.23	0.11	0.16	<LOD	<LOD	<LOD	0.22	0.24	0.16	0.15	0.25
177	1.61	0.81	1.59	0.69	0.62	0.23	0.25	0.20	0.13	0.09	1.01	0.94	0.49	0.83	0.24	0.20	0.16	0.24	0.06	0.19	0.76	0.51	0.23	0.54
173	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
189	0.66	0.58	0.39	0.46	0.09	0.05	0.05	0.04	0.03	0.06	0.13	0.13	0.13	0.17	0.08	0.07	0.08	0.06	0.03	0.09	0.23	0.28	0.14	0.17
180	2.41	3.34	2.78	1.35	0.63	0.33	0.64	0.93	0.36	0.88	1.25	1.58	0.67	0.96	0.39	0.37	0.22	0.20	0.20	0.23	1.10	1.68	1.10	1.63
192	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
170	1.36	2.70	2.24	2.50	0.46	0.28	0.32	0.69	0.21	0.59	0.96	1.06	0.57	0.81	0.59	0.15	0.13	0.13	0.09	0.16	0.84	0.97	0.91	1.17
191	<LOD	0.04	<LOD	0.04	<LOD	<LOD	<LOD	0.08	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.01	<LOD	<LOD	0.07	0.14	<LOD	<LOD
193	0.13	0.05	0.09	0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.01	<LOD	<LOD	<LOD	<LOD	0.02
190	<LOD	0.10	0.02	<LOD	<LOD	0.02	0.02	<LOD	<LOD	0.02	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	0.04	0.04
172	0.05	0.24	<LOD	0.26	<LOD	0.05	0.05	<LOD	<LOD	0.04	0.06	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.06	<LOD
202	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.28	<LOD

204	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
201	1.18	0.93	0.81	0.63	0.13	0.15	0.35	0.23	0.13	0.19	0.41	0.50	0.55	0.44	0.33	0.12	0.07	0.02	0.02	0.14	0.36	0.29	<LOD	0.35
195	0.08	0.09	0.14	0.03	<LOD	<LOD	<LOD	0.03	<LOD	<LOD	0.58	0.09	<LOD	<LOD	0.09	0.02	0.02	<LOD	<LOD	<LOD	0.02	0.08	<LOD	0.02
200	<LOD	<LOD	0.06	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	0.05
198+199	0.23	0.26	0.17	<LOD	<LOD	<LOD	0.05	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.09	0.17	0.03	<LOD
196+203	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.12	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
197	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	0.04	<LOD	<LOD	<LOD	<LOD
194	0.72	<LOD	<LOD	0.17	<LOD	<LOD	0.16	<LOD	0.03	0.03	<LOD	<LOD	<LOD	0.20	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.18	<LOD	0.16	<LOD
205	0.14	0.07	<LOD	<LOD	<LOD	0.07	<LOD	0.05	<LOD	<LOD	0.07	0.13	<LOD	0.07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.02	0.09	<LOD	<LOD
208	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
207	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
206	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
209	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Mono-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Di-CB	2.93	3.12	1.83	2.22	0.48	0.42	0.55	0.44	0.24	0.30	1.30	0.52	0.29	0.47	0.47	0.21	0.07	0.12	0.02	0.19	1.26	1.02	0.40	0.86
Tri-CB	7.22	4.66	5.83	5.00	1.19	1.66	2.59	2.95	1.12	0.79	5.07	5.80	3.44	1.91	1.04	0.78	0.92	0.68	0.49	1.10	3.43	4.52	1.61	1.94
Tetra-CB	13.54	16.46	14.33	12.51	2.84	1.94	4.61	4.51	1.15	1.85	5.41	7.18	3.59	6.96	4.08	1.82	1.77	1.67	1.13	1.76	4.55	5.30	1.94	2.38
Penta-CB	22.89	24.57	18.57	15.29	5.03	2.49	5.05	8.16	1.81	2.90	9.79	11.33	5.30	6.90	5.25	2.81	2.27	1.45	0.87	1.86	5.20	7.04	2.73	3.86
Hexa-CB	17.14	22.26	15.11	15.88	5.23	2.43	3.74	5.10	1.85	2.50	7.91	8.14	6.60	5.64	4.71	2.07	1.52	0.90	0.68	2.24	5.21	6.21	4.17	5.36
Hepta-CB	12.09	13.77	12.25	9.91	2.56	1.31	2.28	3.50	1.22	2.48	5.32	5.95	3.61	4.14	2.11	1.39	0.87	1.00	0.62	1.27	4.54	5.36	2.88	6.04
Octa-CB	2.36	1.35	1.17	0.84	0.13	0.24	0.56	0.32	0.16	0.28	1.06	0.72	0.72	0.78	0.44	0.14	0.09	0.02	0.02	0.20	0.67	0.62	0.47	0.43
Nona-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Deca-CB	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑DL-PCBs	10.86	13.20	9.52	8.45	1.55	1.43	2.53	3.66	0.68	1.03	3.55	4.18	2.56	2.69	2.04	1.32	1.01	0.93	0.42	1.21	3.24	3.12	1.87	1.73
∑iPCBs	14.70	17.15	14.67	11.15	4.41	2.52	3.63	5.24	2.11	3.08	7.00	8.54	4.84	6.41	3.63	1.99	1.31	1.30	0.87	1.74	4.59	6.86	3.61	5.09
∑PCBs	78.16	86.18	69.10	61.65	17.47	10.50	19.39	24.98	7.55	11.11	35.85	39.65	23.54	26.78	18.11	9.21	7.51	5.85	3.82	8.63	24.85	30.06	14.20	20.87

Table A-12. The estimated daily intake (EDI) of ∑PCBs and ∑DL-PCBs via seafood consumption for the coastal residents (adults and children) of Bangladesh.

Location	Type	Seafood consumption[a] (g/day)		$\mathrm{EDI}_{\sum \mathrm{PCBs}}$ (ng/kg–bw/day)		$\mathrm{EDI}_{\sum \mathrm{DL\text{-}PCBs}}$ (pg TEQ/kg–bw/day)	
		Adult[b]	Children[b]	Adult	Children	Adult	Children
Cox's Bazar	Finfish	86.5	40	41.22	45.78	3.46	3.85
	Shellfish	20	12.5	6.38	9.44	2.39	3.41
	$\mathrm{EDI}_{(Finfish+Shellfish)}$			47.59	55.22	5.85	7.26
Chittagong	Finfish	86	43	41.66	50.13	6.56	7.91
	Shellfish	23	14.5	7.79	11.69	3.89	5.77
	$\mathrm{EDI}_{(Finfish+Shellfish)}$			49.46	61.82	10.45	13.68
Bhola	Finfish	68.5	32	38.88	43.63	1.06	1.19
	Shellfish	17	11.5	3.18	4.99	0.27	0.37
	$\mathrm{EDI}_{(Finfish+Shellfish)}$			42.06	48.62	1.34	1.56
Sundarbans	Finfish	70	34	40.80	47.62	0.85	0.98
	Shellfish	20	11	5.78	7.69	1.52	2.06
	$\mathrm{EDI}_{(Finfish+Shellfish)}$			46.58	55.31	2.37	3.04

[a] Data obtained from dietary questionnaire survey; [b] Average body weight was 60 kg for adults and 25 kg for children.

Reference

European Commission. 2011. Commission Regulation (EU) No 1259/2011 of 2 December 2011 amending regulation (EC) no. 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non-dioxin-like PCBs in foodstuffs. Off J Eur Union, 320, 18–23.

APPENDIX B

Table B-1. Analytes, acronyms, supplier, purity, retention time, MS/MS transition, and corresponding internal standards.

Classification	Compound name	Acronym	Supplier of standard and its purity	Retention time (min)	MS/MS transition (m/z)	Corresponding internal standards
PFCAs (perfluoroalkyl carboxylic acids)	Perfluorobutanoate	PFBA	PFC-MXA (Mix) Wellington Laboratories, 98%	6.17	212.7 → 168.7	[^{13}C4]PFBA
	Perfluoropentanoate	PFPeA		13.06	262.7 → 218.7	[^{13}C2]PFHxA
	Perfluorohexanoate	PFHxA		14.11	312.7 → 268.7	[^{13}C2]PFHxA
	Perfluoroheptanoate	PFHpA		15.62	362.7 → 318.7	[^{13}C2]PFHxA
	Perfluorooctanoate	PFOA		17.59	412.7 → 368.7	[^{13}C4]PFOA
	Perfluorononanoate	PFNA		21.23	462.7 → 418.7	[^{13}C4]PFOA
	Perfluorodecanoate	PFDA		25.77	512.7 → 468.7	[^{13}C4]PFOA
	Perfluoroundecanoate	PFUnDA		32.57	562.7 → 518.7	[^{13}C2]PFDoDA
	Perfluorododecanoate	PFDoDA		36.49	612.7 → 568.7	[^{13}C2]PFDoDA
	Perfluorotridecanoate	PFTrDA		37.20	662.7 → 618.7	[^{13}C2]PFDoDA
	Perfluorotetradecanoate	PFTeDA		37.40	712.7 → 668.7	[^{13}C2]PFDoDA
PFSAs (perfluoroalkyl sulfonic acids)	Perfluorobutane sulfonate	PFBS	Tokyo Chemical Ind., 98%	13.34	298.7 → 98.7	[^{13}C4]PFOS
	Perfluorohexane sulfonate	PFHxS	Wellington Laboratories, 98%	21.40	398.7 → 98.7	[^{13}C4]PFOS
	Perfluorooctane sulfonate	PFOS	Kanto Chemical, 98%	15.58	498.7 → 98.7	[^{13}C4]PFOS
	Perfluorodecane sulfonate	PFDS	Wellington Laboratories, 98%	32.30	598.7 → 98.7	[^{13}C4]PFOS
Internal Standard (IS)	Perfluoro-n-[1,2,3,4-^{13}C2]butanoate	[^{13}C4]PFBA	Wellington Laboratories, 98%	6.26	216.7 → 171.7	–
	Perfluoro-n-[1,2-^{13}C2]hexanoate	[^{13}C2]PFHxA	Wellington Laboratories, 98%	14.12	314.7 → 269.6	–
	Perfluoro-n-[1,2,3,4-^{13}C4]octanoate	[^{13}C4]PFOA	Wellington Laboratories, 98%	17.66	416.7 → 371.6	–
	Perfluoro-n-[1,2-^{13}C2]dodecanoate	[^{13}C2]PFDoDA	Wellington Laboratories, 98%	36.53	614.6 → 569.6	–
	perfluoro-1-[1,2,3,4-^{13}C4]octanesulfonate	[^{13}C4]PFOS	Wellington Laboratories, 98%	21.30	502.7 → 98.7	–

Table B-2. Instrumental characteristics used for analytical determination of PFAAs.

LC conditions	
Instrument	HP1100 (Agilent)
Analytical column	Zorbax XDB C-18 (150 mm × 2.1 mm, aperture size 5 μm) (Agilent Tech., Santa Clara, USA)
Mobile phase A	10 mM Ammonium acetate aq.
Mobile phase B	Methanol
Flow rate	0.2 mL/min
Gradient	Time (min) 0 5 15 25 31 42 44.5 B (%) 10 62 65 70 100 70 10
Column Temp.	40 °C
Injection Volume	10 μL
Triple quadrupole MS/MS conditions	
Instrument	Micromass Quattro Ultima (Waters, Milford, USA)
Ionization	Electrospray ionization (ESI) in negative mode
Gas temperature	350 °C
Gas flow (N_2)	Cone: 50 L hr^{-1}; Dissolvation: 500 L hr^{-1}
Capillary voltage	2.70 kV
Scan type	MRM

Table B-3. Recoveries*, LODs and LOQs for individual PFAAs in surface water and sediment.

