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Chapter 18

Contamination by Persistent Toxic Substances in the Asia-Pacific Region

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Abstract

This chapter reviews studies conducted in our laboratory at CMES, Ehime University on various aspects of contamination by persistent toxic substances (PTS) in Asia-Pacific region. The sources, distribution, environmental fate, temporal trends and toxic impacts of PTS on wildlife and humans were examined using archived samples from our environmental specimen bank (*es-BANK*). The hot spots of PTS pollution were found in the areas where they have been heavily used, e.g., elevated HCH residue levels were found in samples from India and South China, while pollution by DDTs was found to be high in China and Vietnam. PBDEs exhibited higher concentrations in coastal waters of Korea and Hong Kong and areas around East China Sea, suggesting the presence of PBDE sources in highly industrialized zones. Our recent study on temporal trends of PBDEs in this region clearly demonstrated a substantial increase in PBDE levels in small cetaceans collected from Hong Kong coastal waters. The role of extensive manufacturing of computers and other electronic appliances as well as dumping of e-waste in some East Asian countries as substantial sources of brominated flame retardants such as PBDEs should be elucidated in future. Based on our monitoring surveys involving different environmental media and biological samples, the transport behavior of PTS could be characterized as "local" and "global". DDTs and PCDD/Fs have less potency for long range atmospheric transport, and their sources are, therefore, located in the proximity of areas where they are used/released. On the other hand, HCHs and, to a lesser extent, PCBs and PBDEs exhibit relatively high ability for transport through atmosphere and their contamination may extend globally in future. As far as toxic impacts of PTS are concerned, the open dumping sites for municipal wastes

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in Asian developing countries are potential sources of various chemical contaminants. Residue levels of dioxins and related compounds were higher in soils, human breast milk and bovine milk from dumping sites when compared to reference sites. Possible impacts of PTS on health of people, especially those living in and around these dumping sites, are of great concern and deserve systematic and long-term investigations. Considering these critical issues of PTS pollution, it is clear that continued and constant efforts should be made to deal with the environmental problems caused by the contaminants not only in developed nations but also in developing countries.

18.1. Introduction

As a closing chapter of the book dealing with various important issues on persistent organic pollutants (POPs), this article reviews the outcomes of comprehensive investigations conducted in our laboratory at CMES, Ehime University over the last three decades on the distribution, sources, transport behavior and fate, pollution trends and toxic impacts of the bioaccumulative persistent toxic substances (PTS) in Asia-Pacific region with a particular emphasis on the developing countries. Results of multi-media monitoring studies were complied and discussed to provide in-depth understanding on the various issues of PTS contamination in both ambient environment and animals, including humans. The prominent contamination by PTS was found in the region where they have been heavily used. The East Asian region is probably a potential source of pollution, particularly by the new contaminants such as polybrominated diphenyls ethers (PBDEs). This group of contaminants, together with polychlorinated biphenyls (PCBs), exhibited slow decreasing or even increasing trends in some highly industrialized areas in Asian developing countries, suggesting the necessity for long-term monitoring. The open dumping sites for municipal wastes in major cities are significant sources of many toxic chemicals, and these areas are probably one of the challenges for future research due to the long-term impacts on the environmental quality and human health. The formation of dioxins and related compounds (DRCs) in such dumping sites and their elevated residue levels found in breast milk of residents living in and around the sites warrant long-term impacts of dioxins on the next generations. Comprehensive and long-term monitoring programs are urgently needed with close collaboration and proper capacity building in the local areas in developing countries in order to mitigate the PTS emission and their risk on ecosystems and human health.

The last decades of the 20th century have witnessed a steady increase in the pace and extent of various environmental problems such as biodiversity loss, changes in water quality, land use and climate. Among these, the issue of environmental pollution caused by PTS has become increasingly important because their usage in large quantities have resulted in many negative impacts including environmental deterioration and health effects on wildlife. In particular, many PTS are known to pose endocrine disrupting effects, which are probably the cause for various reproductive abnormalities observed in wildlife species (Colborn et al., 1996). Among a variety of endocrine disrupters, chemicals with severe toxicity, high accumulation potential in the body and persistence in the environment are the most hazardous for wildlife. Organochlorine (OC) compounds—the typical class of chemicals meeting the above conditions—such as DDTs, hexachlorocyclohexanes (HCHs), chlordane related compounds (CHLs) and PCBs have received considerable attention due to their potential to degrade the environmental quality and to pose ecological risk. Mass production and consumption of these OCs started after the Second World War. DDTs, HCHs and CHLs have been heavily used as insecticides for agriculture and public health purposes including malaria eradication. PCBs are mainly used for industrial purposes such as dielectric fluids in transformers and capacitors. Although many developed nations prohibited production and usage of certain OCs in the 1970s and 1980s due to a deep concern regarding their toxic impact on humans and wildlife, they are still in use in some developing countries.

Dioxins and related compounds including polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and coplanar PCBs should also be paid particular attention due to their highest toxicity among the OCs investigated. They are "unintentionally" generated by refuse incineration and/or byproducts during the manufacturing processes of some other chemicals. According to a comprehensive national survey by the Japanese Ministry of Environment (1999), elevated contamination of DRCs were widely detected in fish-eating animals such as kites and cetaceans collected throughout Japan. In developed countries, residue levels of DRCs in human breast milk have been reported to decrease recently due to the implementation of highly efficient incinerators and strict regulations on the production and usage of hazardous chemicals (LaKind et al., 2001). On the other hand, in developing countries, environmental pollution by DRCs may still increase because of improper management controlling the release of these contaminants. Although it has been suspected that the residents in Asian developing countries have been exposed to relatively high levels of DRCs, there is a lack of information on human exposure to these contaminants.

In addition to OCs, PBDEs, the popular brominated flame retardants, are now a worldwide problem even in remote areas, and Asia-Pacific region is surely no exception (Ikonomou et al., 2002; Birnbaum and Staskal, 2004; Ueno et al., 2004). PBDEs are structurally similar to PCBs and DDT and, therefore, their chemical properties, persistence and distribution in the environment follow similar patterns. Studies on the environmental behavior of PBDEs are chiefly derived from Europe, North America and the Arctic. Despite the usage of vast amounts of these compounds in Asia-Pacific region, there is a paucity of data on the prevalence of PBDEs in Asian environment. Studies are necessary to identify Asian sources of PBDEs as well as to quantify emissions and document their potential environmental fate in this region.

In the previous chapters, various issues of PTS contamination such as magnitude of pollution, environmental transport and fate, features of bioaccumulation in organisms as well as trends of contamination have been discussed with in-depth examples of case studies conducted in particular countries in the Asia-Pacific region. These chapters have provided both general and concrete concept of the ultimate fate of PTS in local perspectives and addressed a number of issues of PTS contamination that specifically exist in each country/area.

Results of multi-media monitoring surveys conducted in our laboratory in collaboration with Asian environmental scientists were compiled in this chapter to discuss the fate of PTS in both ambient environment and biota, including humans, from regional and global point of view. The Asia-Pacific region comprises a number of developing and under developed countries, where considerable quantities of PTS are currently used or have been used until very recently. Therefore, the issue of temporal trends of PTS pollution in this region is critically important. This chapter also provides in-depth analysis of temporal trends of PTS residues in various environmental compartments with the purpose of tracing history, predicting future trends of PTS pollution and exploring new clues for understanding their toxic impacts on environment and human health. We fully utilized archived samples in the Environmental Specimen Bank for Global Monitoring (*es-BANK*) at Ehime University which has a wide array of tissues and organs of about 900 species with 80,000 specimens of wildlife and environmental samples collected throughout the world during the last 40 years.

18.2. Distribution and sources

In addition to the multi-media monitoring of the environmental samples, the use of animal tissues and humans as bioindicators has been proved to be a suitable concept to address various important issues of PTS pollution (Tanabe and Subramanian, 2006). Since bivalves have been suggested as suitable bioindicators for monitoring trace toxic contaminant levels in coastal waters (Goldberg, 1975), we have been using mussel as a sentinel animal to elucidate the pollution sources in the Asia-Pacific region (Sudaryanto et al., 2002; Tanabe et al., 2000; Monirith et al., 2003b; Ramu et al., 2005). The survey conducted by Monirith et al. (2003b) revealed that DDT residue levels were highest in mussel samples from the coastal waters of South China (Fig. 18.1). The higher proportion of *p,p′*-DDT, the main component in the formulation, in mussels may indicate the presence of current emission sources of DDT in China, despite its usage was officially banned in 1983. HCH levels were found to be the

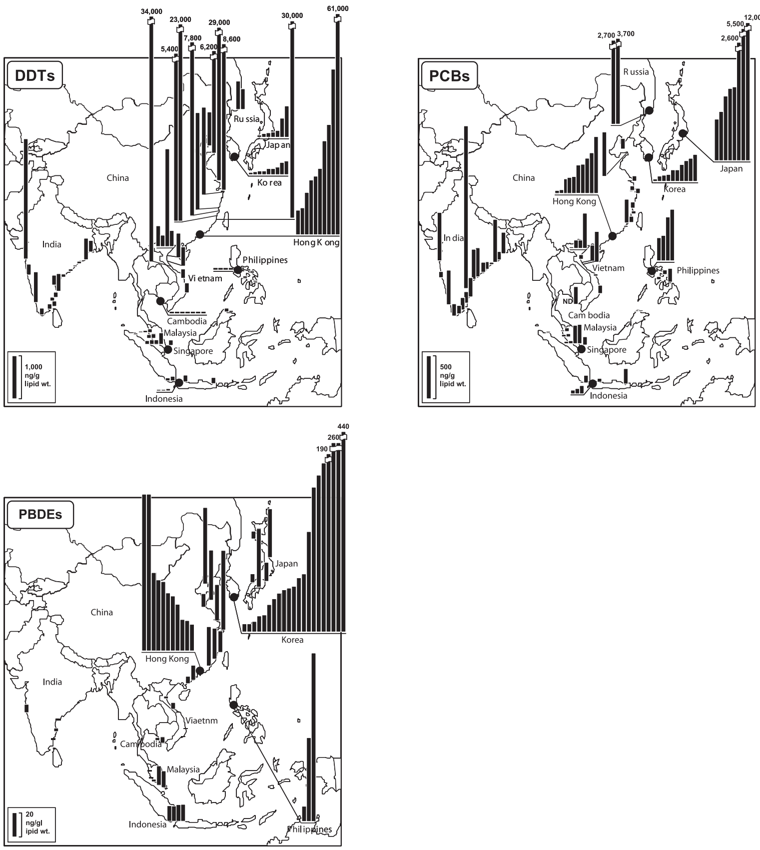

Figure 18.1. DDT, PCB and PBDE concentrations in mussels from coastal waters of some Asian countries.

highest in mussel samples from India, the world's largest consumer of technical HCHs, reflecting continuous usage for public health purposes and on certain food crops (Li et al., 1998; Li, 1999). According to our previous studies on water pollution, considerable contamination by OC insecticides such as HCHs and DDTs was found in developing countries in the Asian tropics (Iwata et al., 1994). On the contrary, PCB levels in mussels in developed nations, including Japan, were clearly higher than those in developing countries (Fig. 18.1). Using resident and migratory birds as bioindicators for monitoring OC contamination, high concentrations of OC insecticides and PCBs were found in birds collected at stopover/wintering areas of tropical Asian developing countries and Russia/Japan, respectively (Tanabe et al., 1998; Kunisue et al., 2002a; Minh et al., 2002). To substantiate our findings with further evidence on pollution sources of OCs, we collected human breast milk from Asian countries and determined residue levels of contaminants. As a result, PCB levels in human breast milk were found to be higher in developed nations like Japan than in developing countries such as China, India, Cambodia and Indonesia (Fig. 18.2). On the other hand, OC insecticides in human breast milk showed apparently higher concentrations in developing countries than in developed nations. Considering all these observations, we can say that, from a global point of view, major emission sources of PCBs are located in developed nations, while that of DDTs and HCHs are in the developing countries.

