論　文　要　旨 Dissertation Abstract

平成 29 年 3 月 1 日
Date (YY/MM/DD): 2017/03/01

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論文題目 Dissertation Title	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 Monitoring and Assessment of Persistent Organic Pollutants (POPs) in Water, Sediment, and Seafood from the Coastal Areas of Bangladesh

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.

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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 C4–14-PFCAs (perfluoroalkyl carboxylates) and C4, C6, C8, and C10-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

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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.

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