PFAAs	% Recoveries [mean (RSD)]		LOD[a]		LOQ[b]		Linearity (r^2)[c]
	Water 10 ng/L (n=3)	Sediment 10 ng/g (n=3)	Water (ng/L)	Sediment (ng/g)	Water (ng/L)	Sediment (ng/g)	
PFBA	113 (6)	87 (1)	0.008	0.031	0.02	0.102	0.999
PFPeA	114 (3)	97 (5)	0.012	0.003	0.032	0.011	0.997
PFHxA	99 (5)	90 (4)	0.008	0.004	0.016	0.013	0.998
PFHpA	102 (6)	99 (4)	0.008	0.004	0.036	0.013	0.998
PFOA	94 (6)	102 (2)	0.120	0.023	0.40	0.076	0.999
PFNA	85 (5)	93 (6)	0.016	0.004	0.04	0.012	0.998
PFDA	83 (4)	95 (2)	0.008	0.002	0.036	0.006	0.999
PFUnDA	111 (6)	106 (3)	0.036	0.009	0.08	0.029	0.999
PFDoDA	93 (6)	107 (5)	0.040	0.003	0.08	0.011	0.999
PFTrDA	102 (2)	98 (5)	0.024	0.003	0.04	0.008	0.998
PFTeDA	95 (9)	103 (3)	0.024	0.002	0.04	0.007	0.999
PFBS	112 (3)	108 (5)	0.080	0.018	0.28	0.06	0.999
PFHxS	87 (4)	94 (5)	0.040	0.012	0.20	0.021	0.999
PFOS	104 (4)	98 (2)	0.080	0.026	0.10	0.088	0.999
PFDS	74 (2)	101 (5)	0.030	0.016	0.10	0.024	0.999

[a] Limit of detection;

[b] Limit of quantification;

[c] Calibration curves (0.01–50 µg/L for each compound);

* The recovery of spiked PFAAs was calculated using the following equation:

spike recovery rate (%) = ($C_{sample + spiked} - C_{sample}$) / C_{spiked} × 100,

where $C_{sample + spiked}$ is the concentration of PFAAs in a spiked sample, C_{sample} is the concentration of PFAAs in the sample (same as above without spiking target compounds, C_{spiked} is the concentration of the spiked target PFAAs.

Table B-4. Physical and chemical characteristics of surface water and sediment collected from the coastal area of Bangladesh.

Season	Site	Water				Sediment							
		pH	Temper-ature (°C)	Salinity (‰)	TSS (mg/L)	pH	TOC (%)	BC (%)	TN (%)	Clay (%)	Silt (%)	Sand (%)	Texture*
Winter	CX1	7.3	19.5	17.3	465	7.1	1.28	0.21	0.3	10.6	18.1	71.2	Sandy Loam
	CX2	6.2	20	18.4	340	6.1	0.66	0.14	0.1	9.1	11.7	79.2	Loamy Fine Sand
	CX3	7.2	20.5	19.6	490	6.9	0.94	0.33	0.1	22.2	34.3	43.5	Loam
	CX4	7.1	20.5	18.6	435	6.5	1.27	0.15	0.2	25.5	27.1	47.4	Sandy Clay Loam
	CT1	6.6	19.5	21.3	280	6.5	0.98	0.39	0.1	13.3	21.4	65.3	Sandy Loam
	CT2	6.3	21	19.5	310	6.1	0.4	0.13	0.1	15.8	70.6	13.5	Silt Loam
	CT3	6.7	22	17.4	230	5.9	0.55	0.14	0.1	44.9	25.4	29.7	Clay
	CT4	6.5	21.5	18.6	260	6.3	1.05	0.53	0.1	13.9	19.7	66.4	Sandy Loam
	ME1	7.7	19	16.8	800	7.5	0.31	0.15	0	2.8	70.8	26.4	Silt Loam
	ME2	7.1	19.5	20.4	650	6.9	0.07	0.06	0	4.2	64.5	31.3	Silt Loam
	ME3	7.8	22	21.6	460	7.3	0.06	0.06	0	6.2	65.4	28.4	Silt Loam
	SN1	6.9	19	17.3	850	6.7	0.39	0.13	0	20	68.8	11.3	Silt Loam
	SN2	7.6	21	18.6	730	7.3	0.24	0.07	0	17.2	73.5	9.3	Silt Loam
	SN3	6.9	23	19.8	960	7.2	0.15	0.06	0	13.5	71.3	15.2	Silt Loam
Summer	CX1	7.2	24.5	22.5	500	7.3	0.69	0.11	0.2	13.4	16.4	70.2	Sandy Loam
	CX2	6.5	23.3	24	350	6.1	0.64	0.13	0.1	7.6	15.8	76.5	Sandy Loam
	CX3	6.8	24.1	12.5	480	6.7	0.49	0.12	0	23.2	32.3	44.5	Loam
	CX4	7.1	23.5	16.5	460	6.6	0.55	0.15	0.1	23.2	31.3	45.5	Loam
	CT1	6.5	22.3	13.5	350	6.3	0.49	0.19	0	5.9	27.4	66.6	Sandy Loam
	CT2	6.2	23.8	15.5	440	6.3	0.43	0.13	0.1	17.8	38.4	43.8	Loam
	CT3	6.3	23.6	17	250	6.1	0.62	0.28	0.1	44.1	28.4	27.6	Clay
	CT4	6.5	23.5	18.5	290	6.3	0.53	0.27	0	20.8	21.3	57.9	Sandy Clay Loam
	ME1	7.5	22	4.5	850	7.5	0.22	0.07	0	10.1	61.7	28.3	Silt Loam
	ME2	6.8	24.3	3.5	630	6.8	0.19	0.06	0	8.3	65.3	26.4	Silt Loam
	ME3	7.3	23.6	7	520	7.3	0.15	0.06	0	6.2	66.3	27.5	Silt Loam
	SN1	6.5	23.5	13.5	900	6.6	0.49	0.28	0.1	18	68.3	13.7	Silt Loam
	SN2	6.2	24.4	14	750	6.7	0.31	0.16	0.1	11.5	77.3	11.3	Silt Loam
	SN3	5.5	24.9	16.5	850	5.9	0.37	0.19	0.1	11.8	69.6	18.6	Silt Loam

*According to the United States Department of Agriculture (USDA) soil texture classification.

Table B-5. Concentrations of individual PFAAs in surface water samples (ng/L) collected from the coastal area of Bangladesh.

Season	Site	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFCAs	∑PFSAs	∑PFAAs
Winter	CX1	0.88	8.07	2.03	<LOD	3.93	0.83	0.37	0.74	0.36	0.15	0.08	3.67	1.43	1.04	0.61	17.5	6.75	24.2
	CX2	<LOD	5.76	1.17	0.26	16.5	0.86	0.67	0.39	0.63	0.08	0.14	2.77	1.81	0.89	<LOQ	26.4	5.46	31.9
	CX3	0.45	1.09	4.07	1.03	24.8	1.40	5.72	0.61	3.30	0.20	1.66	0.34	<LOD	0.16	0.37	44.3	0.87	45.2
	CX4	<LOQ	2.71	0.66	0.17	8.23	1.17	0.28	0.31	<LOQ	0.11	0.12	0.71	<LOD	4.26	0.29	13.8	5.25	19.0
	CT1	1.20	1.56	1.40	0.60	9.39	1.16	1.14	0.68	0.96	0.45	0.46	1.83	0.54	0.41	0.28	19.0	3.07	22.1
	CT2	0.47	1.59	0.86	0.14	10.9	1.15	0.52	0.17	0.17	0.07	0.11	0.99	<LOQ	0.46	0.17	16.1	1.62	17.7
	CT3	1.00	1.25	0.25	0.07	11.4	1.49	0.24	0.35	<LOD	0.05	0.13	0.69	<LOD	0.47	0.17	16.2	1.33	17.5
	CT4	<LOQ	1.03	1.12	0.08	11.3	0.75	0.77	<LOQ	0.21	0.69	0.44	<LOD	<LOQ	0.79	<LOD	16.4	0.79	17.2
	ME1	0.19	0.47	0.16	<LOQ	14.1	1.48	0.25	0.35	<LOD	0.53	0.13	0.36	<LOD	0.24	0.19	17.7	0.78	18.5
	ME2	0.31	0.79	0.22	0.08	12.7	0.57	0.12	0.07	<LOD	<LOD	0.06	0.45	<LOD	0.28	<LOD	14.9	0.73	15.6
	ME3	0.34	0.63	0.67	0.18	8.41	1.11	0.81	0.39	0.48	<LOD	0.26	0.60	<LOD	0.21	0.25	13.3	1.06	14.4
	SN1	<LOD	2.66	0.44	0.31	13.0	1.04	0.20	0.47	<LOQ	0.40	0.20	3.09	<LOD	2.01	<LOD	18.7	5.11	23.8
	SN2	1.10	2.01	0.41	<LOD	3.17	0.84	0.32	0.22	<LOQ	0.08	0.06	1.02	<LOD	1.40	<LOD	8.23	2.42	10.6
	SN3	0.79	1.36	0.22	0.08	14.6	2.08	0.13	0.26	<LOQ	<LOQ	<LOQ	2.38	<LOQ	1.59	<LOD	19.5	3.97	23.5
Summer	CX1	0.07	1.27	0.49	1.35	3.98	1.63	1.54	0.11	0.75	0.57	0.75	<LOD	1.40	0.59	0.52	12.5	2.51	15.0
	CX2	<LOD	2.26	1.29	1.32	5.79	1.07	2.48	<LOQ	1.02	0.26	0.85	<LOQ	0.83	<LOD	<LOD	16.3	0.83	17.2
	CX3	1.17	2.73	0.60	0.94	27.8	1.27	<LOQ	<LOQ	1.34	0.49	0.46	1.44	2.59	5.10	0.86	36.8	9.99	46.8
	CX4	0.86	3.24	1.22	5.12	25.4	0.46	1.53	0.28	0.55	0.37	0.27	0.39	0.00	1.70	0.84	39.3	2.93	42.2
	CT1	<LOD	2.38	0.61	0.70	27.3	1.70	1.55	0.11	0.62	0.42	0.59	0.43	1.93	1.74	<LOQ	36.0	4.09	40.1
	CT2	1.82	2.16	1.13	0.96	12.2	1.67	5.32	<LOD	0.52	0.54	0.36	1.06	0.79	1.70	0.39	26.7	3.95	30.7
	CT3	1.10	1.60	0.83	1.04	15.3	4.17	3.59	0.26	0.57	0.46	0.36	0.65	<LOD	4.29	0.66	29.3	5.60	34.9
	CT4	0.41	1.64	0.87	0.60	12.6	1.37	1.40	0.31	0.83	0.62	0.65	1.00	<LOQ	0.71	<LOD	21.3	1.71	23.0
	ME1	0.31	1.51	0.55	0.14	8.30	0.96	0.67	<LOD	0.17	<LOD	<LOD	1.50	<LOD	<LOD	<LOD	12.6	1.50	14.1
	ME2	<LOD	1.94	0.93	<LOQ	5.92	0.76	0.85	<LOD	0.06	0.31	<LOD	<LOD	<LOD	0.51	0.19	10.8	0.70	11.5
	ME3	<LOD	1.27	0.86	0.24	7.94	1.00	0.55	<LOD	<LOD	<LOD	0.18	<LOQ	0.45	1.25	<LOD	12.0	1.70	13.7
	SN1	0.24	0.69	0.65	0.21	7.93	1.08	1.61	0.14	0.30	0.09	0.07	0.31	0.15	1.19	<LOQ	13.0	1.65	14.7
	SN2	0.25	1.03	0.67	0.44	8.71	1.51	1.32	0.24	0.63	0.37	0.15	<LOD	<LOD	0.87	<LOQ	15.3	0.87	16.2
	SN3	<LOD	0.92	1.48	0.78	4.67	3.03	2.01	0.70	0.53	0.00	0.56	<LOD	<LOQ	0.69	0.09	14.8	0.78	15.5

Note: Values <LOD were considered as ‘0’ while calculating ∑PFCAs, ∑PFSAs, and ∑PFAAs.