Noteworthy, widespread contamination by PBDEs in the coastal waters of Asia became apparent with the highest concentration in mussel samples from Hong Kong (Ramu et al., 2005) (Fig. 18.1). Another study focused on cetaceans (Kajiwara et al., 2006) also showed the highest PBDE concentrations in samples from Hong Kong, suggesting that developing nations may also have significant pollution sources of PBDEs (Fig. 18.3). 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 (Zheng et al., 2004). Estimates suggest that one out of three computers manufactured in the world are assembled in this region (Mai et al., 2005). As PBDEs are commonly incorporated into polymers for use in electronic components, the high levels of PBDEs in wildlife from Hong Kong may be due to the discharge of effluents derived from materials used in the production or the dismantling of electronic equipments, the so-called "e-waste". In addition, our recent survey on more than 20 sites along Korean coastal waters (Ramu et al., 2007) revealed that natural blue mussels contained very high concentrations

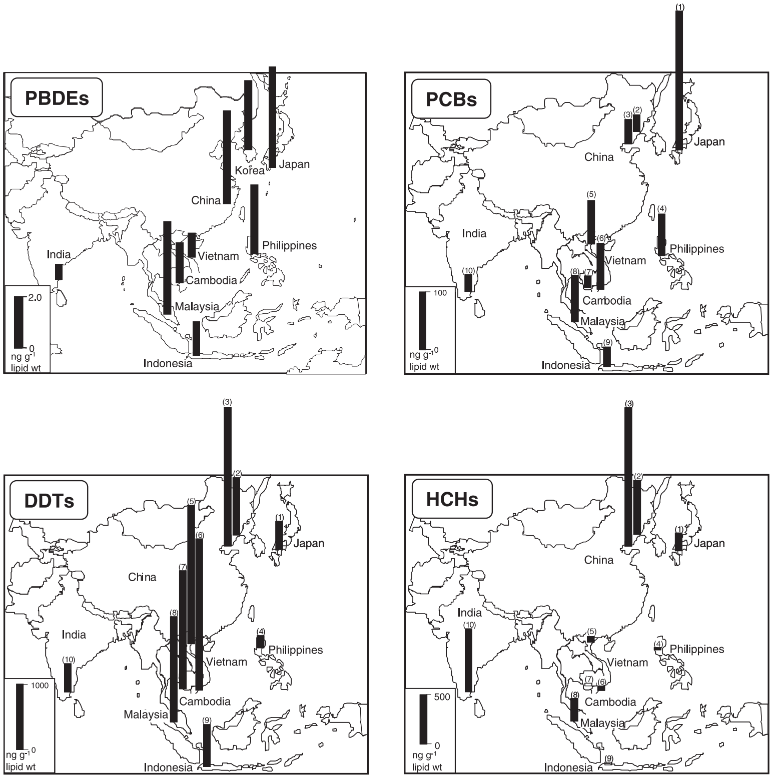

Figure 18.2. Concentrations of PBDEs, PCBs, DDTs and HCHs in human breast milk collected from general public in Asian countries. (1) Ehime, Japan (Kunisue et al., 2004c), (2) Shenyang, China (Kunisue et al., 2004a), (3) Dalian, China (Kunisue et al., 2004a), (4) Quezon, Philippines (Kunisue et al., 2002b), (5) Hanoi, Vietnam (Minh et al., 2004), (6) Hochiminh, Vietnam (Minh et al., 2004), (7) Phnom Penh, Cambodia (Kunisue et al., 2004b), (8) Penang, Malaysia (Sudaryanto et al., 2005b), (9) Jakarta, Indonesia (Sudaryanto et al., 2006), (10) Palaverkadu, India (Kunisue et al., 2002b). Data for PBDEs were from Sudaryanto et al. (2005a).

of PBDEs (mean = 91 ng g^{-1}; range = 6.6–440 ng g^{-1} lipid wt; Fig. 18.1), which are among the highest levels ever reported for the mussels from Asia-Pacific. Elevated residues of PBDEs in mussels from Korea indicate that this region is a potential source of these new chemicals in eastern Asia.

To elucidate whether the DRC pollution sources are also present in developing countries, human breast milk samples were employed for chemical analysis. As summarized in Fig. 18.4, significant levels of DRCs were detected in human breast milk from developing countries,

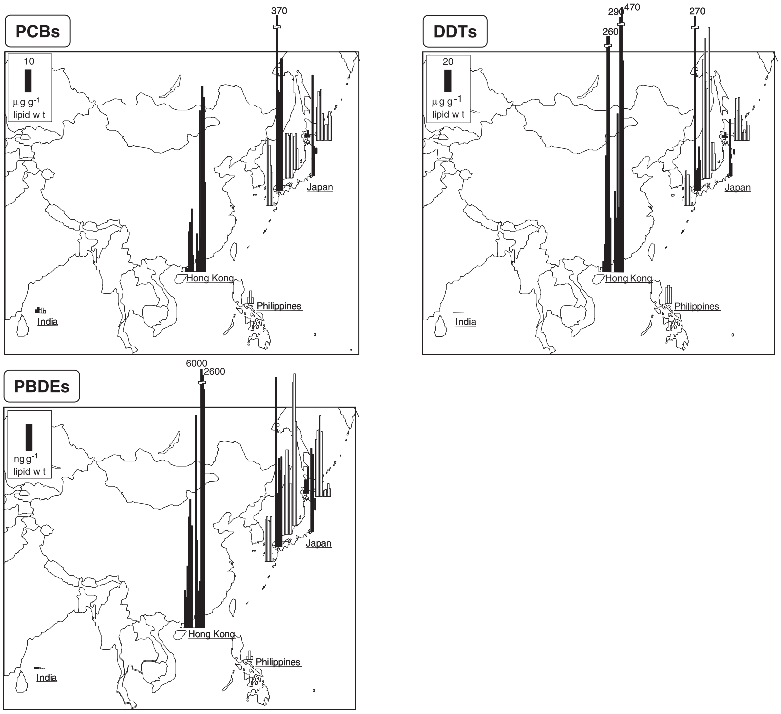

Figure 18.3. Geographical distribution of PCBs, DDTs and PBDEs in Asian cetaceans. Black and white bars indicate coastal and offshore species, respectively.

indicating considerable contamination in some developing countries as well as in developed nations. Our further survey on DRC contamination in soils revealed critical emission sources in the open dumping sites of municipal wastes in Asian developing countries (Minh et al., 2003). A variety of municipal wastes are dumped there continuously and burnt under low temperature spontaneously and/or intentionally generated fire, implying DRCs formation by such uncontrolled combustion with very little waste management. Additionally, PCBs can leach out from old electric appliances dumped there. When comparing PCDD/F levels in various soil types on global terms, open dumping sites in Asian developing countries showed apparently higher concentrations than general background soils and comparable levels to the dioxin-contaminated sites in developed nations so far reported (e.g. de Jong et al., 1993; Lorber et al., 1998). Toxic equivalents (TEQs) concentrations detected in soils from open dumping sites in Asian developing countries exceeded the environmental quality standards set forth by the Japanese

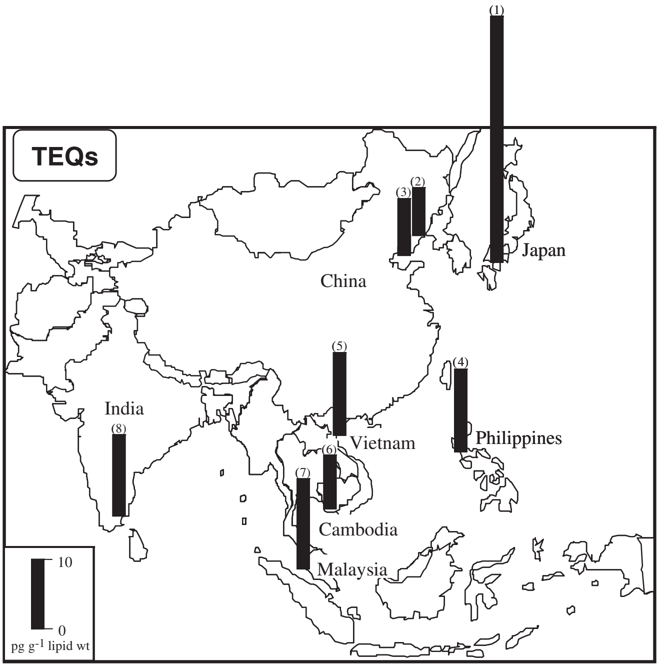

Figure 18.4. TEQs of DRCs (PCDDs, PCDFs, coplanar PCBs) in human breast milk collected from general public in Asian countries. (1) Ehime, Japan (Kunisue et al., 2004c), (2) Shenyang, China (Kunisue et al., 2004a), (3) Dalian, China (Kunisue et al., 2004a), (4) Quezon, Philippines (Kunisue et al., 2004d), (5) Hanoi, Vietnam (Kunisue et al., 2004d), (6) Phnom Penh, Cambodia (Kunisue et al., 2004d), (7) Penang, Malaysia (Sudaryanto et al., 2005b), (8) Palaverkadu, India (Kunisue et al., 2004d).

government and the US Department of Health. More importantly, in Asian developing countries, a stratum of impoverished people earn a living by collecting dumped recyclable and valuable resources and form slums in and around the dumping sites. Since they spend most of the days there, risks on their health through the exposure to significant amount of dioxins and other anthropogenic chemicals are of particular concern. Figure 18.5 illustrates the mean and range concentrations of DRCs in human breast milk collected from residents around dumping sites and reference sites in India, Cambodia and Vietnam (Kunisue et al., 2004d). In these countries, higher levels were observed in dumping sites, particularly, the concentrations in Indian dumping sites were significantly higher than those in control sites, which was not observed in the cases of Cambodia and Vietnam. This result prompted us to consider the presence of extraordinary exposure pathway of DRCs in

