Asian Mussel Watch Program: Sources and Distribution of Polybrominated Diphenyl Ethers (PBDEs) and Organochlorines Contamination in Coastal Waters of Asian Countries

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Introduction

Polybrominated diphenyl ethers (PBDEs) are widely used as flame retardants and have applications in a variety of commercial and household products. They are of environmental concern due to their persistence, potential for bioaccumulation and widespread distribution via atmospheric transport, and possible adverse effects in wildlife and humans.[1] Studies on environmental behavior of PBDEs are chiefly derived from Europe and North America and there are only a few reports from Asia. As a result, there is an urgent research need to identify Asian sources of PBDEs as well as to quantify emissions and document their potential environmental fate in this region.

Bivalves such as mussels have been suggested as suitable bioindicator for monitoring trace toxic contaminant levels in coastal waters.[2] In particular, green mussels (*Perna viridis*) are widely distributed in the Asian coastal waters and recognized as commercially valuable seafood in this region. The present study involves coastal monitoring of PBDEs and organochlorine compounds (OCs) using sentinel organisms such as mussels as bioindicators to ascertain the quality of coastal waters in the Asia-Pacific region and to identify Asian sources of PBDEs.

Materials and Methods

Samples

Green mussel and blue mussel (*Mytilus edulis*) were collected from various locations in the Asian countries such as India, Indonesia, China, Hong Kong, Japan, Cambodia, Vietnam and Philippines from 1998 to 2004. The frozen mussel samples were thawed and biometric measurements were made. After shucking the whole soft tissues of mussels from each location, they were pooled, homogenized, transferred to polyethylene bags and frozen at -20°C until chemical analysis.

Chemical analysis

Analysis of PBDEs was performed following the procedure described by Ueno *et al*.[3] with slight modification. OCs including polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane and its metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), chlordane related compounds (CHLs), and hexachlorobenzene (HCB) were analyzed following the method described by Kajiwara*et al*.[4]

Results and Discussion

Contamination status of PBDEs

PBDE concentrations detected in the soft tissue homogenates of mussel samples from the coastal waters of Asian region are shown in Figure 1. PBDEs were detected in all the mussel samples analyzed in this study, indicating widespread contamination by these compounds in the coastal waters of Asia. Contamination status of PBDEs in mussels varied depending on countries and the local sites of sampling. Total concentrations of PBDE congeners

ranged from 0.82 to 120 ng/g lipid weight, with the highest levels found in mussel samples from Hong Kong. The Pearl River Delta in Hong Kong has a number of electronic and telecommunication industries, as well as a number of private manufacturing operations which have transformed this delta into one of the fastest growing industrial manufacturing areas in the world.[5] Estimates suggest that one out of three computers manufactured in the world are assembled in this region.[6] As PBDEs are commonly incorporated into polymers for use in electronic components, the high levels of PBDEs in mussel tissues from Hong Kong may be due to the discharge of effluents derived from materials used in the production or the dismantling of electronic equipment. Our previous study also demonstrated high PBDE levels in cetaceans from Hong Kong when compared to cetaceans from Asia-Pacific region.[7] The results of the present study suggest the existence of considerable sources of PBDEs in and around Hong Kong in Asia. Comparing the present study with the only available study on PBDEs in mussels from Asia (Singapore), PBDE levels in green mussels from Singapore were high and varied from 39 to 430 ng/g lipid weight.[8] The reason could be due to the proximity of the sampling locations to industrial activities. While, PBDE levels in mussel tissues from Asia-Pacific region were comparable to the levels reported for blue mussel tissues from European region like Belgium[9], Norway[10], Greenland[11] and Denmark[12] they were far lower than in blue mussel tissues from San Francisco Estuary, USA.[13]

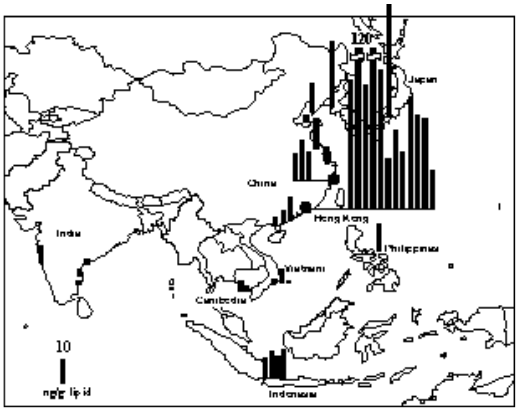

Figure 1. PBDE concentrations in mussels from coastal waters of some Asian countries.