Table B-6. Concentration of individual PFAAs in sediment samples (ng/g dw) collected from the coastal area of Bangladesh.

Season	Site	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFCAs	∑PFSAs	∑PFAAs
Winter	CX1	1.00	1.01	0.41	0.38	0.82	0.09	0.12	0.31	0.17	0.04	<LOQ	0.87	0.17	1.25	0.43	4.36	2.71	7.08
	CX2	0.46	0.79	0.21	0.21	0.14	0.17	0.14	0.19	0.17	0.08	<LOD	0.79	0.96	1.13	0.58	2.54	3.46	6.01
	CX3	0.19	0.26	0.51	0.19	0.45	0.44	1.00	0.18	0.49	0.09	0.17	0.59	0.29	1.19	<LOD	3.97	2.07	6.04
	CX4	1.37	0.29	0.15	0.07	0.69	0.23	0.12	0.55	0.23	0.05	0.22	0.68	0.38	1.82	0.38	3.97	3.27	7.24
	CT1	0.50	0.17	0.18	0.24	1.35	0.32	0.23	0.29	0.20	0.12	0.17	0.59	0.68	0.97	0.76	3.79	3.00	6.79
	CT2	0.34	0.09	0.14	0.08	1.42	0.14	0.14	0.16	0.04	<LOQ	0.10	0.55	0.09	0.68	<LOD	2.66	1.32	3.98
	CT3	0.40	0.12	0.11	0.20	0.78	0.53	0.22	<LOD	0.27	0.02	0.10	0.36	<LOD	0.75	0.99	2.74	2.09	4.83
	CT4	0.27	0.32	0.16	0.02	1.49	0.43	0.24	0.53	0.13	0.01	0.23	0.75	<LOD	3.56	<LOD	3.84	4.31	8.15
	ME1	0.22	0.15	0.09	0.04	0.99	0.10	0.08	0.28	0.03	0.02	0.15	0.43	0.02	0.92	<LOD	2.13	1.37	3.50
	ME2	0.80	0.10	0.07	0.03	0.88	0.04	0.04	<LOD	<LOQ	<LOQ	0.01	<LOD	<LOD	0.86	<LOD	1.97	0.86	2.83
	ME3	<LOQ	0.16	0.13	0.03	0.47	0.06	0.05	<LOD	0.10	0.01	<LOD	0.44	0.02	0.95	0.06	1.01	1.47	2.48
	SN1	0.53	0.30	0.10	0.08	0.71	0.16	0.11	0.43	0.03	0.04	0.13	0.49	<LOD	0.44	<LOD	2.63	0.93	3.56
	SN2	0.36	0.25	0.09	0.02	0.71	0.04	0.06	0.22	<LOQ	<LOD	<LOQ	0.44	0.14	0.78	<LOD	1.75	1.36	3.10
	SN3	0.20	0.16	0.07	<LOQ	0.59	0.04	<LOD	0.33	<LOD	0.02	<LOD	0.44	<LOD	0.73	0.27	1.41	1.44	2.85
Summer	CX1	0.19	0.09	0.10	0.09	0.27	0.05	0.08	0.06	0.08	0.08	0.10	<LOD	0.11	0.59	<LOD	1.21	0.70	1.91
	CX2	<LOD	0.13	0.17	0.10	0.69	0.17	0.14	<LOD	0.13	0.08	<LOD	0.10	0.06	0.42	<LOD	1.61	0.58	2.19
	CX3	0.47	0.12	0.08	0.51	0.29	0.10	0.05	0.03	0.05	0.03	0.04	0.19	<LOD	1.64	0.21	1.77	2.04	3.81
	CX4	<LOQ	0.16	0.13	<LOD	0.25	0.12	0.06	<LOQ	0.06	0.07	0.04	0.35	<LOD	0.53	0.29	0.89	1.17	2.06
	CT1	0.14	0.13	0.10	<LOD	0.36	0.13	0.05	0.02	0.07	0.03	0.03	0.23	0.03	0.49	0.18	1.06	0.94	2.00
	CT2	0.17	0.11	0.16	0.05	0.26	0.11	0.06	<LOQ	0.08	0.04	0.04	0.22	<LOD	0.46	<LOD	1.08	0.68	1.76
	CT3	0.33	0.17	0.08	0.07	0.30	0.21	0.11	0.02	0.08	0.04	0.04	0.21	<LOD	0.56	<LOD	1.47	0.77	2.23
	CT4	0.55	0.17	0.10	0.03	0.20	0.11	0.07	0.09	0.09	0.09	0.07	0.15	<LOD	1.01	0.12	1.56	1.28	2.84
	ME1	<LOD	0.09	0.11	0.03	0.30	0.11	0.05	0.11	0.07	0.04	0.02	0.18	0.03	0.31	<LOD	0.93	0.52	1.46
	ME2	0.14	0.10	0.12	0.02	0.24	0.11	0.05	0.47	0.07	0.04	0.02	0.11	<LOD	0.34	<LOD	1.38	0.45	1.84
	ME3	<LOD	0.13	0.11	0.02	0.10	0.10	0.04	<LOD	0.07	0.07	<LOD	<LOD	0.03	0.35	0.06	0.64	0.43	1.07
	SN1	0.26	0.12	0.09	0.02	0.09	0.09	0.03	0.08	0.01	<LOD	<LOD	0.18	<LOD	0.30	<LOD	0.78	0.48	1.26
	SN2	0.23	0.09	0.11	0.02	0.27	0.09	0.08	<LOD	0.02	<LOD	0.03	<LOD	0.04	0.85	<LOD	0.93	0.90	1.83
	SN3	<LOD	0.06	0.13	0.04	0.64	0.11	0.16	0.02	0.12	0.02	0.05	0.17	<LOD	0.57	0.03	1.33	0.77	2.10

Note: Values <LOD were considered as ‘0’ while calculating ∑PFCAs, ∑PFSAs, and ∑PFAAs.

Table B-7. Test of statistical significance by one way ANOVA (p <0.05) for seasonal variation (winter Vs summer) among the individual PFAAs in the investigated surface water and sediment samples.

PFAAs	**Water**		**Sediment**	
	P-value	**Significant**	**P-value**	**Significant**
PFBA	0.850	No	0.011	**Yes**
PFPeA	0.459	No	0.021	**Yes**
PFHxA	0.709	No	0.104	No
PFHpA	0.034	**Yes**	0.343	No
PFOA	0.757	No	0.0001	**Yes**
PFNA	0.151	No	0.077	No
PFDA	0.093	No	0.118	No
PFUnDA	0.024	**Yes**	0.004	**Yes**
PFDoDA	0.659	No	0.110	No
PFTrDA	0.161	No	0.458	No
PFTeDA	0.461	No	0.025	Yes
PFBS	0.022	**Yes**	< 0.0001	**Yes**
PFHxS	0.279	No	0.036	**Yes**
PFOS	0.376	No	0.024	**Yes**
PFDS	0.351	No	0.055	No

Table B-8. Spearman rank correlations among the individual PFAAs and some physical chemical properties [pH, temperature (T), salinity, total suspended solids (TSS)] of surface water samples (*n=14*).

Seasons	Variables	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	pH	T	Salinity	TSS
Winter	PFBA	**1**																		
	PFPeA	0.00	**1**																	
	PFHxA	0.06	0.39	**1**																
	PFHpA	-0.32	0.02	0.48	**1**															
	PFOA	-0.41	-0.28	-0.12	0.38	**1**														
	PFNA	0.23	-0.19	-0.27	0.10	0.35	**1**													
	PFDA	0.13	-0.01	**0.83**	0.47	-0.12	-0.08	**1**												
	PFUnDA	0.16	0.35	0.53	0.37	0.07	0.20	0.37	**1**											
	PFDoDA	0.07	0.23	**0.90**	0.59	0.02	-0.13	**0.89**	0.53	**1**										
	PFTrDA	-0.17	0.07	0.32	0.10	0.04	-0.04	0.33	0.24	0.22	**1**									
	PFTeDA	-0.22	-0.22	0.57	**0.68**	0.23	0.07	**0.74**	0.44	0.62	0.55	**1**								
	PFBS	0.15	**0.82**	0.13	-0.04	-0.18	-0.07	-0.20	0.46	0.13	-0.11	-0.33	**1**							
	PFHxS	0.11	0.48	0.52	0.02	0.00	-0.21	0.33	0.23	0.52	0.14	0.00	0.50	**1**						
	PFOS	-0.16	**0.73**	-0.16	-0.24	-0.26	-0.08	-0.45	-0.09	-0.25	0.06	-0.44	0.63	0.18	**1**					
	PFDS	0.22	0.14	0.56	0.21	-0.22	0.27	0.53	0.64	0.47	0.19	0.33	-0.01	0.10	-0.31	**1**				
	pH	0.14	-0.30	-0.24	-0.31	-0.29	0.06	-0.07	0.16	-0.17	-0.06	-0.11	-0.20	-0.58	-0.21	0.32	**1**			
	T	0.30	-0.18	-0.09	-0.17	-0.11	0.27	0.10	-0.40	0.04	-0.50	-0.15	-0.21	-0.02	0.04	-0.19	-0.10	**1**		
	Salinity	0.30	-0.34	0.15	0.42	-0.08	0.00	0.31	-0.17	0.36	-0.45	0.08	-0.23	0.00	-0.38	0.01	-0.02	0.42	**1**	
	TSS	-0.13	-0.05	-0.46	-0.15	0.25	0.08	-0.49	0.02	-0.30	-0.07	-0.39	0.21	-0.33	0.18	-0.25	0.54	-0.28	-0.11	**1**
Summer	PFBA	**1**																		
	PFPeA	0.29	**1**																	
	PFHxA	-0.16	0.17	**1**																
	PFHpA	0.31	0.41	0.21	**1**															
	PFOA	0.61	0.63	-0.18	0.13	**1**														
	PFNA	0.11	-0.25	-0.07	0.29	0.08	**1**													
	PFDA	0.06	-0.06	0.42	0.49	-0.17	0.58	**1**												
	PFUnDA	0.10	-0.22	0.24	0.35	0.08	0.39	0.30	**1**											
	PFDoDA	0.23	0.34	-0.13	0.57	0.26	0.32	0.09	0.39	**1**										
	PFTrDA	0.56	0.40	-0.21	0.49	0.42	0.37	0.17	0.20	0.61	**1**									
	PFTeDA	-0.09	0.26	0.14	**0.68**	-0.04	0.47	0.40	0.32	**0.74**	0.50	**1**								
	PFBS	**0.69**	0.46	-0.26	0.02	**0.69**	-0.01	-0.06	-0.21	0.10	0.28	-0.03	**1**							
	PFHxS	-0.12	0.25	-0.25	0.30	0.05	0.27	0.07	-0.25	0.44	0.31	0.65	0.12	**1**						
	PFOS	0.52	0.35	-0.14	0.28	**0.78**	0.38	0.08	0.12	0.14	0.40	0.04	0.41	0.27	**1**					
	PFDS	0.50	0.32	-0.06	0.53	0.31	0.20	0.12	0.18	0.19	0.45	0.03	0.04	0.06	0.55	**1**				
	pH	-0.10	0.19	-0.45	-0.14	-0.03	-0.70	-0.66	-0.46	-0.22	-0.14	-0.23	0.16	0.05	-0.22	-0.09	**1**			
	T	-0.03	-0.37	0.11	0.11	-0.36	0.33	-0.02	0.19	0.04	0.14	0.04	-0.58	-0.01	0.02	0.48	-0.32	**1**		
	Salinity	0.20	0.37	0.18	**0.85**	0.09	0.33	0.47	0.34	**0.68**	0.61	**0.86**	0.10	0.37	0.10	0.15	-0.11	-0.10	**1**	
	TSS	-0.30	-0.64	-0.17	-0.57	-0.50	-0.30	-0.34	-0.13	-0.52	-0.67	-0.60	-0.35	-0.23	-0.39	-0.15	0.15	0.28	**-0.78**	**1**

Values in bold are significant at the 0.01 level (2-tailed).