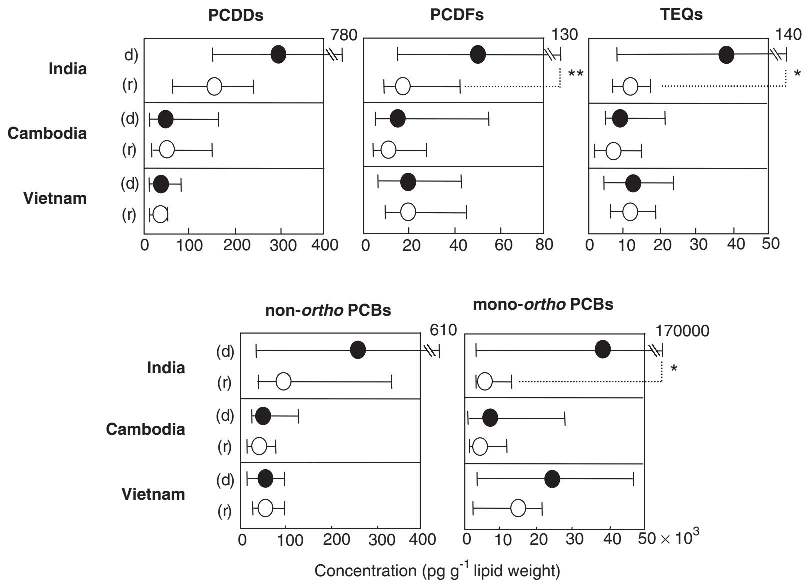

Figure 18.5. Comparison of the concentrations of DRCs in human breast milk from dumping (d) and reference (r) sites. The circles and bars represent mean and range values, respectively. *$p<0.05$, **$p<0.01$, Mann-Whitney U test.

the dumping sites of India. Although, in Cambodia and Vietnam, domestic livestock are not reared around the dumping sites, residents around the Indian dumping site consume milk from the buffaloes and cows that graze daily on the garbage in the waste dumping sites. To elucidate whether bovine milk is a potential source of DRCs for the residents around the dumping site in India, residue levels of these contaminants in milk from the buffaloes and cows there were examined (Kunisue et al., 2004d). As expected, concentrations of DRCs in bovine milk collected from the dumping site were significantly higher than those from the reference site (Fig. 18.6), indicating such a dairy operation led to significant accumulation of DRCs in residents around dumping site. For assessing the health risk, we estimated daily intakes by infants based on dioxin, HCH and DDT concentrations in human breast milk (Fig. 18.7). The estimated daily intakes of dioxins by infants in all the four developing countries exceeded tolerable daily intake (TDI) proposed by World Health Organization (WHO) (Van Leeuwen et al., 2000). Epidemiological studies coupled with monitoring of contaminants should be conducted on dumping site populations in developing countries.

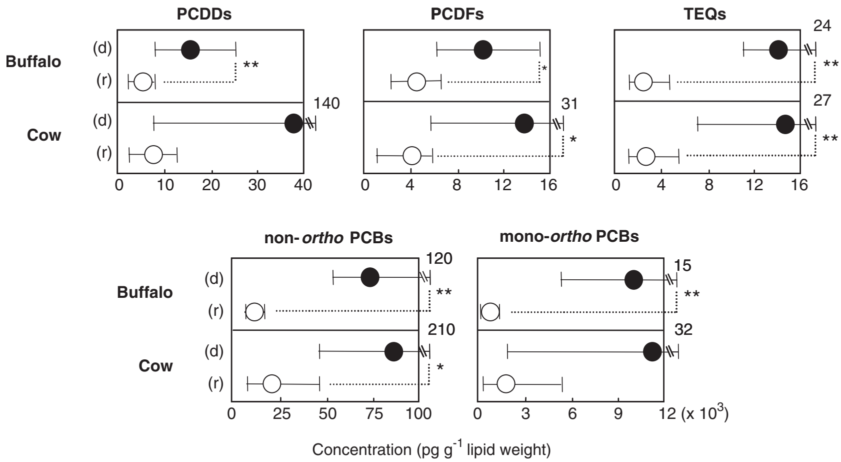

Figure 18.6. Comparison of the concentrations of DRCs in bovine milk from dumping (d) and reference (r) sites. The circles and bars represent mean and range values, respectively. *$p<0.05$, **$p<0.01$, Mann-Whitney *U* test.

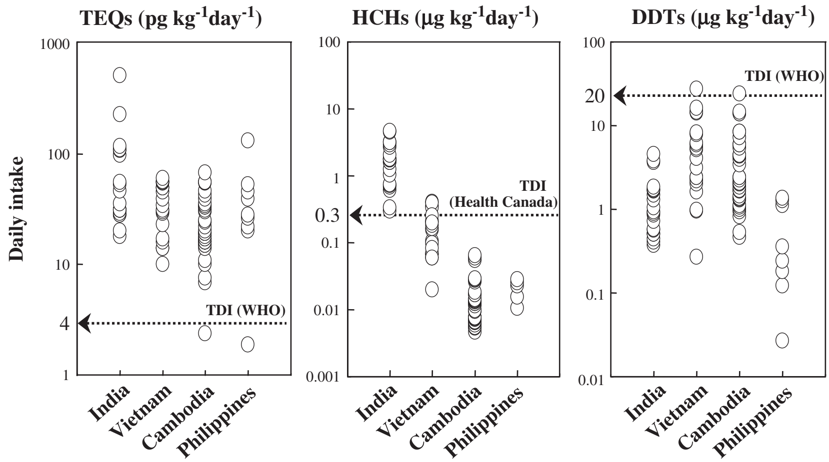

Figure 18.7. Estimated daily intake of OCs by infants in Asian developing countries.

18.3. Transport behavior and fate

Multi-media monitoring studies not only helped increase knowledge of the status of contamination, but also provided in-depth insights into the transport behavior and fate of PTS in tropical ecosystems in Asian developing countries. On a global scale, the fate of persistent OCs in open oceans was different from that in coastal environment (Iwata et al., 1993, 1994; Tanabe et al., 1994). Open oceans play the role as a final sink for PTS, and cold waters from high latitudes like Arctic Ocean, which are located far from pollution sources, serve as significant reservoirs (Iwata et al., 1993). On the other hand, pronounced contamination by OC insecticides was observed in coastal waters of Asian countries as a result of extensive usage for agriculture and public health purposes (Iwata et al., 1994). The latitudinal distribution of persistent OCs such as PCBs, DDTs, HCHs in sediments from the coastal waters of Asian countries is a typical example for understanding the transport behavior of PTS on a smaller scale (Iwata et al., 1994). Iwata et al. (1994) suggested a concept of using the relationship between the concentration ratio of a compound in water phase (concentration in water sample) and in particulate phase (organic carbon normalized concentration in sediment) (S/W ratio) and latitude. The degree of correlation and the ratio (S/W) are indications of the efficiency of their transport.

Analytical results showed relatively uniform distribution of HCHs along with the latitudes, and the ratio (S/W) showed the highest degree of positive correlation with latitude. This phenomenon indicates that HCH isomers pose higher potency for atmospheric transport and global redistribution. Other compounds such as DDTs, CHLs and PCBs with lower correlations and gradients exhibited less efficiency for atmospheric transport.

Another case study conducted by Ramesh et al. (1989) and Takeoka et al. (1991) provided quantitative information on the budget and fluxes of HCHs in the Vellar River watershed, South India, a typical tropical area in Asia. The model showed that 99.6% of the HCH, which was applied to the catchment area, was volatilized and that only 0.4% drained into the estuary. In addition, 75% of the flux to the estuary was volatilized and, thus, only approximately 0.1% of the applied HCH finally reached the Bay of Bengal by aquatic pathways. Despite the fact that HCH was applied into paddy fields in large quantities in India, only very small amounts of this insecticide retained in the aquatic phase due to the extraordinary high degree of volatilization. Elevated temperature in the tropical watershed in South India is favorable factor for the rapid volatilization of highly volatile HCH isomers. Thus, results of this case study on a specific watershed site in South India is consistent with those in the case study on small scale latitudinal distribution of HCHs conducted from various sites along the estuaries of Mekong River Delta, Southern Vietnam (Iwata et al., 1994), which provided insights into the transport and fate of persistent semi-volatile organic compounds.

In general, the degree of usage, the climate and the physico-chemical properties of persistent organic compounds are the major factors controlling the magnitude of contamination in the environment. When comparing the distribution of OCs in different environmental media in Asia-Pacific region, we found that distribution patterns in fish and sediment were relatively similar and showed less spatial variation. The pattern in air and water exhibited greater geographical variability. A recent extensive survey of OC insecticides from Red and Duong rivers, the two largest rivers flowing through the northern delta region of Vietnam, and various lakes in Hanoi has elucidated that DDT and HCH residue levels in rivers are apparently greater than those in lakes (Hung and Thiemann, 2002). This observation suggests different behaviors of semi-volatile compounds in tropical lakes and in rivers, showing shorter retention time in lakes than in rivers. Again, elevated temperatures may enhance volatilization of such compounds, leading to shorter residence time in tropical water phase, lower residue levels and relatively uniform distribution in sediments and aquatic biota. In this context, the role of the tropics in Asia-Pacific region as potential emission sources for higher latitude areas deserves further attention.