Congener profiles of PBDEs

Of the 10 congeners analyzed, a total of nine congeners from di- to deca-BDE were identified in mussel samples from the Asia-Pacific region. BDE 3 (mono-BDE) was not found above the detection limit of analysis for all the samples. Among the PBDE congeners found in the environment, BDE 47, BDE 99 and BDE 100 usually make up approximately 90% of the total level of PBDEs in aquatic biota and BDE 47 constitutes more than 60% of the total level.[14] In the present study, BDE 209 (deca-BDE) was detected in mussel samples from China, Hong Kong and Indonesia at varying levels. BDE 209 because of its high log K_{ow}, is primarily bound to particulate organic carbon and mussels which are filter feeder organisms accumulate contaminants from suspended particles and from phytoplankton.[15] Mussels in the present study were not depurated before analysis thus the detection of BDE 209 could be due to the presence of sediment in the gut of mussels. In China, deca-BDE is one of the most produced flame retardants and high levels of deca-BDE were detected in the riverine and coastal sediments of South China.[6] The detection of BDE 209 in mussel samples from these regions suggests the prevalence of deca-BDE in the marine environment and further suggests that mussels are a good matrix for monitoring PBDEs in the aquatic environment. Studies carried out on PBDEs in mussels have concentrated mainly on tetra to hexa-BDE congeners. Considering the above mentioned facts, for calculation of total concentration of PBDEs the sum of mono-BDE to hepta-BDE was considered. Excluding deca-BDE congener values from the samples, the congener profile was dominated by BDE 47 followed by BDE 99 > BDE 100 > BDE 28 ≥ BDE 153 and BDE 154 as had been observed in various studies conducted elsewhere Figure 2.

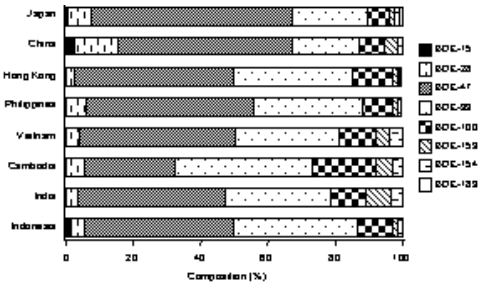

Figure 2. PBDE congener profiles in mussels from coastal waters of some Asian countries.

Contamination status of OCs

Among the OCs analyzed in this study, DDTs were the highest and those of the other OCs were in the order of PCBs > CHLs > HCHs > HCB. Similar trend was observed in the previous mussel watch program.[16] DDT concentrations as high as 40,000 ng/g lipid weight, were found in mussel samples from the coastal waters of South China. Elevated concentrations of DDT have been reported in mussels from China earlier.[16,17] The higher ratio of *p,p'*-DDT in mussel samples from South China may indicate the presence of current emission sources of DDT in China, even though usage of DDT was officially banned in 1983. Levels of PCBs in mussel sample from Osaka, Japan were the highest

(2,200 ng/g lipid weight) suggesting that emission sources of PCBs still existed in Japan even though PCBs production was prohibited in 1972. For HCH levels as high as 230 ng/g lipid weight, were found in mussel samples from India. India has been the largest user of technical HCH in the world and though the usage had been banned in agriculture since 1983, it is still used for public health purposes and on certain food crops.[18]CHLs which are largely used as termiticide were the highest for mussel samples from Japan (900 ng/g lipid weight).

In conclusion, this is the first time that the available mussel species in these regions have been used for screening PBDEs. The results suggest that mussels are good matrixes for monitoring the momentary concentrations of PBDEs in the aquatic environment. Further, from the results it can be concluded that there exist significant sources of PBDEs in Hong Kong, DDT in China, PCBs and CHLs in Japan and HCHs in India. The contaminant residues in mussels pose a great concern on human health as mussels are important seafood in the Asian developing countries. Given the increasing production and usage of PBDEs, more emphasis should be given for continuous monitoring of these compounds.

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