Table B-9. Spearman rank correlations among the individual PFAAs and some physical chemical properties [pH, total organic carbon (TOC), black carbon (BC), total nitrogen (TN)] of sediment samples (*n=14*).

Seasons	Variables	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	pH	TOC	BC	TN
Winter	PFBA	**1**																		
	PFPeA	0.31	**1**																	
	PFHxA	0.09	0.60	**1**																
	PFHpA	0.44	0.25	**0.67**	**1**															
	PFOA	0.17	-0.29	-0.14	0.01	**1**														
	PFNA	0.08	0.20	0.60	0.54	0.09	**1**													
	PFDA	0.02	0.31	**0.78**	0.58	0.16	**0.92**	**1**												
	PFUnDA	0.31	0.64	0.15	-0.12	0.11	0.07	0.09	**1**											
	PFDoDA	0.12	0.28	0.79	**0.68**	-0.16	**0.85**	**0.82**	-0.05	**1**										
	PFTrDA	0.23	0.52	0.61	0.62	-0.37	0.54	0.51	0.37	0.63	**1**									
	PFTeDA	0.17	0.09	0.27	0.13	0.49	0.66	0.61	0.44	0.42	0.28	**1**								
	PFBS	0.22	**0.80**	**0.87**	0.46	-0.12	0.35	0.55	0.50	0.49	0.55	0.21	**1**							
	PFHxS	0.21	0.38	0.69	0.55	-0.29	0.21	0.39	0.06	0.53	0.53	0.05	0.60	**1**						
	PFOS	0.13	0.59	0.67	0.21	-0.03	0.38	0.47	0.32	0.58	0.37	0.37	**0.67**	0.49	**1**					
	PFDS	0.32	0.18	0.33	0.55	-0.22	0.28	0.21	-0.01	0.51	0.51	-0.22	0.24	0.37	0.20	**1**				
	pH	-0.33	-0.06	-0.39	-0.48	-0.17	**-0.70**	-0.66	0.03	-0.52	-0.27	-0.29	-0.31	-0.10	-0.05	-0.38	**1**			
	TOC	0.45	0.63	**0.79**	0.62	0.18	0.65	**0.75**	0.51	**0.71**	0.59	0.56	**0.79**	0.49	**0.67**	0.37	-0.49	**1**		
	BC	0.13	0.52	**0.71**	0.49	0.28	**0.74**	**0.82**	0.49	**0.69**	0.59	0.72	**0.62**	0.37	**0.67**	0.21	-0.31	**0.88**	**1**	
	TN	0.50	0.59	**0.76**	**0.71**	-0.10	0.61	0.67	0.32	**0.75**	0.62	0.27	**0.71**	0.55	0.50	0.56	-0.58	**0.90**	0.67	**1**
Summer	PFBA	**1**																		
	PFPeA	0.29	**1**																	
	PFHxA	**0.75**	-0.25	**1**																
	PFHpA	0.14	-0.11	-0.04	**1**															
	PFOA	-0.32	-0.23	0.17	0.44	**1**														
	PFNA	-0.22	0.46	0.28	0.07	0.53	**1**													
	PFDA	-0.06	-0.09	0.27	0.41	**0.64**	0.33	**1**												
	PFUnDA	0.36	-0.15	-0.41	0.09	-0.21	-0.18	-0.34	**1**											
	PFDoDA	-0.19	0.16	0.31	0.48	0.51	0.48	0.64	-0.08	**1**										
	PFTrDA	-0.06	0.52	0.20	0.28	-0.06	0.27	0.22	-0.03	0.56	**1**									
	PFTeDA	0.37	0.01	-0.18	0.24	0.19	0.08	0.53	0.17	0.41	0.30	**1**								
	PFBS	0.08	0.27	-0.06	-0.15	0.17	0.61	-0.13	0.10	-0.11	-0.17	0.21	**1**							
	PFHxS	-0.34	-0.25	0.15	0.04	0.36	-0.13	0.20	-0.30	0.10	0.21	-0.19	-0.51	**1**						
	PFOS	0.52	0.05	-0.36	0.27	0.22	-0.09	**0.52**	-0.13	0.18	0.10	**0.72**	-0.05	-0.02	**1**					
	PFDS	0.02	0.37	-0.17	-0.28	-0.06	0.13	-0.17	-0.13	-0.11	0.08	0.24	0.40	-0.28	0.41	**1**				
	pH	-0.07	-0.31	-0.12	-0.15	-0.45	-0.60	-0.55	0.30	-0.60	0.02	-0.27	-0.35	0.30	-0.22	-0.10	**1**			
	TOC	0.35	0.44	0.17	0.37	0.25	0.25	0.45	-0.01	0.36	0.45	0.44	0.16	0.12	0.34	0.01	-0.40	**1**		
	BC	0.42	0.34	0.37	-0.19	0.09	0.23	0.31	-0.06	0.12	-0.23	0.28	0.31	-0.29	0.24	0.08	-0.71	0.44	**1**	
	TN	-0.04	0.03	0.18	0.29	0.25	0.00	0.56	-0.33	0.31	0.12	0.24	-0.17	0.13	0.10	-0.36	-0.35	0.59	0.24	**1**

Values in bold are significant at the 0.01 level (2-tailed).

Table B-10. Spearman rank correlations ($p < 0.01$) among individual PFAAs between water and sediment samples (considering both seasons).

		water														
		PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS
sediment	PFBA	0.075	0.001	0.069	0.368	0.043	0.085	-0.009	-0.214	0.019	0.031	-0.112	0.095	-0.022	-0.065	0.302
	PFPeA	**0.439**	**0.877**	**0.455**	**0.535**	-0.074	-0.098	-0.065	0.190	0.152	0.217	-0.215	**0.511**	0.427	0.085	0.372
	PFHxA	-0.040	0.331	**0.914**	0.307	0.006	**0.418**	**0.847**	0.050	**0.771**	**0.446**	0.204	0.344	0.303	0.161	-0.005
	PFHpA	-0.277	-0.151	0.033	-0.050	-0.268	-0.031	0.053	-0.342	0.029	0.377	-0.051	-0.156	-0.086	-0.164	0.005
	PFOA	-0.091	-0.067	0.103	0.239	-0.182	0.195	0.286	-0.189	0.170	0.160	0.085	0.075	0.026	0.118	0.102
	PFNA	-0.174	-0.232	-0.149	-0.074	-0.143	0.041	0.033	-0.259	0.008	-0.041	-0.025	-0.219	-0.174	-0.190	-0.105
	PFDA	-0.318	-0.145	**0.452**	-0.080	-0.274	0.248	**0.562**	-0.304	**0.445**	0.326	0.049	-0.132	-0.014	-0.161	-0.272
	PFUnDA	0.269	**0.448**	**0.531**	0.317	0.249	0.237	**0.411**	0.165	**0.480**	0.216	0.232	**0.520**	**0.396**	0.043	**0.401**
	PFDoDA	-0.148	0.020	**0.666**	**0.394**	-0.168	0.352	**0.808**	-0.213	**0.673**	**0.487**	0.173	0.060	0.237	0.104	-0.047
	PFTrDA	-0.050	-0.104	-0.087	0.068	0.054	0.143	0.006	0.128	-0.077	0.305	**0.405**	-0.067	-0.120	0.322	-0.151
	PFTeDA	-0.226	-0.108	**0.541**	0.177	-0.060	**0.394**	**0.758**	-0.195	**0.635**	**0.528**	0.299	-0.036	0.080	0.126	-0.154
	PFBS	**0.435**	**0.699**	0.261	**0.492**	0.110	-0.090	-0.091	0.328	0.001	0.135	-0.104	**0.570**	**0.399**	0.015	**0.399**
	PFHxS	0.083	0.344	0.117	**0.638**	-0.228	-0.152	-0.126	-0.201	0.001	0.242	-0.232	0.039	0.291	0.086	0.216
	PFOS	**0.390**	0.032	-0.217	0.313	-0.229	-0.066	-0.198	0.085	-0.118	-0.022	0.073	0.035	-0.039	0.183	0.047
	PFDS	0.127	0.132	0.237	**0.539**	-0.158	0.020	0.093	-0.152	0.160	0.242	0.055	0.088	-0.020	0.072	0.145

Values in bold are significant at the 0.01 level (2-tailed).

Table B-11. Recoveries*, LODs and LOQs for individual PFAAs in seafood.

PFAAs	% Recoveries [mean (RSD)] Spiked at 10 ng/g (n=3)	LOD[a] (ng/g ww)	LOQ[b] (ng/g ww)	Linearity (r^2)[c]
PFBA	88 (3)	0.05	0.19	0.9991
PFPeA	117 (4)	0.03	0.07	0.9974
PFHxA	113 (2)	0.03	0.10	0.9984
PFHpA	82 (6)	0.04	0.13	0.9987
PFOA	114 (4)	0.01	0.03	0.999
PFNA	82 (5)	0.03	0.10	0.9986
PFDA	113 (5)	0.02	0.07	0.9988
PFUnDA	105 (4)	0.02	0.04	0.9992
PFDoDA	119 (2)	0.01	0.03	0.9993
PFTrDA	87 (2)	0.02	0.05	0.9985
PFTeDA	86 (6)	0.03	0.08	0.999
PFBS	85 (3)	0.02	0.09	0.999
PFHxS	113 (2)	0.01	0.03	0.9993
PFOS	110 (5)	0.02	0.07	0.999
PFDS	106 (2)	0.02	0.07	0.9993

[a] Limit of detection;

[b] Limit of quantification;

[c] Calibration curves (0.01 – 50 µg/L for each compound);

*The spike recovery of PFAAs was calculated using the equation: spike recovery = ($C_{\text{sample + spiked}}$ - C_{sample}) / C_{spiked} × 100%, where $C_{\text{sample + spiked}}$ is the concentration of PFAAs in a spiked sample, C_{sample} is the concentration of PFAAs in the sample (same as above without spiking target compounds, C_{spiked} is the concentration of the spiked target compound.

Table B-12. Concentration of individual PFAAs in seafood (finfish and shellfish) samples (ng/g ww) collected from the coastal area of Bangladesh in winter 2015.