In addition to the environmental samples, the use of biological tissues has been proved to be a reliable tool for understanding the distribution patterns and transport behavior of PTS (Tanabe and Subramanian, 2006). Recent studies using the skipjack tuna (*Katsuwonus pelamis*) for monitoring semi-volatile organic compounds in open seas and oceans can be a typical example (Ueno et al., 2003). Skipjack tuna is widely distributed all over the world oceans; its ecology is well studied as it is one of the target species of commercial fishing in Japan. We attempted to make clear the distribution and transport of these compounds using this species as a bioindicator (Ueno et al., 2003). Interestingly, HCH and hexachlorobenzene (HCB) concentrations were higher in the high-latitude areas than in the low-latitude areas (Fig. 18.8). Similar to these insecticides, significant levels of PCBs were detected not only in the samples from coastal waters but also in the open seas. According to various investigations conducted so far, PCBs as well as HCHs and HCB are known to have a highly transportable nature through atmosphere, contributing to global pollution. On the other hand, higher levels of DDTs were found in tunas collected from tropical and subtropical open sea waters, while northern tunas showed much lower DDT levels, which is also similar to the pattern found in dolphins and whales (Prudente et al., 1997). As for DRCs, Ueno et al. (2005) found measurable levels of PCDDs only in the samples from coastal waters around Japan and Korea, but very low levels below detection limit were found in samples from open seas (Fig. 18.9), which is

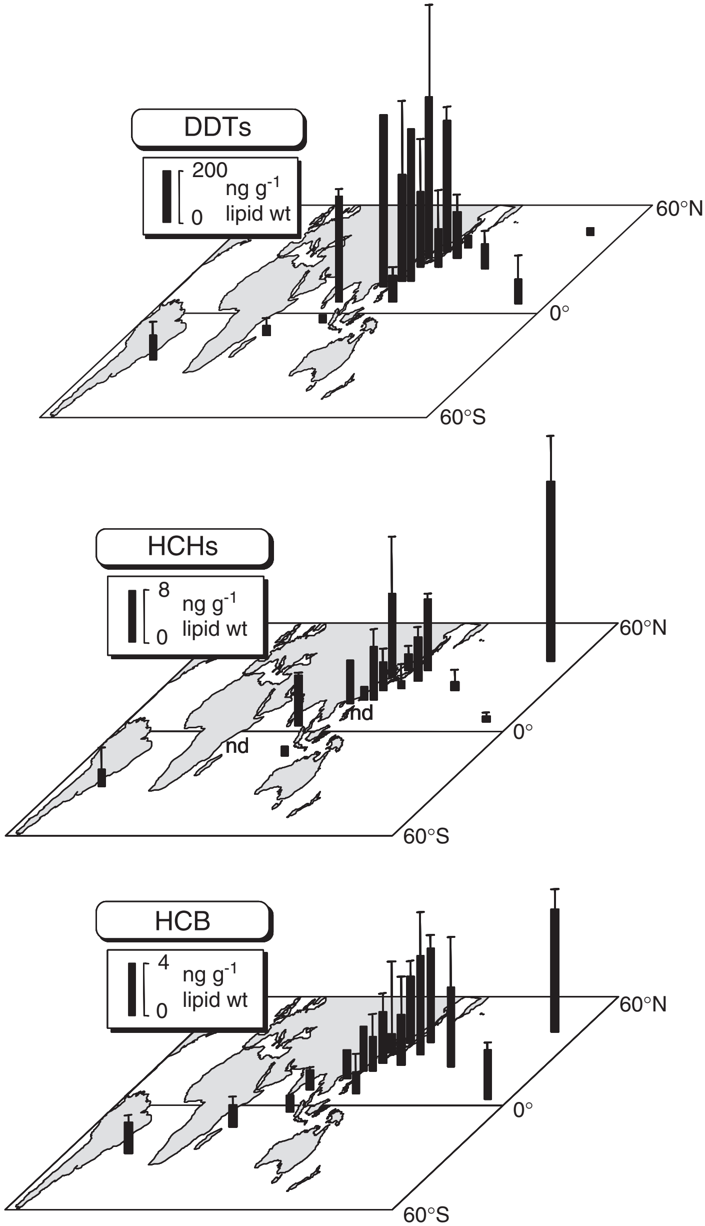

Figure 18.8. Geographical distribution of OC concentrations in the muscle of skipjack tuna. Data from Ueno et al. (2003).

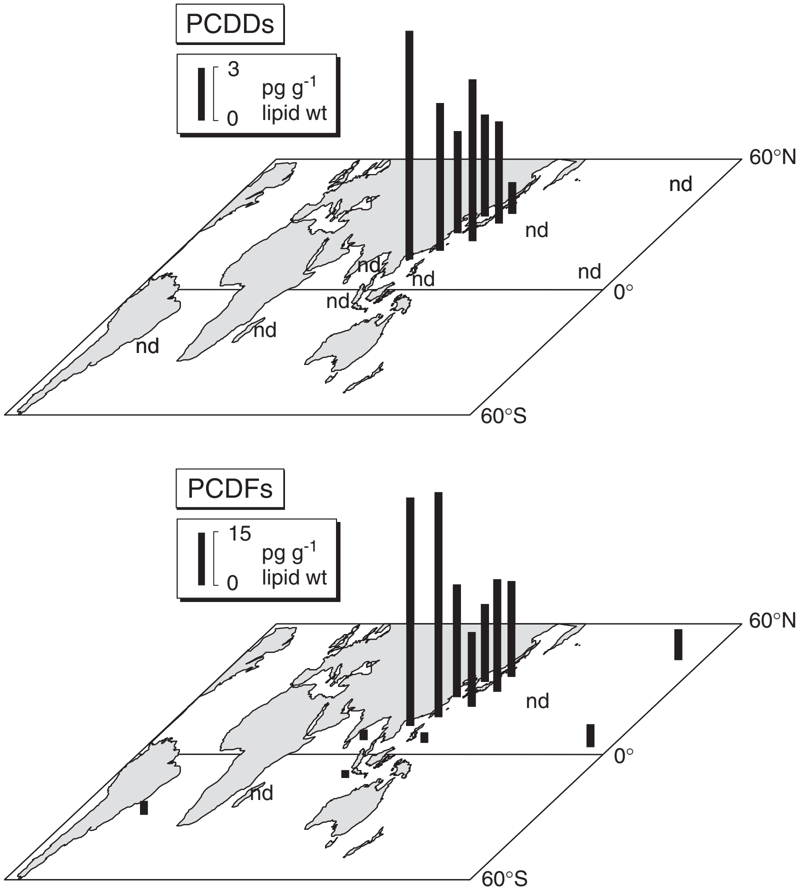

Figure 18.9. Geographical distribution of DRC concentrations in the muscle of skipjack tuna. Data from Ueno et al. (2005). nd: under detection limit.

similar to DDT distribution (Fig. 18.8). These observations on DDTs and dioxins confirm the less transportable and highly localized nature of these contaminants. As a further finding of spatial distribution, PBDEs were also detected in tunas from remote oceans even though their residue levels were relatively low, indicating PBDEs retain transportable nature to some extent similar to PCBs (Ueno et al., 2004). The highest concentration of PBDEs was detected in the sample from off-Taiwan, and relatively higher concentrations of these compounds were observed around the East China Sea and the South China Sea (Fig. 18.10).

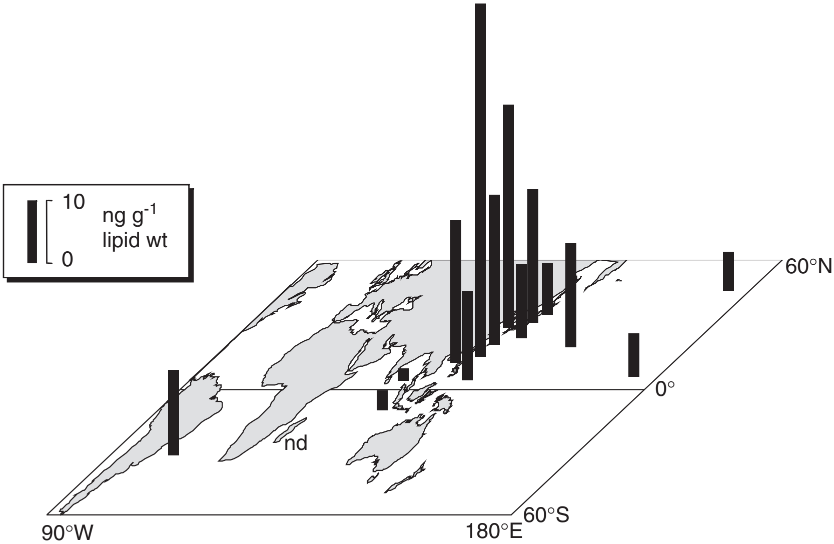

Figure 18.10. Geographical distribution of PBDE concentrations in the muscle of skipjack tuna. Data from Ueno et al. (2004). nd: under detection limit.

The distribution pattern of organohalogen compounds found in skipjack tunas is in association with the long-range atmospheric transport of the compounds. According to Wania and Mackay (1993), chemicals used in developing and developed nations evaporate at different rates, carried out globally by long-range atmospheric transport and eventually deposit mostly in the water phase of open seas, particularly in polar regions. In this context, volatile chemicals such as HCB, HCHs, PBDEs and PCBs are relatively transportable through atmosphere and deposit in open oceans, and significant residue levels were found in skipjack tunas and dolphins from open seas. On the other hand, less volatile contaminants such as DDT and dioxins have low transportability through air, and higher levels still exist in coastal wildlife. Considering these facts, DDTs and dioxins may be designated as regional pollution type contaminants, and PCBs, PBDEs, HCHs and HCB as global pollution type contaminants. In any case, the toxic impacts of POPs affect not only the local ecosystems nearby the emission sources, but also unexpected areas such as the Arctic and the Antarctic regions. Therefore, their influence to the environment should be examined on a global scale as has been done for issues like the ozone depletion by chlorofluorocarbon (CFC) and global warming by carbon dioxide.

18.4. Temporal trends and future prospects of contamination

It is essential to understand the future trends of contamination in order to evaluate forthcoming risks posed by PTS. Hence, we have examined the historical trends in residue levels of organohalogen compounds in some aquatic mammals. We first carried out the investigations to make clear the temporal trends of OC levels in inland areas and open seas. As shown in Fig. 18.11, most OCs in the archived samples of Baikal seals (*Phoca sibirica*), a representative species living in inland water body, were on a declining trend during 1992–2002 (Tanabe et al., 2003; Tsydenova et al., 2004). This result indicates that the regulation of OC production and usage following the recommendation by WHO, Food and Agriculture Organization (FAO), United Nation Environment Programme (UNEP), etc. showed significant effects on inland areas. Recently, Aguilar and Borrell (2005) reported similar decreasing trends of OC contamination in the Mediterranean Sea using striped dolphin (*Stenella coeruleoalba*) as an indicator. In the case of open seas, a study on the archived blubber samples of minke whales (*Balaenoptera acutorostrata*) collected from Antarctic Ocean, a remote area far away from emission sources of pollutants, during 1984–2001, showed that the concentrations of OC insecticides and PCBs did not clearly decline during this period (Fig. 18.12; Aono et al., 1997 with additional data). Considering all these facts, it can be said that the recent contamination by OCs in inland and coastal areas

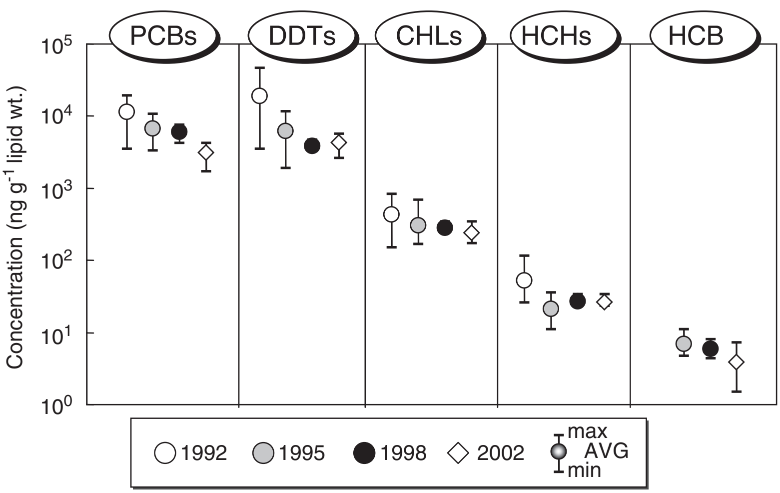

Figure 18.11. Temporal variation of OCs in Baikal seals collected from Lake Baikal in 1992–2002. Data from Tanabe et al. (2003) and Tsydenova et al. (2004).

