

学 位 論 文 要 旨

Distribution and Debromination of Polybrominated Diphenyl Ethers (PBDEs) in Tropical Asian Countries

(熱帯アジア諸国における臭素化ジフェニルエーテル類 (PBDEs) の分布と
脱臭素化)

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Polybrominated diphenyl ethers (PBDEs) are extensively used as additive flame retardants in polymers for the manufacture of many consumer products. Tetra- to hepta-BDE congeners have been regulated under the Stockholm Convention because of their persistence, toxicity, potential to bioaccumulate and their capacity for long-range environmental transport. However, deca-BDE has not been regulated. Although there have been indications of the debromination of deca-BDE to lower and more toxic congeners, the evidences have been limited. Thus, the present study emphasized on providing evidence on the debromination of deca-BDE through environmental observation.

Another focus of this study is the relationship between socio-economic setting and pollution. Tropical Asian countries have experienced unprecedented population and rapid economic growths in the last decade. These are driving factors in the consumption of PBDE-laden goods, e.g. electrical and electronic equipment. Consequently, high volumes of PBDE-containing wastes may be generated in the region. Some tropical Asian countries may be recipients of electronic waste. Thus, there is a need to reveal the environmental distribution of PBDEs, and to understand the transformation processes of deca-BDE in these environments.

This study investigated the distribution, transport and fate of PBDEs, including debromination in eight tropical Asian countries and their highly urbanized cities, such as Lao PDR (Vientiane), Cambodia (Phnom Penh), Vietnam (Hanoi and Can Tho), India (Kolkata, Chennai and Mumbai), Indonesia

(Jakarta), Thailand (Bangkok), the Philippines (Metro Manila) and Malaysia (Kuala Lumpur). The study covered three topics: PBDEs in the leachates of municipal solid waste dumping sites (MSWDS), sedimentary PBDEs in the canals or rivers of urban areas, and understanding the historical trends of PBDE emissions in tropical Asian coastal environments using sediment cores. Twenty three to 46 PBDE congeners were measured in 24 leachate samples from 10 MSWDS, and in 44 surface sediment samples collected from rivers, and coastal waters among the 8 countries. Forty five PBDE congeners were measured in sediment cores from Manila Bay, Philippines and the upper Gulf of Thailand.

The total PBDE concentrations in the leachates from the MSWDS ranged from 3.7 to 133,000 ng/L. The total PBDE concentrations in the sediments ranged from 0.83 to 3,140 ng/g dry wt. The PBDE concentrations in most of the leachates from the MSWDS, and the sedimentary BDE-209 concentrations showed a trend toward higher concentrations in the more industrialized Asian countries, and were similar to or higher than those reported for highly urbanized and industrialized countries worldwide. Thailand, Cambodia, India, the Philippines and Malaysia are possible hot spot areas of PBDE pollution.

The PBDE congener profiles in the leachates and in the sediments reflected the composition of the technical penta-, octa- and deca-BDE products, and the higher importation of technical deca-BDE than octa- and penta-BDE products into Asia. The occurrence of congeners which are not contained, or in trace concentrations, in the technical PBDE products (e.g., BDEs 208, 207, 206, 202, 188, 49, 17/25, 8, 1) was observed in most of the leachate samples and surface sediments, suggesting the occurrence of debromination of BDE-209 in the MSWDS and in the sedimentary environments of tropical Asian countries. Increasing ratios of BDE-202/209, 206/209, 207/209 and decreasing % BDE-209 down the core layers (relative to their proportions in the technical deca-BDE mixtures) provided further evidence in the anaerobic debromination of BDE-209. Debrominated higher PBDE congeners can be the source of lower but more bioavailable and toxic congeners to the environment.

PBDEs were predominantly found in the adsorbed phase of the leachates. Partitioning of PBDEs in the dissolved phase in some leachates was associated with the presence of dissolved organic matter (DOM). Dissolution of PBDEs with DOM in the leachates may facilitate the transport of PBDEs from the MSWDS to the aquatic environments.

The historical trends of PBDE emissions in typical tropical Asian environments were captured by the sediment cores. Increasing concentrations of total PBDEs toward the surface suggested increasing consumption of PBDEs. The doubling times of total PBDEs (6–7 years) and BDE-209 (6–7.5 years) were comparable to or faster than those reported in other water bodies around the world.