Location	Species	Moisture (%)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFCA	∑PFSA	∑PFAAs
Cox's Bazar	Ilish	72	0.42	0.80	<LOD	<LOD	<LOD	<LOD	<LOD	0.74	<LOD	1.77	<LOD	0.55	<LOQ	0.93	0.25	3.72	1.72	5.45
	Rupchanda	71	<LOD	0.19	<LOD	<LOD	0.10	<LOQ	<LOQ	<LOD	0.04	<LOD	<LOD	0.16	<LOD	0.44	<LOD	0.32	0.60	0.92
	Loitta	79	<LOD	0.37	<LOD	<LOD	<LOD	<LOQ	<LOQ	<LOD	0.07	0.10	<LOD	<LOD	<LOD	0.38	<LOQ	0.53	0.38	0.91
	Sole	77	<LOD	<LOD	0.73	<LOD	0.09	0.23	0.12	0.76	0.09	0.11	<LOD	<LOQ	<LOQ	0.39	<LOQ	2.12	0.39	2.52
	Poa	80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.32	0.48	0.09	0.67	<LOD	0.47	<LOD	0.90	1.14	2.04
	Shrimp	84	<LOD	<LOD	<LOQ	<LOD	0.12	<LOQ	0.14	<LOD	0.08	0.09	<LOQ	0.13	<LOD	0.75	<LOD	0.43	0.88	1.31
	Crab	68	<LOD	<LOD	<LOD	<LOD	1.12	1.77	0.58	<LOD	0.13	0.27	<LOD	0.50	<LOD	0.46	<LOD	3.87	0.96	4.83
Chittagong	Ilish	58	<LOD	0.39	<LOD	<LOD	0.10	<LOD	<LOD	0.68	1.71	0.72	0.77	0.73	<LOQ	2.41	1.20	4.37	4.34	8.71
	Rupchanda	76	<LOD	0.19	<LOD	1.12	0.21	<LOQ	<LOQ	0.07	0.18	0.32	<LOD	0.21	<LOD	0.44	<LOD	2.08	0.65	2.74
	Loitta	81	<LOD	0.31	0.15	<LOD	0.09	<LOQ	<LOQ	<LOD	<LOD	0.24	<LOQ	<LOD	<LOD	0.24	<LOD	0.79	0.24	1.02
	Sole	79	<LOD	<LOQ	0.39	<LOQ	0.09	0.24	0.11	<LOD	0.08	0.12	<LOQ	0.11	<LOD	0.34	<LOD	1.04	0.44	1.48
	Poa	75	<LOD	<LOD	<LOD	<LOD	0.06	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	0.05	0.21	<LOQ	0.06	0.26	0.32
	Shrimp	77	0.64	<LOD	<LOD	<LOD	1.16	0.16	0.12	0.47	0.08	<LOQ	<LOQ	<LOQ	<LOD	0.47	<LOD	2.62	0.47	3.10
	Crab	75	<LOQ	<LOD	<LOD	<LOD	1.26	2.11	1.34	0.98	0.10	0.15	<LOQ	0.40	<LOD	0.85	<LOQ	5.93	1.25	7.18
Bhola	Ilish	59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.90	1.39	<LOD	0.55	0.09	0.73	1.46	3.29	2.82	6.11
	Poa	77	0.60	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.90	<LOD	<LOD	0.48	<LOQ	0.76	<LOD	1.50	1.24	2.74
	Loitta	75	0.56	<LOD	<LOD	0.85	0.06	0.29	0.50	<LOD	0.03	0.30	<LOD	0.32	<LOD	0.57	<LOD	2.61	0.89	3.49
	Shrimp	86	<LOD	<LOD	<LOD	0.66	0.11	0.26	0.19	<LOD	<LOD	<LOQ	<LOQ	0.19	<LOD	0.45	<LOQ	1.22	0.64	1.87
	Crab	67	<LOD	<LOD	<LOD	<LOD	0.61	<LOD	1.01	<LOD	0.31	0.49	<LOD	0.78	<LOD	0.72	0.71	2.42	2.21	4.62
Sundarbans	Ilish	65	<LOD	0.32	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.92	0.49	<LOD	0.73	<LOD	1.06	<LOD	1.73	1.79	3.52
	Poa	68	<LOD	0.62	0.45	<LOD	<LOD	0.12	<LOQ	<LOD	0.05	<LOQ	<LOD	<LOD	<LOD	0.64	<LOD	1.24	0.64	1.88
	Loitta	87	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	0.09	<LOQ	<LOD	<LOD	0.23	0.08	0.09	0.31	0.41
	Shrimp	84	<LOD	<LOD	<LOD	<LOD	0.07	<LOD	0.15	<LOD	<LOD	<LOQ	<LOQ	0.80	<LOD	0.75	<LOD	0.22	1.55	1.77
	Crab	62	<LOD	<LOD	<LOD	<LOD	0.23	<LOD	0.22	<LOD	<LOD	<LOQ	<LOQ	0.18	<LOD	1.61	0.09	0.45	1.88	2.33

Note: For LOD and LOQ please refer to Table B-11.

Table B-13. Concentration of individual PFAAs in seafood (finfish and shellfish) samples (ng/g ww) collected from the coastal area of Bangladesh in summer 2015.

Location	Species	Moisture (%)	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS	∑PFCA	∑PFSA	∑PFAAs
Cox's Bazar	Ilish	63	0.72	0.38	<LOD	<LOD	0.27	0.14	0.41	0.82	1.03	1.50	1.23	0.69	0.42	1.94	0.84	6.49	3.89	10.38
	Rupchanda	79	<LOD	0.64	0.43	<LOD	0.09	<LOQ	<LOQ	0.09	<LOD	0.07	<LOD	<LOD	<LOD	0.81	0.17	1.31	0.98	2.29
	Loitta	84	<LOD	<LOD	<LOD	<LOD	0.09	0.10	0.08	<LOD	0.03	0.04	<LOD	0.20	0.16	0.10	<LOQ	0.35	0.45	0.80
	Sole	76	<LOD	<LOD	0.28	<LOD	0.08	0.45	0.08	<LOD	<LOQ	<LOQ	<LOD	<LOD	0.13	0.35	<LOD	0.88	0.48	1.36
	Poa	78	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.17	0.26	0.49	0.57	1.83	<LOD	0.44	2.89	3.32
	Shrimp	76	0.48	<LOD	<LOD	<LOD	0.24	0.27	0.20	<LOD	0.17	0.54	<LOD	<LOD	<LOD	0.20	<LOD	1.90	0.20	2.10
	Crab	59	0.66	0.15	<LOD	<LOD	0.88	0.64	0.50	<LOD	0.20	0.14	0.12	1.58	<LOD	0.26	<LOD	3.30	1.84	5.14
Chittagong	Ilish	74	<LOD	0.11	<LOD	<LOD	<LOD	0.92	<LOD	0.88	1.57	1.98	1.34	1.21	0.69	3.86	2.02	6.80	7.78	14.58
	Rupchanda	82	<LOD	0.65	<LOD	1.07	<LOQ	<LOD	0.07	<LOD	<LOD	0.50	0.38	1.11	0.60	0.79	<LOD	2.68	2.49	5.18
	Loitta	85	<LOD	0.30	<LOQ	1.08	0.05	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	0.32	<LOD	0.34	<LOD	1.43	0.66	2.08
	Sole	81	<LOD	<LOD	0.65	<LOD	0.18	1.56	0.52	<LOD	0.22	0.15	0.10	0.77	<LOD	0.59	<LOD	3.38	1.36	4.74
	Poa	78	<LOD	<LOD	<LOD	0.73	0.40	0.77	0.82	<LOD	0.28	0.41	0.18	1.47	0.55	0.61	<LOD	3.59	2.63	6.22
	Shrimp	81	<LOD	<LOD	<LOD	<LOD	1.25	0.27	0.20	0.56	0.04	<LOQ	<LOD	0.03	<LOD	0.24	<LOD	2.32	0.27	2.60
	Crab	63	<LOD	<LOD	<LOD	<LOD	2.39	0.49	<LOD	1.17	<LOD	0.21	<LOD	1.00	<LOD	1.99	1.08	4.26	4.08	8.34
Bhola	Ilish	70	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.41	<LOD	2.01	0.78	<LOD	<LOD	0.07	2.07	0.31	3.20	2.45	5.65
	Poa	75	0.60	0.38	<LOD	<LOD	<LOD	<LOD	0.27	<LOD	0.25	<LOD	<LOD	<LOD	<LOD	0.81	<LOD	1.50	0.81	2.31
	Loitta	81	0.27	0.25	<LOD	<LOQ	<LOD	0.20	0.89	0.07	0.21	0.13	<LOD	1.02	<LOD	0.65	<LOD	2.02	1.67	3.69
	Shrimp	75	<LOD	<LOD	<LOD	<LOD	0.46	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	0.68	<LOD	0.10	0.16	0.46	0.94	1.41
	Crab	74	<LOD	<LOD	<LOD	<LOD	0.87	<LOD	0.30	<LOD	0.24	0.18	<LOD	<LOD	0.36	0.16	0.28	1.59	0.80	2.40
Sundarbans	Ilish	65	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.99	0.85	0.83	0.43	<LOD	2.54	<LOD	2.67	2.97	5.63
	Poa	69	<LOD	<LOD	0.28	<LOD	<LOD	0.23	1.20	<LOD	0.29	0.19	<LOD	<LOD	<LOD	0.59	<LOD	2.18	0.59	2.78
	Loitta	80	<LOD	<LOD	<LOD	<LOD	<LOD	0.54	0.52	<LOD	<LOD	0.12	<LOD	<LOD	<LOD	1.79	<LOD	1.18	1.79	2.97
	Shrimp	73	<LOD	0.19	<LOD	<LOQ	0.28	0.13	0.25	0.05	0.12	0.23	<LOD	<LOD	<LOD	0.27	0.07	1.25	0.34	1.60
	Crab	59	<LOD	<LOD	<LOD	<LOD	0.26	0.42	0.37	<LOD	0.11	<LOD	<LOD	0.75	<LOD	0.30	<LOD	1.16	1.05	2.21

Note: For LOD and LOQ please refer to Table B-11.

Table B-14. Spearman rank correlation between individual PFAAs.

i. Finfish

Variables	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS
PFBA	**1**														
PFPeA	0.2857	**1**													
PFHxA	-0.2932	0.0132	**1**												
PFHpA	0.0701	0.1018	-0.0460	**1**											
PFOA	-0.0998	0.0245	0.2761	0.3108	**1**										
PFNA	-0.0870	-0.2977	0.4438	0.1935	0.4012	**1**									
PFDA	0.1686	-0.1906	0.2862	0.3159	0.3432	**0.6342**	**1**								
PFUnDA	0.2592	0.3949	0.0125	-0.0152	0.2582	0.1242	-0.0749	**1**							
PFDoDA	0.1140	-0.1324	-0.2304	-0.1890	-0.0628	-0.1229	-0.0332	0.1720	**1**						
PFTrDA	0.0097	0.0270	-0.3369	0.0317	-0.0585	-0.1551	-0.1656	0.3827	**0.5714**	**1**					
PFTeDA	-0.1427	-0.0461	-0.1652	-0.0153	0.2072	0.0143	-0.1999	0.1519	0.1856	**0.5844**	**1**				
PFBS	0.1682	0.0930	-0.3598	0.3134	0.1551	-0.0503	-0.1538	0.2922	0.3555	**0.5466**	**0.5122**	**1**			
PFHxS	0.0132	-0.1171	-0.2758	-0.0705	0.1336	-0.0334	-0.0868	0.2285	0.1523	0.3410	0.3823	0.4020	**1**		
PFOS	0.2365	0.2490	-0.3300	-0.2106	-0.3103	-0.2907	-0.1689	0.3641	**0.5647**	**0.5766**	0.2814	0.3736	0.2618	**1**	
PFDS	0.0034	0.1101	-0.1918	-0.3921	0.0087	-0.1188	-0.2765	**0.5564**	0.2575	0.3757	0.1372	0.0350	0.4442	0.2347	**1**

Values in bold are significant at the 0.01 level (2-tailed).