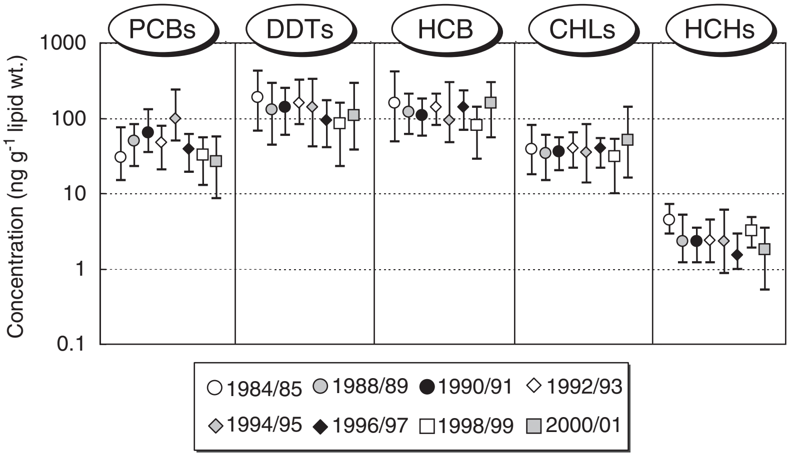

Figure 18.12. Temporal variation of OCs in minke whales collected from the Antarctic in 1984–2001.

have decreased in comparison with their severely polluted status in the 1970s. On the contrary, in remote oceans, their declining trends were very slow in the past several decades. This might be due to the nature of the oceans which are vast reservoirs and act as the final sinks for these persistent chemicals. From these observations we can conclude that the OC pollution and potential risks in marine ecosystems will continue for a considerable period of time in the future, and, hence, long-term monitoring for toxic contaminants is necessary in the marine environment, particularly in open oceans.

Interest in PBDE flame retardants has been as exponential as their apparent increase in the environment over the past 20–25 years in North America (Ikonomou et al., 2002; Norstrom et al., 2002; Rayne et al., 2003) and Europe (Norén and Meironyté, 2000; Thomsen et al., 2002). However, there is still very little information on PBDE contamination and its temporal trend from other regions of the world, including the Asia-Pacific region. Therefore, we have carried out comprehensive investigations to provide an in-depth understanding of the historical trends of contamination by PBDEs since 1972, in the fat tissues of northern fur seals (*Callorhinus ursinus*) inhabiting and migrating in the North Pacific, in comparison to classical OCs (Tanabe et al., 1994; Kajiwara et al., 2004). The lowest PBDE levels were in the fur seals collected in 1972, with the peak concentration around 1991–1994, and then, decreased to about 50% in 1997–1998 (Fig. 18.13). On the other hand, OCs in northern fur seals showed the highest levels in 1970s to early 1980s and then decreased.

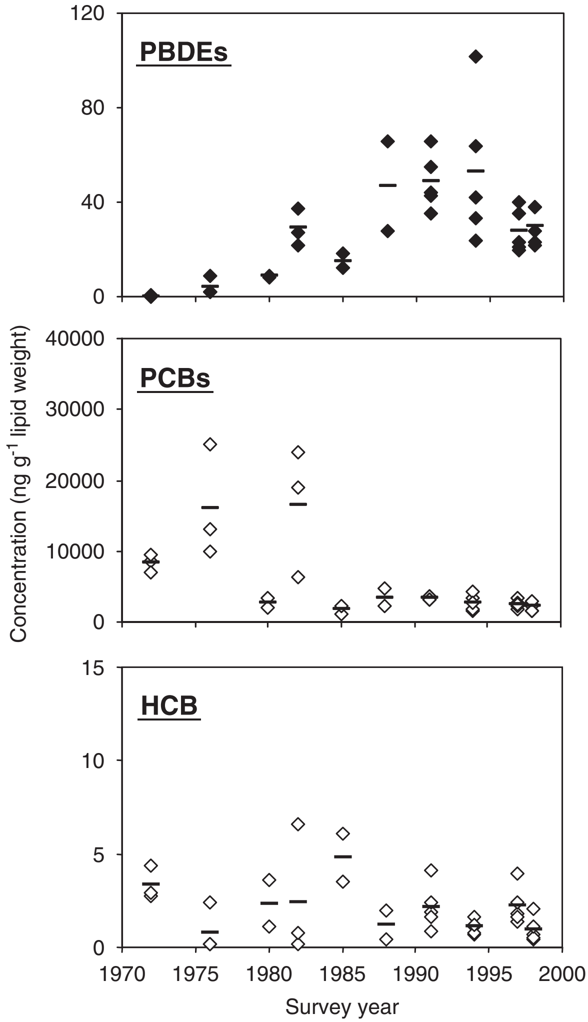

Figure 18.13. Temporal trends in concentrations of PBDEs and OC residue in female northern fur seals collected from 1972 to 1998. Horizontal bars indicate mean values. Data from Tanabe et al. (1994) and Kajiwara et al. (2004).

Compositions of higher brominated congeners increased since 1972, while some lower brominated congeners decreased, implying a change in the pattern of use, particularly the increased usage of highly brominated diphenyl ethers during 1972–1998. In the meantime, PCB compositions in fur seals showed no temporal variation, suggesting a continuous input of

PCBs into the marine environment in significant quantities. As peak concentrations of PBDEs occurred later than OCs, it is essential to follow-up the patterns of PBDE pollution that may be of great concern in the future.

Furthermore, to examine temporal changes in the past 20 years around Japan and 10 years around China, we analyzed PBDEs and OCs in the blubber of melon-headed whales (*Peponocephala electra*) stranded along Japanese coasts in 1982 and 2001 and finless porpoises (*Neophocaena phocaenoides*) from Chinese waters in 1990/91 and 2000/01, respectively. In whales from Japan, PBDE levels in 2001 were significantly higher than 1982, showing an increase of about 10 times (Fig. 18.14). Similarly, finless porpoise from China showed five times increase during the past 10 years, indicating that PBDEs usage in China also increased drastically in the past decade (Ramu et al., 2006). When we compared PBDE congener patterns, higher contributions of hexa-BDEs were observed in whales collected in 2001 than those in 1982. This is similar to the trend found in northern fur seals from the Pacific coast of Japan (Kajiwara et al., 2004), indicating a possible change in the source products. On the other hand, congener profiles in porpoises from China did not shift to higher BDE congeners, implying a continuous discharge of lower BDE commercial mixtures such as Penta-BDE.

Asian countries consumed about half of the annual worldwide consumption of PBDEs in 2001 (Watanabe and Sakai, 2003; BSEF, 2004). In Asia, the major portion of PBDE formulations was estimated to be

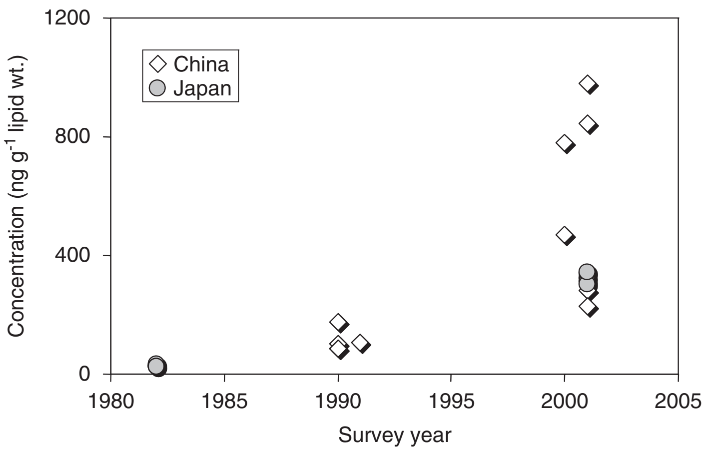

Figure 18.14. Temporal changes in PBDE levels in the cetaceans found stranded along the coasts of China and Japan.

consumed by the countries other than Japan, suggesting that considerable amounts of PBDEs have been also used in some developing countries in Asia, probably in nations with high economic growth rate. The high population and economic growth rate in China (Li and Daler, 2004) undoubtedly accelerated the industrial and human activities involving the usage of modern chemicals including PBDEs. Higher levels of PBDEs in mussels and small cetaceans from Hong Kong, mussels from Korea (Figs. 18.1 and 18.3) and fishes from the East China Sea (Fig. 18.10) as well as the increasing temporal trend of PBDE levels in wildlife from Chinese coasts may suggest their expanding usage in China and nearby countries. Moreover, the export of waste electric products like computers, televisions, etc. used in developed nations to Asian developing countries such as China as trash has been reported (Hileman, 2002; Schmidt, 2002). In some cases, the wastes were burnt in open dumping sites, which may lead to the formation of chlorinated and brominated dioxins. A series of our investigations underlines the need for long-term monitoring of PBDE pollution in Chinese coastal waters as well as other countries in Asia-Pacific region. It is also imperative to find out the pollution sources of PBDEs in the entire Asian region.

18.5. Human exposure and human health implications

18.5.1. Monitoring the status of contamination using breast milk as bioindicator

The presence of PTS in humans is always viewed with concerns because of their potential for various biological effects such as teratogenicity, carcinogenicity, neurotoxicity and immunotoxicity (Nicholson and Landrigan, 1994). Human exposure to toxic chemicals occurs throughout the entire life span of humans, from the prenatal period in the mother's womb and continues until death. Particularly, persistent chemicals such as OC insecticides, PCBs, PBDEs and DRCs with relatively high lipophilicity tend to accumulate in large quantities in lipid rich tissues. Among these, their accumulation in human milk has been of great concern and has received considerable attention during the past four decades. In the previous review, we pointed out that human tissue samples, particularly breast milk, collected in a non-destructive manner, are among the best samples for measuring both spatial and temporal variations of POPs. The advantage of using human tissue samples is that background information such as age, reproduction and possible route of exposure can be accurately obtained from the subjects (Tanabe and Subramanian, 2006).

Recently, we have conducted various studies on the contamination status of PTS comprising PCDDs, PCDFs, PCBs and OC insecticides such as DDTs, HCHs, CHLs and HCB in human breast milk collected from general public and the residents around open dumping sites of municipal wastes in Asian developing countries such as India, Cambodia, Vietnam, Philippines, Malaysia, Indonesia and China during 1999–2003 (Kunisue et al., 2002b, 2004a, b, c, d; Minh et al., 2004; Sudaryanto et al., 2005b, 2006).

Mean lipid-normalized concentrations of POPs and TEQs of DRCs, which were estimated based on human/mammal toxic equivalency factors (TEFs) proposed by WHO (Van den Berg et al., 1998), in human breast milk collected from general public in Asian countries such as India, Cambodia, Vietnam, Philippines, Malaysia, Indonesia, China and Japan during 1999–2003 (Kunisue et al., 2002b, 2004a, b, c, d; Minh et al., 2004; Sudaryanto et al., 2005b, 2006) were illustrated in Figs. 18.4 and 18.15–18.19. In general, relatively high concentrations of DDTs were detected in human breast milk from Asian developing countries, while elevated residue levels of TEQs of DRCs, PCBs and CHLs were observed in Japanese breast milk. This trend of pollution was also observed in the Asia-Pacific Mussel Watch Program of POPs, in which mussels were employed as bioindicators (Monirith et al., 2003b). In Japan, it has been demonstrated in our recent studies on wildlife that TEQs, PCBs and CHLs were predominant (Kunisue et al., 2003, 2006; Kubota et al., 2004).

Among Asian developing countries, concentrations of DDTs in human breast milk from Vietnam, China, Cambodia and Malaysia were relatively higher than those from other countries (Fig. 18.16). It has been previously reported that DDTs were predominant in environmental media, biota and foodstuff from these Asian developing countries. In Vietnam, higher levels of DDTs in sediments from densely populated areas as compared to those from paddy field were reported, indicating recent application of DDTs for public health purposes such as malaria control rather than for agriculture (Nhan et al., 2001). In addition, our previous studies also demonstrated the elevated residue levels of DDTs in wild avian species and foodstuffs from Vietnam (Kannan et al., 1992, 1997; Minh et al., 2002; Kunisue et al., 2003). Although fewer comprehensive investigations on pollution by POPs in Cambodia are available due to lack of monitoring of such pollutants affected by a long-term civil war, it was reported that DDT has been used to control parasites of fish in cage culture (Tana, 1996) and relatively high residue levels of DDTs were observed in fish (Monirith et al., 1999). Higher levels of DDTs were also detected in inland resident avian species (Monirith et al., 2003a). In China, it has been recently reported that not only DDTs but also

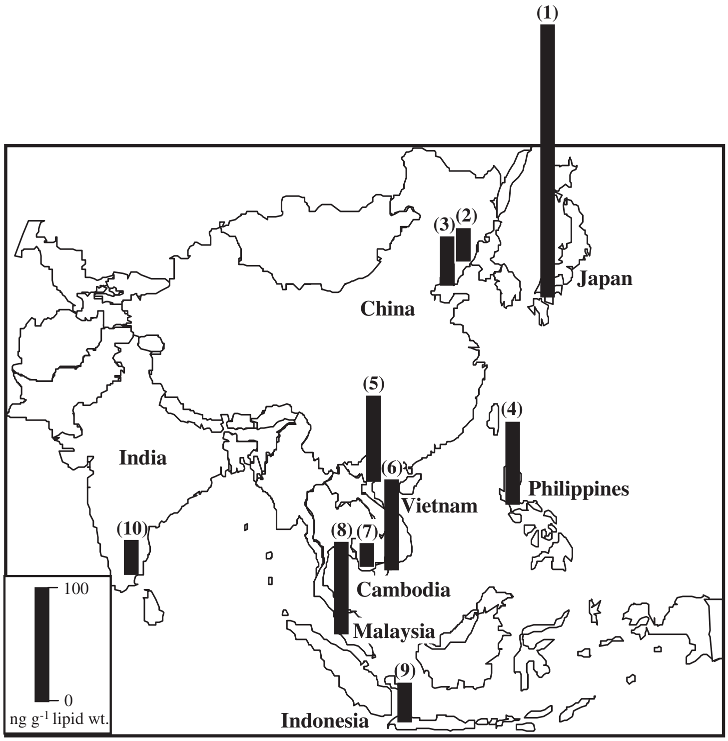

Figure 18.15. Concentrations of PCBs in human breast milk collected from general public in Asian countries. (1) Ehime, Japan (Kunisue et al., 2004c), (2) Shenyang, China (Kunisue et al., 2004a), (3) Dalian, China (Kunisue et al., 2004a), (4) Quezon, Philippines (Kunisue et al., 2002b), (5) Hanoi, Vietnam (Minh et al., 2004), (6) Hochiminh, Vietnam (Minh et al., 2004), (7) Phnom Penh, Cambodia (Kunisue et al., 2004b), (8) Penang, Malaysia (Sudaryanto et al., 2005b), (9) Jakarta, Indonesia (Sudaryanto et al., 2006), (10) Palaverkadu, India (Kunisue et al., 2002b).

HCHs were predominant in various environmental samples along the coastal areas and foodstuffs (Zhou et al., 2001; Chen et al., 2002; Nakata et al., 2002). The investigation on pollution of DDTs and HCHs in sediment cores indicated recent input of these contaminants into coastal areas from inland (Zhang et al., 2002). In addition, the investigation on pollution of DDTs in fish and mussels collected from aquaculture cages in coastal waters showed high proportions of *p,p′*-DDT and also indicated recent releases of this chemical to the environment (Klumpp et al., 2002). Although studies on POPs pollution in Malaysia are limited, Zakaria et al. (2003) reported relatively high levels of *p,p′*-DDT in water and sediments from agricultural areas compared with other POPs.

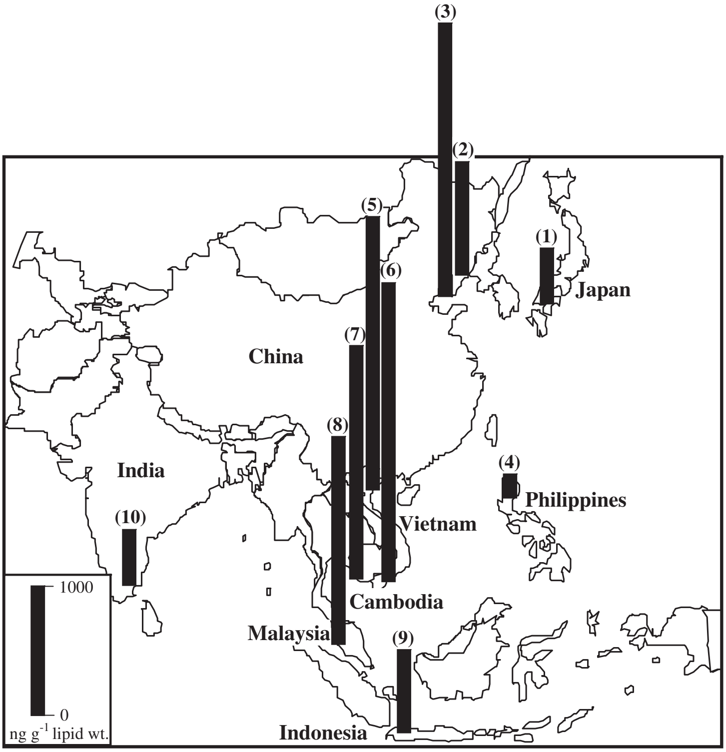

Figure 18.16. Concentrations of DDTs in human breast milk collected from general public in Asian countries. References cited are similar to those in Fig. 18.15.

Considering the above observations, it can be suspected that DDT may be illegally in use in some Asian developing counties and the residents are constantly exposed to this contaminant.

In Chinese and Indian human breast milk, notably higher concentrations of HCHs were observed compared with other Asian countries (Fig. 18.17). It seems that this reflects a huge amount of usage in the past and/or recent illegal use of technical HCH in these two countries. The total usage of technical HCH was estimated as 4.46 and 1.0 million metric tons in China and India, respectively, which are the largest and second largest consumers in the world (Li et al., 1998). As described earlier, relatively high residue levels of HCHs have been recently reported in Chinese environmental media and foodstuffs (Zhou et al., 2001; Chen et al., 2002; Nakata et al., 2002). Our studies revealed elevated residue levels of HCHs in wild avian species and foodstuffs

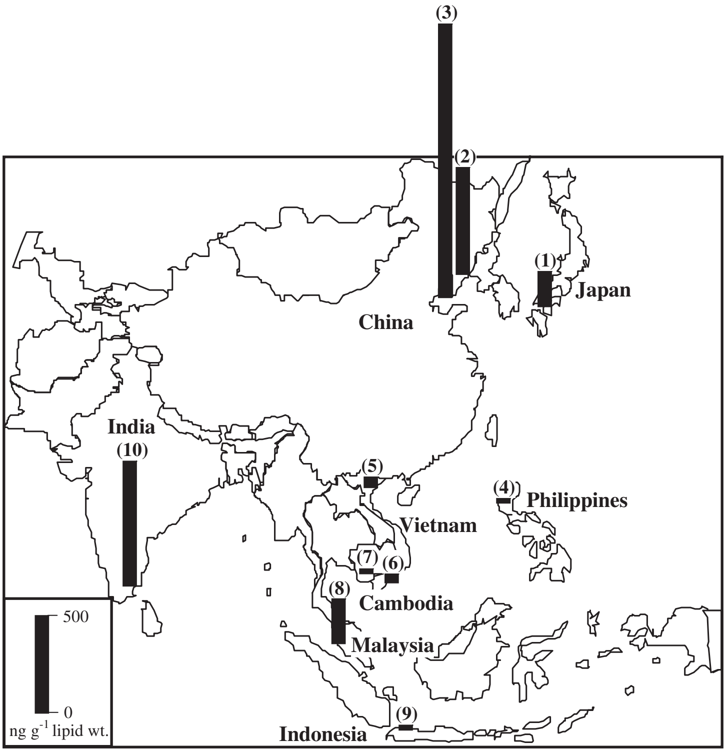

Figure 18.17. Concentrations of HCHs in human breast milk collected from general public in Asian countries. References cited are similar to those in Fig. 18.15.

from India (Kannan et al., 1997; Tanabe et al., 1998; Kunisue et al., 2003). These results indicate that considerable sources of HCHs are still present in the environment of these countries, and that technical HCH may be illegally in use for public health purposes in these two counties and residents are exposed to HCHs.

In human breast milk from China, HCB levels were also higher than other Asian countries (Fig. 18.19). HCB has been produced in a large chemical factory along Ya-Er Lake, which is located in the eastern part of Wuhan, Hubei province, and elevated levels of HCB were detected in soils and sediments from this lake (Wu et al., 1997). In addition, high concentrations of HCB were also detected in tea leaves, indicating recent use of this chemical as a fungicide (Nakata et al., 2002). These results imply that use of HCB is still continuing in China. In addition to being a fungicide and an impurity of pesticides, HCB is known to be formed during waste burning.