ii. Shellfish

Variables	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS
PFBA	**1**														
PFPeA	0.2637	**1**													
PFHxA	-0.1472	-0.0974	**1**												
PFHpA	-0.2150	0.4244	-0.0974	**1**											
PFOA	0.3014	0.0307	-0.3081	-0.3400	**1**										
PFNA	0.4499	0.1469	-0.1694	-0.0361	**0.5471**	**1**									
PFDA	0.1294	0.2378	-0.2521	-0.0511	0.1912	0.2580	**1**								
PFUnDA	0.1785	0.0684	-0.1703	0.0311	**0.7152**	0.4038	-0.1466	**1**							
PFDoDA	0.3000	0.3244	-0.0284	-0.1453	0.1941	0.1174	**0.6986**	-0.2396	**1**						
PFTrDA	0.1609	0.2181	-0.0284	0.0208	0.2510	0.1755	0.3735	0.0835	**0.6475**	**1**					
PFTeDA	0.4617	0.1542	0.2550	0.0451	-0.2343	0.0143	-0.0351	-0.0814	-0.2531	-0.3128	**1**				
PFBS	-0.0155	0.0051	-0.1404	-0.2718	0.0973	0.2156	0.0590	-0.1076	-0.1587	-0.1633	0.2266	**1**			
PFHxS	-0.1472	-0.0974	-0.0667	-0.0974	0.0840	-0.3388	0.1400	-0.1703	0.3695	0.1422	-0.2231	-0.3651	**1**		
PFOS	-0.1005	-0.2429	0.2521	-0.1176	0.0353	0.0267	-0.0412	0.3004	-0.3493	0.0627	0.3983	**0.5693**	-0.3641	**1**	
PFDS	-0.3945	-0.0478	-0.2395	0.1490	0.1824	-0.3439	-0.0346	0.1491	-0.0622	0.2100	-0.3319	0.0883	0.3293	0.1399	**1**

Values in bold are significant at the 0.01 level (2-tailed).

iii. All seafood samples (finfish+shellfish)

Variables	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	PFOS	PFDS
PFBA	**1**														
PFPeA	0.2124	**1**													
PFHxA	-0.2585	0.1145	**1**												
PFHpA	-0.0139	0.2097	-0.0057	**1**											
PFOA	0.0788	-0.2833	-0.0932	0.0305	**1**										
PFNA	0.1237	-0.2523	0.2056	0.1314	**0.5324**	**1**									
PFDA	0.2397	-0.2829	0.0287	0.1608	**0.5943**	**0.6846**	**1**								
PFUnDA	0.2511	0.2496	-0.0554	-0.0137	0.2917	0.2395	-0.0762	**1**							
PFDoDA	0.1775	0.0212	-0.1536	-0.1353	-0.0765	-0.0735	0.1367	0.0691	**1**						
PFTrDA	0.0361	0.1086	-0.2106	0.0598	-0.1081	-0.0804	-0.0596	0.2468	**0.5899**	**1**					
PFTeDA	0.0222	-0.0079	-0.0764	0.0184	0.0306	0.0029	-0.1122	0.0618	0.0984	0.2836	**1**				
PFBS	0.1057	0.0333	-0.3316	0.1546	0.1568	0.0505	-0.0322	0.1515	0.2134	0.2981	**0.5023**	**1**			
PFHxS	-0.0300	0.0344	-0.1320	-0.0326	-0.1760	-0.1719	-0.1968	0.1088	0.2053	0.3366	0.2614	0.1859	**1**		
PFOS	0.0945	0.1951	-0.1368	-0.1382	-0.3094	-0.2288	-0.1252	0.3079	0.2623	**0.5120**	0.2948	**0.5076**	0.1955	**1**	
PFDS	-0.1162	0.0265	-0.2207	-0.2386	0.1156	-0.1834	-0.1837	**0.5218**	0.1066	0.3099	0.0064	0.0485	0.3587	0.2111	**1**

Values in bold are significant at the 0.01 level (2-tailed).

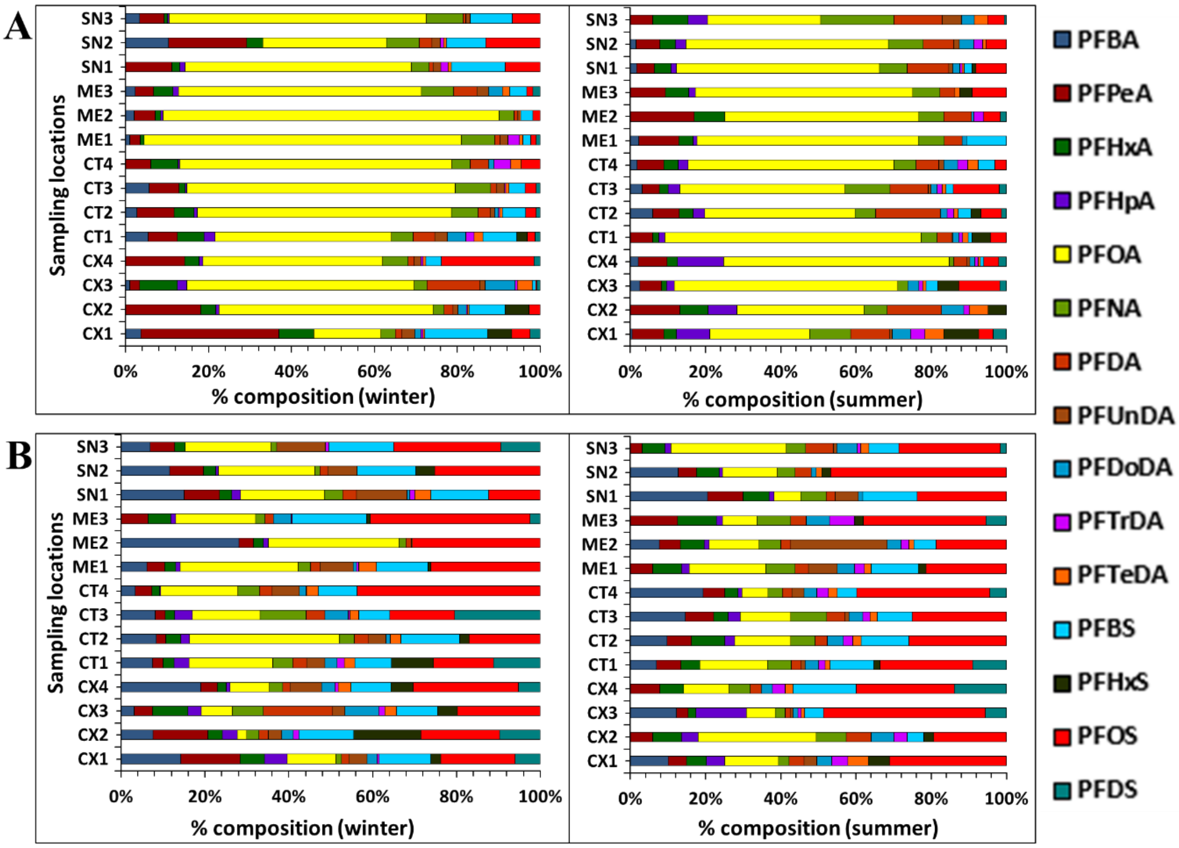

Figure B-1. Patterns of the relative contribution of individual PFAAs (% composition) to the total PFAAs in water (A) and sediment (B) of the coastal area of Bangladesh in winter and summer.

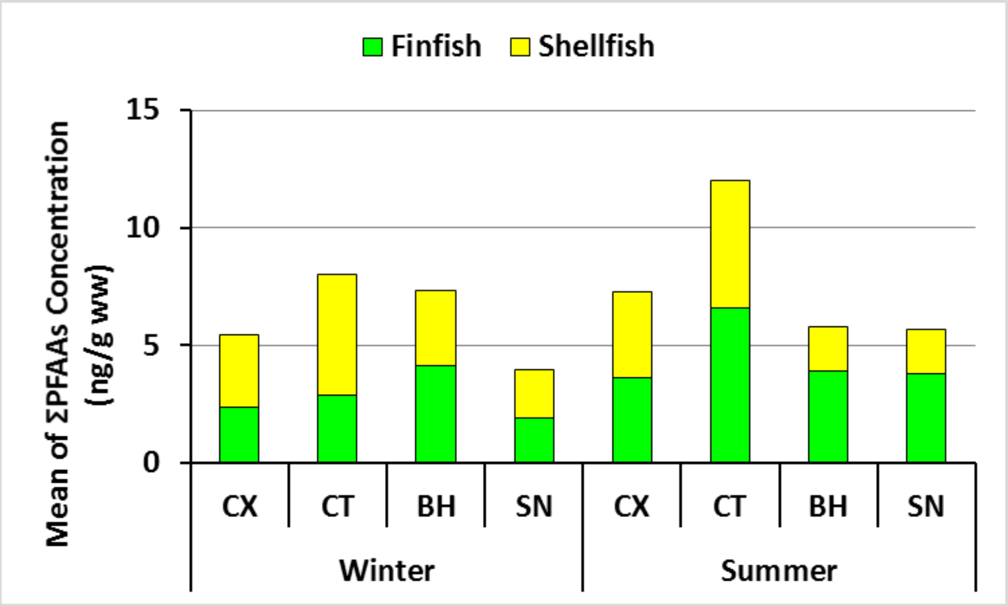

Figure B-2. Mean concentrations of total PFAAs (sum of 15 target PFAAs) in seafood (finfish and shellfish) collected from the coastal areas of Bangladesh in winter and summer of 2015. In figure, CX: Cox's Bazar, CT: Chittagong, BH: Bhola (Meghna estuary), SN: Sundarbans.

APPENDIX C

Table C-1. 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℃(1 min)-10℃/min-300℃(4min)-10℃/min-310℃(2min)
Inlet Temperature	300℃
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℃
Ion source temperature	300℃
Quadrupole temperature	150℃
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
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 C-2. Recoveries*, LODs, LOQs and linearity of calibration curves for the analytical determination of PAHs.

PAHs	% Recoveries [mean (RSD)]			LOD[a]			LOQ[b]			Linearity (r^2)[c]
	Water 10 ng/L (n=3)	Sediment 10 ng/g dw (n=3)	Seafood 10 ng/g ww (n=3)	Water (ng/L)	Sediment (ng/g dw)	Seafood (ng/g ww)	Water (ng/L)	Sediment (ng/g dw)	Seafood (ng/g ww)	
Nap	97 (6)	70 (3)	71 (4)	0.05	0.06	0.04	0.15	0.18	0.12	0.9986
Acel	114 (4)	97 (6)	97 (2)	0.10	0.27	0.01	0.29	0.82	0.03	0.9999
Ace	98 (1)	89 (4)	98 (4)	0.05	0.07	0.04	0.14	0.21	0.12	0.9999
Flu	103 (4)	98 (5)	101 (2)	0.11	0.11	0.06	0.34	0.33	0.18	0.9999
Phe	94 (4)	103 (1)	100 (4)	0.16	0.1	0.03	0.48	0.30	0.09	0.9997
Ant	84 (5)	92 (3)	113 (2)	0.10	0.16	0.07	0.32	0.48	0.21	0.9999
Flt	86 (4)	94 (5)	101 (4)	0.04	0.05	0.03	0.12	0.15	0.09	0.9992
Pyr	107 (6)	108 (2)	104 (1)	0.05	0.02	0.02	0.15	0.06	0.06	0.9993
BaA	93 (5)	111 (3)	99 (2)	0.03	0.07	0.03	0.09	0.21	0.09	0.9999
Chr	103 (4)	94 (5)	107 (8)	0.02	0.02	0.01	0.06	0.06	0.03	0.9999
BbF	94 (2)	102 (6)	93 (2)	0.12	0.02	0.02	0.36	0.06	0.06	0.9998
BkF	111 (7)	112 (4)	90 (7)	0.12	0.05	0.02	0.36	0.15	0.06	0.9992
BaP	88 (9)	93 (5)	85 (1)	0.19	0.08	0.01	0.59	0.24	0.03	0.9996
DahA	103 (4)	99 (2)	79 (5)	0.01	0.06	0.03	0.04	0.18	0.09	0.9996
BghiP	76 (4)	103 (4)	77 (2)	0.02	0.23	0.01	0.07	0.70	0.03	0.9998
IP	109 (7)	75 (5)	75 (5)	0.01	0.04	0.02	0.04	0.12	0.06	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:

Spike recovery rate (%) = ($C_{sample + spiked}$ − C_{sample}) / C_{spiked} × 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.