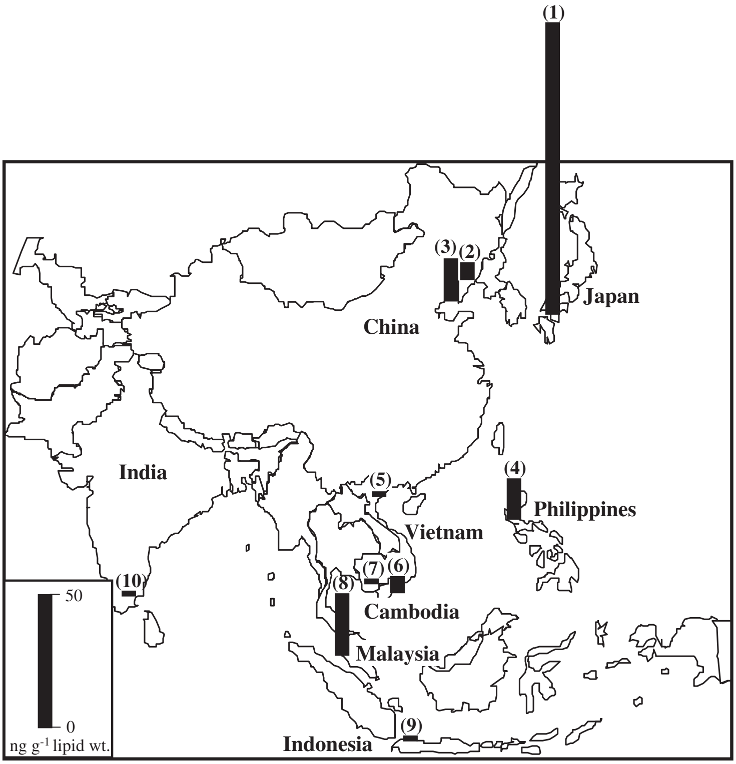

Figure 18.18. Concentrations of CHLs in human breast milk collected from general public in Asian countries. References cited are similar to those in Fig. 18.15.

To understand the magnitude of contamination in human breast milk from Asian countries, levels of PTS were compared to those reported recently in other developed, developing and former socialist countries (Schecter et al., 1990a, b; Dewailly et al., 1992; Hernández et al., 1993; Fürst et al., 1994; Becher et al., 1995; Liem et al., 1995; Gonzalez et al., 1996; Cok et al., 1997, 1999; Hooper et al., 1997; Hansen, 1998; Kinyamu et al., 1998; Schade and Heinzow, 1998; Kiviranta et al., 1999; Newsome and Ryan, 1999; Schuhmacher et al., 1999; Norén and Meironyté, 2000; Paumgartten et al., 2000; Harris et al., 2001; Waliszewski et al., 2001) (Table 18.1). In general, contamination by DDTs in human breast milk is higher in developing and former socialist countries than in developed countries, while levels of TEQs of DRCs, PCBs and CHLs in Asian developing countries were notably lower than those in developed nations. Among human breast milk from Asian developing countries, levels of DDTs in Vietnam, China, Cambodia and Malaysia, HCHs in China and India, and HCB in China were

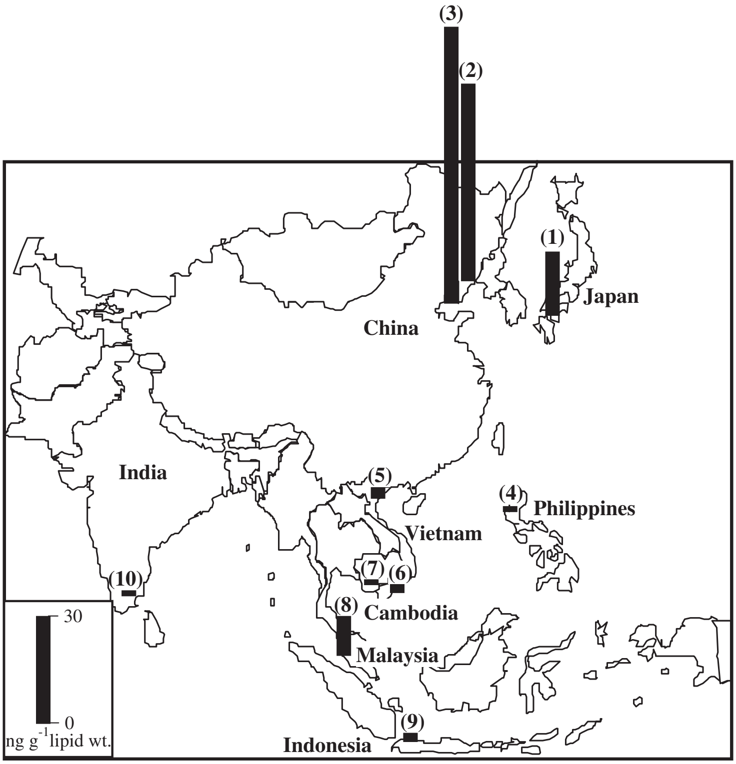

Figure 18.19. Concentrations of HCB in human breast milk collected from general public in Asian countries. References cited are similar to those in Fig. 18.15.

relatively higher compared with other countries, again suggesting recent use of these OC insecticides and fungicides and that these Asian developing countries may be potential sources of these contaminants to the global environment. Moreover, given the uncontrolled and low temperature burning of dumped solid wastes under anaerobic conditions is common in open dumping sites in various Asian countries; the formation of contaminants such as HCB during combustion is another issue that needs particular attention. Further investigations on human exposure and temporal trends of PTS, especially OC insecticides, are needed to elucidate future pollution trends.

18.5.2. Dioxin contamination in open dumping sites in Asian developing countries

In Asian developing countries, the presence of large open dumping sites of municipal wastes in the suburban areas of major cities is the prime

Table 18.1. International comparison of persistent toxic substances in human breast milk

Country	Survey year	TEQs[a]	PCBs[b]	DDTs[b]	HCHs[b]	CHLs[b]	HCB[b]	References
Asian countries								
Japan	1999	36	240	430[d]	180[h]	110[l]	18	Kunisue et al. (2004c)
China	2002	8.2	42	2100[d]	1400[h]	16[l]	81	Kunisue et al. (2004a)
Philippines	2000	12	–	–	–	–	–	Kunisue et al. (2004d)
Philippines	2000	–	72	190[d]	4.7[h]	15[l]	<0.56	Kunisue et al. (2002b)
Vietnam	2000	12	–	–	–	–	–	Kunisue et al. (2004d)
Vietnam	2000	–	74	2100[d]	58[h]	2.0[l]	3.9	Minh et al. (2004)
Cambodia	1999–2000	7.8	–	–	–	–	–	Kunisue et al. (2004d)
Cambodia	1999–2000	–	20	1800[d]	5.6[h]	1.6[l]	1.6	Kunisue et al. (2004b)
Malaysia	2003	13	80	1600[d]	230[h]	23[l]	11	Sudaryanto et al. (2005b)
Indonesia	2001	–	33	640[d]	14[h]	2.0[l]	2.2	Sudaryanto et al. (2006)
India	2000	12	–	–	–	–	–	Kunisue et al. (2004d)
India	2000	–	30	430[d]	640[h]	0.91[l]	1.0	Kunisue et al. (2002b)
Other developing and former socialist countries								
Turkey	1995–1996	–	–	2400[e]	480[h]	–	50	Cok et al. (1997)
Iran	1991	–	–	2000[e]	600[h]	–	61	Cok et al. (1999)
Brazil	1992	9.7[c]	150	1700[d]	280[h]	–	12	Paumgartten et al. (2000)
Mexico	1997–1998	–	–	4700[f]	60[h]	–	30	Waliszewski et al. (2001)
Kenya	1991	–	–	470[f]	96[i]	–	–	Kinyamu et al. (1998)
Kazakhstan	1994	–	380	2300[e]	2300[i]	–	91	Hooper et al. (1997)
Russia	1996	–	520	2000[e]	560[m]	27[m]	99	Hansen (1998)
Russia	1989	24[c]	–	–	–	–	–	Schecter et al. (1990a)
South Africa	–	15[c]	–	–	–	–	–	Schecter et al. (1990b)

Other developed countries								
Sweden	1997	–	320	170[e]	–	–	12	Norén and Meironyté (2000)
Germany	1995–1997		550	240[e]	40[j]	–	80	Schade and Heinzow (1998)
Germany	1986–1991	34[c]	–	–	–	–	–	Fürst et al. (1994)
Canada	1996		250	470[g]	23[h]	140[l]	43	Newsome and Ryan (1999
Canada	1990	24	–	–	–	–	–	Dewailly et al. (1992)
Spain	1991		–	610[e]	280[h]	–	0.6	Hernández et al. (1993)
Spain	1996	14[c]	–	–	–	–	–	Schuhmacher et al. (1999)
UK	1997–1998		–	470[e]	100[k]	–	43	Harris et al. (2001)
USA	1980s	19[c]	–	–	–	–	–	Schecter et al. (1990a)
France	1990	23[c]	–	–	–	–	–	Gonzalez et al. (1996)
Norway	1992–1993	36	–	–	–	–	–	Becher et al. (1995)
Finland	1992–1994	39	–	–	–	–	–	Kiviranta et al. (1999)
Netherlands	1993	47	–	–	–	–	–	Liem et al. (1995)

[a] $pg\,g^{-1}$ lipid wt.
[b] $ng\,g^{-1}$ lipid wt.
[c] PCDD/DFs only.
[d] p,p'-DDE + p,p'-DDT + p,p'-DDD.
[e] p,p'-DDE + p,p'-DDT.
[f] p,p'-DDE + p,p'-DDT + p,p'-DDD + o,p'-DDT.
[g] p,p'-DDE + p,p'-DDT + o,p'-DDT.
[h] α-HCH + β-HCH + γ-HCH.
[i] α-HCH + β-HCH.
[j] β-HCH only.
[k] β-HCH + γ-HCH.
[l] Oxychlordane + *trans*-nonachlor + *cis*-nonachlor.
[m] Total.

source of critical DRC pollution. In those dumping sites, varieties of municipal wastes are burnt under low temperature by spontaneous combustion or intentional incineration, leading to secondary formation of DRCs. We found that the residue levels of DRCs were significantly higher in soils from those dumping sites than the agricultural and urban soils collected far from these areas (reference sites), indicating that the dumping sites are potential sources of DRCs (Minh et al., 2003). To understand the possible toxic impacts of DRCs on humans, we attempted to elucidate the contamination status of DRCs in human breast milk collected from the residents around the dumping sites of municipal wastes in India, Cambodia, and Vietnam and compared with those from the reference sites (Kunisue et al., 2004d).