Table C-3. 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	3	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		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	4	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		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	5	<LOD	<LOD	49.5	<LOD	64.7	31.0	120.1	142.9	<LOD	<LOD	<LOD	<LOD	19.1	<LOD
BkF		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); [e] Sum of 16 USEPA PAHs.

Table C-4. 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	3	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		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	4	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		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	5	<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		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); [e] Sum of 16 USEPA PAHs

Table C-5. 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
		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
		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 C-6. Concentrations (ng/g dw) of 16 USEPA priority PAHs in surface sediment 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	31.7	28.0	64.6	75.0	202.2	84.1	130.5	82.2	137.1	40.0	13.1	11.5	43.7	27.2
Acel	3	9.3	19.8	22.7	95.8	81.8	74.8	16.6	246.8	23.7	11.3	4.2	40.6	43.2	9.1
Ace		42.5	30.3	19.7	20.6	31.5	25.5	32.7	8.7	3.5	0.9	0.5	106.7	40.1	12.7
Flu		61.1	9.8	68.5	5.3	117.2	78.8	101.0	102.8	13.0	2.9	1.2	126.1	29.0	15.7
Phe		77.3	93.1	689.0	487.8	1054.7	798.7	1132.4	1439.1	679.9	39.9	17.3	198.4	77.2	327.3
Ant		12.7	18.2	183.4	12.0	189.2	176.1	207.3	334.2	38.9	10.4	4.7	49.7	25.6	20.6
Flt	4	195.5	269.8	1032.6	443.5	3479.2	1069.7	1839.4	1921.8	1988.2	214.2	59.9	402.5	868.6	1870.3
Pyr		137.7	205.0	1470.3	975.0	2083.1	2024.7	2619.4	2116.4	1349.3	159.0	46.4	571.2	741.8	2242.1
BaA		33.1	13.6	222.6	257.5	203.4	255.6	51.3	341.9	31.1	116.4	85.3	39.6	483.2	26.1
Chr		48.0	28.9	780.2	404.0	1436.8	906.2	1973.9	719.9	141.8	158.7	90.3	218.8	439.1	567.8
BbF	5	7.9	9.6	520.4	601.0	1108.6	512.5	417.6	370.1	102.7	46.1	13.0	90.5	369.6	154.6
BkF		4.0	1.8	157.6	131.3	347.3	163.2	138.0	112.0	22.6	12.6	3.0	84.1	265.6	106.0
BaP		90.4	25.4	183.6	129.4	373.5	164.5	115.7	54.8	32.2	19.6	4.4	105.0	307.2	243.3
DahA		19.8	2.4	25.4	36.6	35.4	50.6	20.9	1.6	8.1	2.1	0.6	89.0	314.4	152.4
BghiP	6	13.0	3.7	149.1	32.8	181.7	188.3	75.3	66.9	33.7	10.3	3.4	33.0	271.0	22.1
IP		19.6	2.9	40.3	33.9	133.3	185.2	34.7	62.8	28.0	9.9	2.8	135.4	77.3	17.7
∑2-Ring PAHs		31.7	28.0	64.6	75.0	202.2	84.1	130.5	82.2	137.1	40.0	13.1	11.5	43.7	27.2
∑3-Ring PAHs		202.9	171.1	983.3	621.6	1474.4	1153.8	1490.0	2131.6	758.9	65.5	27.8	521.5	215.0	385.4
∑4-Ring PAHs		414.3	517.3	3505.8	2080.0	7202.4	4256.3	6483.9	5100.0	3510.4	648.3	281.8	1232.1	2532.6	4706.3
∑5-Ring PAHs		122.1	39.2	886.9	898.3	1864.8	890.7	692.2	538.4	165.5	80.4	21.0	368.6	1256.8	656.2
∑6-Ring PAHs		32.5	6.6	189.4	66.7	314.9	373.6	110.0	129.8	61.7	20.2	6.1	168.4	348.3	39.8
∑LMW-PAHs[a]		234.6	199.1	1047.9	696.6	1676.6	1237.9	1620.5	2213.8	896.1	105.5	40.9	533.0	258.7	412.6
∑HMW-PAHs[b]		569.0	563.1	4582.1	3044.9	9382.2	5520.6	7286.1	5768.2	3737.7	748.9	309.0	1769.1	4137.7	5402.4
∑C-PAHs[c]		222.8	84.5	1930.1	1593.7	3638.2	2237.8	2752.1	1663.1	366.4	365.4	199.3	762.3	2256.3	1267.8
∑PAHs[d]		803.6	762.1	5630.0	3741.5	11058.8	6758.5	8906.6	7982.0	4633.7	854.4	349.8	2302.0	4396.4	5815.0

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

Table C-7. Concentrations (ng/g dw) of 16 USEPA priority PAHs in surface sediment 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	156.5	75.8	677.0	321.5	1092.3	89.3	553.8	638.1	87.5	60.5	18.5	253.5	331.9	271.0
Acel	3	77.7	32.9	420.5	90.0	337.0	54.3	89.3	221.4	64.0	78.4	26.3	47.8	242.7	7.8
Ace		6.0	7.7	91.8	60.7	816.7	215.3	293.6	103.0	9.1	2.7	2.1	12.6	563.0	197.9
Flu		2.7	0.6	24.3	75.3	140.0	128.0	192.6	53.9	12.2	4.0	1.2	16.5	545.6	171.3
Phe		23.9	60.1	1538.3	898.8	1040.0	579.6	569.1	443.7	561.8	24.2	14.7	325.9	75.6	600.9
Ant		52.0	55.8	564.3	85.4	180.7	116.6	60.3	26.5	70.1	117.9	15.4	50.1	11.3	36.0
Flt	4	114.7	84.7	906.8	701.9	3664.7	2114.6	2587.6	1859.5	886.2	131.5	30.6	509.5	747.7	1553.4
Pyr		46.3	20.8	1304.7	2025.2	4857.9	4706.7	3528.3	6190.3	490.4	10.8	4.6	1350.0	110.5	947.1
BaA		106.0	85.7	104.8	124.2	451.9	411.9	473.4	184.6	57.7	52.5	14.7	85.5	73.6	201.4
Chr		106.8	123.4	231.2	134.9	1407.7	916.8	2907.9	263.5	155.9	49.8	10.8	266.0	46.3	1301.9
BbF	5	32.3	19.7	201.4	187.5	657.2	247.6	312.5	75.7	28.8	5.4	4.6	22.3	12.0	256.3
BkF		38.4	28.1	96.5	88.1	185.9	184.9	584.9	512.3	30.8	16.1	6.7	62.4	4.7	391.5
BaP		65.6	50.1	308.7	64.8	352.7	93.7	142.7	22.0	71.7	36.3	17.8	23.7	60.3	130.1
DahA		25.4	15.7	85.4	81.2	221.1	51.9	106.6	89.4	31.8	10.8	6.4	11.1	295.0	7.1
BghiP	6	27.2	22.0	126.1	83.1	207.8	34.8	184.6	34.7	44.7	6.7	9.5	15.7	40.9	2.6
IP		16.5	8.9	179.5	23.9	1475.5	3.0	380.9	49.4	102.3	4.9	15.9	111.7	10.0	5.0
∑2-Ring PAHs		156.5	75.8	677.0	321.5	1092.3	89.3	553.8	638.1	87.5	60.5	18.5	253.5	331.9	271.0
∑3-Ring PAHs		162.3	157.1	2639.2	1210.2	2514.4	1093.9	1204.9	848.5	717.3	227.3	59.7	453.0	1438.2	1013.8
∑4-Ring PAHs		373.9	314.6	2547.6	2986.2	10382.2	8150.0	9497.2	8497.9	1590.1	244.5	60.7	2211.1	978.1	4003.9
∑5-Ring PAHs		161.7	113.6	691.9	421.6	1417.0	578.2	1146.7	699.4	163.1	68.6	35.6	119.5	371.9	785.0
∑6-Ring PAHs		43.7	30.8	305.6	107.0	1683.3	37.8	565.4	84.1	147.1	11.7	25.4	127.4	50.9	7.6
∑LMW-PAHs[a]		318.8	232.9	3316.3	1531.7	3606.6	1183.2	1758.7	1486.5	804.8	287.8	78.3	706.5	1770.1	1284.8
∑HMW-PAHs[b]		579.3	459.1	3545.1	3514.8	13482.4	8766.0	11209.3	9281.5	1900.3	324.8	121.7	2458.0	1400.9	4796.4
∑C-PAHs[c]		391.1	331.7	1207.5	704.6	4752.0	1909.8	4908.8	1197.0	479.0	175.8	76.9	582.7	501.7	2293.3
∑PAHs[d]		898.1	692.0	6861.4	5046.5	17089.1	9949.2	12968.0	10768.0	2705.0	612.6	199.9	3164.4	3170.9	6081.3

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

Table C-8. Concentrations (ng/g ww) of 16 USEPA priority PAHs in seafood from the coastal area of Bangladesh in winter 2015.

PAHs	PAH Ring	Finfish																Shellfish							
		Ilish				Rupchanda		Loitta				Sole		Poa				Shrimp				Crab			
		CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	BH	SN	CX	CT	BH	SN
Nap	2	1185.0	1209.6	1540.8	1051.2	83.9	130.9	301.6	489.6	271.7	343.2	264.3	417.9	338.2	337.4	189.9	246.5	59.0	68.7	48.6	81.0	202.8	281.0	126.0	213.1
Acel	3	89.5	35.4	28.4	45.2	4.3	10.0	27.6	14.1	13.8	4.2	2.0	<LOD	48.4	10.6	21.5	47.2	0.8	4.2	<LOD[a]	0.7	17.1	12.7	<LOD	2.4
Ace		34.2	5.9	4.7	63.6	2.6	6.7	9.5	45.6	53.3	9.1	48.6	163.8	8.4	17.2	13.8	16.0	15.2	24.0	28.5	21.2	108.5	210.8	72.1	71.8
Flu		110.8	266.5	35.5	106.8	11.3	13.1	33.2	36.9	6.8	45.4	33.2	58.2	20.7	44.2	36.3	61.3	19.1	19.1	17.1	9.8	102.5	169.4	35.1	200.7
Phe		718.6	745.9	322.2	357.2	45.6	55.7	150.8	275.8	87.5	216.3	311.6	145.6	90.6	141.7	79.3	127.7	69.3	89.2	62.8	102.7	401.8	378.3	113.2	761.1
Ant		29.9	83.7	56.9	86.2	8.9	14.8	67.9	22.8	53.3	18.5	3.1	6.1	14.6	2.9	4.1	33.3	39.5	6.7	24.7	21.8	78.5	28.5	85.2	209.0
Flt	4	129.0	27.8	82.9	23.2	5.3	26.2	40.3	28.2	34.9	71.1	126.6	92.9	63.7	51.8	68.6	66.8	33.1	80.3	24.4	30.4	85.3	191.7	83.1	293.0
Pyr		160.6	61.9	97.1	30.8	10.9	18.6	66.3	97.7	61.9	41.3	136.0	118.3	109.0	74.5	77.3	27.9	34.2	57.3	28.8	50.7	165.7	264.7	135.2	188.2
BaA		34.2	38.3	49.7	22.6	0.6	0.3	11.1	33.7	0.7	0.9	47.0	<LOD	17.7	25.4	10.7	6.9	1.4	4.7	1.8	2.8	11.4	12.7	5.4	23.2
Chr		65.8	82.5	75.8	69.8	5.7	10.5	32.4	25.0	35.5	8.5	77.9	95.4	129.5	169.9	9.7	27.3	1.0	8.8	3.3	2.1	20.0	25.8	4.1	66.3
BbF	5	<LOD	8.4	<LOD	1.6	0.1	<LOD	9.5	1.1	8.6	0.1	<LOD	47.8	<LOD	9.0	0.2	1.3	6.8	<LOD	9.5	6.0	30.0	34.9	5.7	8.9
BkF		2.6	4.8	<LOD	<LOD	<LOD	<LOD	0.6	<LOD	4.6	<LOD	0.1	20.6	0.5	0.2	<LOD	<LOD	2.3	0.8	3.7	2.4	15.7	21.2	3.5	11.3
BaP		2.4	4.0	0.9	1.4	<LOD	0.3	0.1	0.0	0.1	<LOD	<LOD	0.2	<LOD	<LOD	0.2	0.1	0.6	0.1	0.3	0.7	3.1	2.4	5.1	2.0
DahA		2.1	0.4	9.5	<LOD	2.3	<LOD	<LOD	13.0	2.0	8.7	0.4	6.2	0.3	<LOD	<LOD	<LOD	0.4	<LOD	0.6	1.4	30.0	3.4	<LOD	3.0
BghiP	6	18.4	199.2	<LOD	2.1	1.6	0.1	204.2	2.4	<LOD	6.3	0.9	<LOD	<LOD	13.0	<LOD	0.3	0.8	4.6	<LOD	<LOD	106.6	163.9	<LOD	6.5
IP		10.5	32.4	64.7	49.3	1.3	3.5	33.2	2.2	23.7	15.9	<LOD	<LOD	18.4	9.8	<LOD	2.7	2.6	5.7	<LOD	<LOD	57.4	36.0	<LOD	37.8
∑2-Ring PAHs		1185.0	1209.6	1540.8	1051.2	83.9	130.9	301.6	489.6	271.7	343.2	264.3	417.9	338.2	337.4	189.9	246.5	59.0	68.7	48.6	81.0	202.8	281.0	126.0	213.1
∑3-Ring PAHs		983.0	1137.3	447.7	659.0	72.6	100.3	289.0	395.2	214.7	293.6	398.5	373.6	182.7	216.6	155.1	285.6	143.8	143.2	133.1	156.3	708.6	799.6	305.6	1245.0
∑4-Ring PAHs		389.6	210.6	305.6	146.4	22.6	55.5	150.0	184.6	132.9	121.8	387.5	306.6	319.9	321.6	166.3	128.9	69.6	151.1	58.3	86.0	282.4	494.8	227.8	570.7
∑5-Ring PAHs		7.1	17.4	10.4	3.1	2.4	0.3	10.1	14.2	15.2	8.8	0.6	74.7	0.8	9.2	0.4	1.5	10.1	0.9	14.0	10.4	78.8	61.9	14.4	25.1
∑6-Ring PAHs		29.0	231.6	64.7	51.3	2.9	3.6	237.4	4.6	23.7	22.3	0.9	<LOD	18.4	22.9	<LOD	2.9	3.3	10.3	<LOD	<LOD	164.0	199.9	<LOD	44.3
∑LMW-PAHs[b]		2168.0	2346.9	1988.5	1710.2	156.5	231.2	590.6	884.8	486.4	636.8	662.8	791.6	520.9	554.0	345.0	532.1	202.8	212.0	181.7	237.2	911.4	1080.6	431.7	1458.2
∑HMW-PAHs[c]		425.6	459.7	380.7	200.8	27.9	59.4	397.6	203.3	171.8	152.9	388.9	381.2	339.1	353.6	166.8	133.3	83.1	162.2	72.3	96.4	525.2	756.7	242.2	640.1
∑C-PAHs[d]		117.7	170.7	200.6	144.7	10.1	14.6	86.8	75.0	75.1	34.2	125.4	170.0	166.3	214.3	20.9	38.3	15.0	20.1	19.1	15.3	167.6	136.4	23.9	152.4
∑PAHs[e]		2593.7	2806.6	2369.2	1911.0	184.5	290.6	988.2	1088.2	658.2	789.7	1051.7	1172.8	860.0	907.6	511.7	665.4	285.9	374.2	254.0	333.6	1436.6	1837.3	673.9	2098.3

[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); [e] Sum of 16 USEPA PAHs.

Table C-9. Concentrations (ng/g ww) of 16 USEPA priority PAHs in seafood from the coastal area of Bangladesh in summer 2015.

PAHs	PAH Ring	Finfish																Shellfish							
		Ilish				Rupchanda		Loitta				Sole		Poa				Shrimp				Crab			
		CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	CX	CT	BH	SN	CX	CT	BH	SN	CX	CT	BH	SN
Nap	2	1938.1	1870.7	1466.3	1718.3	98.6	161.8	563.0	840.9	714.0	990.3	357.7	587.4	405.1	608.9	356.0	461.7	25.3	27.3	22.7	38.2	124.9	205.0	90.3	225.1
Acel	3	76.6	140.3	111.1	59.0	4.1	4.1	52.9	55.0	57.2	36.6	2.4	4.4	45.9	39.7	4.4	13.3	1.5	6.3	3.1	0.5	38.6	2.3	18.9	5.9
Ace		7.3	116.7	1.3	22.5	8.0	4.5	59.4	66.7	70.9	9.5	100.2	108.0	29.5	44.4	9.5	43.0	8.8	2.5	2.1	4.3	52.0	69.1	25.2	20.7
Flu		231.4	419.8	101.2	160.0	34.3	30.8	96.9	139.5	46.3	117.6	84.5	73.7	77.5	52.8	47.1	34.8	7.0	3.7	2.7	22.1	99.2	193.0	67.7	34.1
Phe		781.1	1133.7	824.5	491.3	136.7	129.4	217.5	324.6	252.1	178.0	505.1	572.1	169.2	137.6	255.8	260.0	32.8	30.9	31.2	28.6	215.9	328.9	103.1	453.1
Ant		95.3	74.2	14.5	39.3	12.6	10.7	109.2	3.3	64.0	90.6	149.4	127.5	8.7	6.5	19.2	21.5	5.1	14.0	3.0	2.2	20.7	201.0	72.4	35.5
Flt	4	171.5	152.4	311.0	182.5	20.2	12.4	156.6	136.2	42.2	100.8	459.9	404.0	102.6	74.4	73.7	26.6	16.6	8.1	24.4	51.1	72.4	85.2	136.2	173.2
Pyr		186.1	202.0	81.5	36.5	27.9	24.0	192.0	151.7	60.0	20.7	180.8	302.2	209.6	226.3	42.8	96.2	21.1	11.2	15.0	9.2	147.0	181.0	41.7	270.9
BaA		40.9	11.0	153.0	22.5	1.0	1.7	63.4	131.1	1.4	7.9	41.3	104.4	22.9	22.1	1.0	31.7	1.7	13.3	4.3	2.2	51.7	22.3	59.5	80.0
Chr		83.0	67.7	51.8	55.0	5.3	11.2	34.5	320.9	3.7	25.4	66.8	173.1	20.7	46.9	0.6	194.9	3.2	34.2	6.5	8.7	75.2	95.4	47.2	50.3
BbF	5	3.6	11.9	37.0	1.1	0.4	0.9	0.6	<LOD[a]	0.3	<LOD	<LOD	<LOD	0.4	<LOD	0.1	1.0	<LOD	<LOD	<LOD	0.2	15.2	8.6	29.8	34.1
BkF		10.9	6.8	27.2	22.5	0.4	0.9	<LOD	2.3	0.5	<LOD	1.2	0.2	<LOD	0.7	0.8	<LOD	0.3	0.2	1.8	<LOD	21.3	10.8	31.8	50.3
BaP		2.6	3.7	1.6	1.1	0.3	0.5	0.6	0.5	0.1	<LOD	0.4	<LOD	0.1	<LOD	0.2	0.1	0.0	0.0	<LOD	0.0	2.9	5.1	0.8	7.6
DahA		1.5	0.5	29.6	0.3	0.2	0.4	3.1	<LOD	<LOD	1.6	<LOD	1.0	<LOD	<LOD	0.3	12.3	<LOD	2.1	<LOD	1.9	25.3	2.9	32.8	3.0
BghiP	6	40.1	3.1	<LOD	2.8	0.0	0.1	45.1	18.7	<LOD	<LOD	2.0	2.1	1.1	11.0	0.2	1.3	1.7	2.1	<LOD	0.1	23.8	6.3	6.8	218.0
IP		2.6	2.2	19.7	5.6	0.1	0.6	40.0	14.1	12.3	17.5	5.9	<LOD	<LOD	1.8	0.1	2.0	2.1	1.8	1.1	0.7	81.4	1.7	20.5	32.6
∑2-Ring PAHs		1938.1	1870.7	1466.3	1718.3	98.6	161.8	563.0	840.9	714.0	990.3	357.7	587.4	405.1	608.9	356.0	461.7	25.3	27.3	22.7	38.2	124.9	205.0	90.3	225.1
∑3-Ring PAHs		1191.7	1884.7	1052.6	772.1	195.7	179.4	535.9	589.1	490.5	432.4	841.6	885.6	330.8	280.9	335.9	372.6	55.3	57.3	42.2	57.7	426.3	794.2	287.3	549.3
∑4-Ring PAHs		481.6	433.1	597.4	296.4	54.5	49.3	446.4	739.9	107.2	154.8	748.8	983.7	355.9	369.7	118.1	349.5	42.7	66.8	50.2	71.1	346.3	383.8	284.6	574.5
∑5-Ring PAHs		18.6	23.0	95.4	25.0	1.2	2.7	4.3	2.8	1.0	1.6	1.6	1.2	0.5	0.7	1.5	13.4	0.3	2.3	1.8	2.1	64.7	27.4	95.2	94.9
∑6-Ring PAHs		42.7	5.4	19.7	8.4	0.2	0.7	85.1	32.8	12.3	17.5	7.9	2.1	1.1	12.8	0.4	3.4	3.8	3.9	1.1	0.8	105.2	8.0	27.4	250.6
∑LMW-PAHs[b]		3129.8	3755.3	2518.9	2490.4	294.4	341.2	1098.9	1430.0	1204.5	1422.7	1199.3	1473.0	735.9	889.8	691.9	834.3	80.6	84.6	64.9	95.9	551.2	999.2	377.6	774.4
∑HMW-PAHs[c]		542.9	461.4	712.5	329.9	55.8	52.7	535.8	775.5	120.5	173.9	758.3	987.1	357.6	383.2	120.0	366.2	46.8	73.0	53.0	74.0	516.1	419.2	407.2	919.9
∑C-PAHs[d]		145.0	103.9	320.1	108.1	7.6	16.3	142.1	468.9	18.3	52.5	115.6	278.8	44.2	71.6	3.2	242.1	7.4	51.5	13.6	13.7	272.9	146.7	222.4	257.8
∑PAHs[e]		3672.7	4216.8	3231.4	2820.2	350.2	393.9	1634.7	2205.5	1324.9	1596.6	1957.5	2460.0	1093.5	1273.0	811.9	1200.5	127.4	157.6	117.9	169.9	1067.4	1418.4	784.7	1694.3

[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); [e] Sum of 16 USEPA PAHs.

Table C-10. Seafood consumption[a] data (g/day) for the Bangladeshi coastal residents (adults and children) in winter and summer.

Coastal area	Adults[b]		Children[b]	
	Winter	Summer	Winter	Summer
Cox's Bazar	106	107	51	54
Chittagong	111	107	56	59
Bhola (Meghna Estuary)	85	86	41	46
Sundarbans	88	92	44	46

[a] Data obtained from dietary questionnaire survey; [b] Average body weight was 60 kg for adults (≥18 years) and 25 kg for children (6−17 years).

“Make the environment pollution-free earlier, make your life healthier and happier.”

- Md. Habibullah-Al-Mamun