As already shown previously while discussing the different sources of DRCs, it is noteworthy that the concentrations of DRCs in human breast milk from the dumping site in India were significantly higher than those from the reference site and those from Cambodia and Vietnam (Fig. 18.5), indicating that significant pollution sources of DRCs are present in the dumping site of India and the residents around there have been exposed to relatively higher levels of these contaminants than the other two developing countries. To understand the magnitude of contamination in human breast milk from the dumping sites in India, Cambodia and Vietnam, TEQ levels were compared with the values for human breast milk from general public of other countries since 1990, selected from publications in which concentrations of all the isomers were reported (Fig. 18.20). Because I-TEFs were mainly used to calculate TEQs, the reported data were recalculated by using WHO-TEFs for comparison. The levels of TEQs in human breast milk from India (38 pg TEQs g^{-1} lipid wt.) were comparable to or higher than those from developed countries (Schecter et al., 1990b; Dewailly et al., 1992; Fürst et al., 1994; Becher et al., 1995; Liem et al., 1995; Gonzalez et al., 1996; Kiviranta et al., 1999; Schuhmacher et al., 1999; Kunisue et al., 2004c) and the former Soviet Union (Schecter et al., 1990a). This suggests that the residents around the dumping sites of India have been exposed to comparable levels of DRCs with the general public of developed countries. On the other hand, the levels of TEQs in human breast milk from Cambodia (9.2 pg TEQs g^{-1} lipid wt.) and Vietnam (13 pg TEQs g^{-1} lipid wt.) were lower than those from developed countries and comparable to those from other developing countries (Schecter et al., 1990b; Paumgartten et al., 2000; Kunisue et al., 2004a; Sudaryanto et al., 2005b). In this international comparison, however, there are some uncertainties such as age and parity of the mother, sampling period, sample number and accuracy of the analytical techniques involved. In addition, only very little

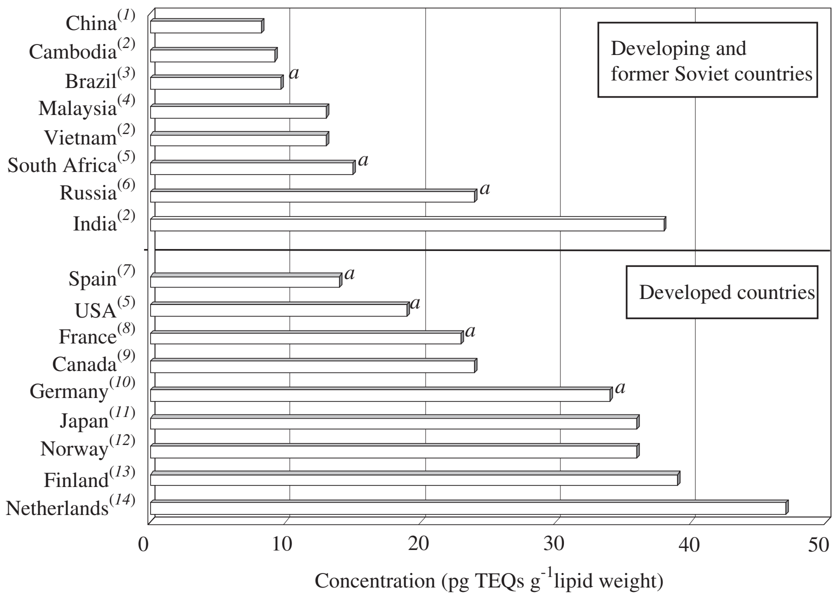

Figure 18.20. Comparison of TEQ levels in human breast milk from the residents around dumping sites in India, Cambodia and Vietnam with those from general public in other countries. [a]PCDD/Fs only. [1]Kunisue et al. (2004a), [2]Kunisue et al. (2004d), [3]Paumgartten et al. (2000), [4]Sudaryanto et al. (2005b), [5]Schecter et al. (1990b), [6]Schecter et al. (1990a), [7]Schuhmacher et al. (1999), [8]Gonzalez et al. (1996), [9]Dewailly et al. (1992), [10]Fürst et al. (1994), [11]Kunisue et al. (2004c), [12]Becher et al. (1995), [13]Kiviranta et al. (1999), [14]Liem et al. (1995).

data is available on non- and mono-*ortho* PCBs in the literature. Because of such uncertainties, it was difficult to draw any firm conclusion. However, the observation that TEQs of DRCs in human breast milk from the dumping site of India were comparable to or higher than those from some developed countries including Japan is noteworthy. In developed countries, it is claimed that the residue levels of DRCs in human breast milk decreased recently (LaKind et al., 2001), because of the installation of highly efficient incinerators and strict regulations on the production and usage of various chemicals. On the other hand, in Asian developing countries, it can be anticipated that the pollution by DRCs may increase further and hence the residue levels in human breast milk may also increase in future, because the release of these contaminants are not at all controlled even now.

Although residue levels of DRCs in soils collected from the open dumping sites in Asian developing countries were apparently greater than those from the reference sites (Minh et al., 2003), the levels of DRCs in human breast milk from the residents around the dumping sites in Cambodia and Vietnam were not significantly higher than those from reference sites. This implies that the residents around the dumping sites in Cambodia and Vietnam have not been greatly exposed to DRCs originating from the dumping sites. However, residue levels of these contaminants in Indian samples around the dumping site were notably higher (Fig. 18.5). In India, buffaloes and cows reared near the waste dumping site mainly feed on dumped leftovers. The residents around the dumping site constantly drink the milk collected from these bovines. On the other hand, in Cambodia and Vietnam, livestock such as buffaloes and cows are not reared around the dumping sites. To elucidate whether bovine milk is a potential source of DRCs for the residents around the dumping site in India, residue levels of these contaminants in the milk of buffaloes and cows collected there were estimated (Kunisue et al., 2004d).

DRCs were detected with elevated levels in the samples from dumping sites in all the bovine milk samples analyzed (Fig. 18.6). This indicates that buffaloes and cows feeding in the dumping site of India consume greater amounts of DRCs through contaminated soils and/or garbage, and that daily intake of these bovine milk by the residents around the dumping site in India is one of the possible reasons why TEQ levels in human breast milk collected from the dumping site were significantly higher than those from the reference site. In India, dietary consumption of dairy products is generally higher than that of other countries, and average consumption of milk in India by a person per day rose from 135 g in 1980 to 176 g in 1990 (John et al., 2001). The residents around the dumping site in India constantly drink the milk collected from reared buffaloes and cows. Assuming that an adult weighing 60 kg drinks 176 g of buffalo or cow milk per day, estimated daily intake of TEQs from bovine milk from the dumping site was from 1 to 4 pg TEQs kg^{-1} day^{-1}, as investigated in our study—the range of TDI proposed by WHO (Van Leeuwen et al., 2000), and only in one buffalo milk sample the value exceeded TDI (Fig. 18.21). Even though the values are within TDI, the residents around the dumping site in India are exposed to considerably high levels of DRCs and hence may be at greater risk of exposure to these contaminants via bovine milk.

18.5.3. Risk assessment for infant

It is realized that the presence of POPs in human breast milk is of great concern because these lipophilic chemicals are readily transferred and

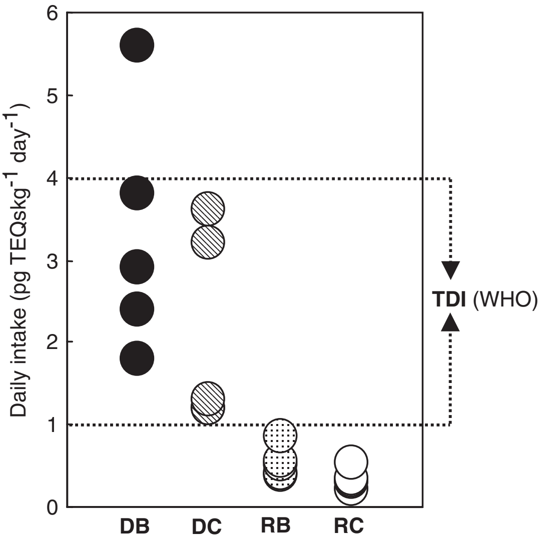

Figure 18.21. Estimated daily intake of TEQs by adults through bovine milk collected from the dumping and reference sites in India. Daily intake was estimated based on the assumption that an adult (60 kg) ingests 176 g of bovine milk per day (John et al., 2001). DB = Buffalo milk from the dumping site. DC = Cow milk from the dumping site. RB = Buffalo milk from the reference site. RC = Cow milk from the reference site.

absorbed to infants. In case of dioxins, it is reported that one to three months old infants absorb above 90% of most dioxin congeners present in their mothers' milk (McLachlan, 1993; Pluim et al., 1993; Dahl et al., 1995), hence they may be exposed to relatively high levels of POPs during this period. To understand the magnitude of exposure to PTS by infants, we estimated daily intake (DI) from the levels of these contaminants in human breast milk observed in Asian countries, based on the assumption that an infant ingests 700 ml milk per day and the weight of an infant is 5 kg (Kunisue et al., 2002b, 2004a, b, c, d; Minh et al., 2004; Sudaryanto et al., 2005b, 2006) and compared to the guideline standards proposed by WHO and Health Canada (Oostdam et al., 1999; Van Leeuwen et al., 2000).

As expected, relatively higher DIs of TEQs were observed in the infants from Japan and the dumping site in India compared with those from other countries and DIs in all the cases exceeded 1–4 pg TEQs kg^{-1} day^{-1}, the TDI (Table 18.2). In addition, DIs of PCBs, CHLs by Japanese, DDTs by Chinese, Vietnamese, Cambodian, and Malaysian, HCHs by Chinese and Indian, and HCB by Chinese infants were relatively high, and the DIs of these contaminants by some individuals exceeded the TDIs (Table 18.2).

Table 18.2. Estimated daily intakes of persistent toxic substances by infants in Asian countries

Country	TEQs[a]	PCBs[b]	DDTs[b]	HCHs[b]	CHLs[b]	HCB[b]
Japan	140 (28–630)	1.0 (0.17–7.8)	2.0 (0.19–22)	0.83 (0.12–6.6)	0.56 (0.038–7.8)	0.069 (0.019–0.34)
China	20 (17–26)	0.075 (0.008–0.45)	3.2 (0.39–18)	1.9 (0.21–15)	0.023 (0.002–0.12)	0.15 (0.029–0.68)
Philippines	29 (1.9–52)	0.14 (0.003–0.30)	0.58 (0.027–1.4)	0.015 (<0.002–0.028)	0.048 (0.003–0.12)	0.002 (<0.002–0.009)
Vietnam	35 (10–60)	0.29 (0.030–1.2)	8.9 (0.78–110)	0.11 (0.010–0.43)	0.018 (<0.002–0.080)	0.012 (<0.002–0.040)
Cambodia	26 (2.4–67)	0.080 (0.012–0.42)	4.5 (0.47–23)	0.016 (<0.002–0.065)	0.006 (<0.002–0.018)	0.006 (<0.002–0.032)
Malaysia	19 (3.3–40)	0.21 (0.022–1.7)	4.0 (0.19–19)	0.43 (0.063–1.8)	0.050 (0.006–0.18)	0.031 (<0.002–0.29)
Indonesia	–	0.080 (0.020–0.32)	1.2 (0.030–8.9)	0.040 (0.005–0.25)	0.010 (<0.002–0.030)	0.010 (<0.002–0.020)
India	28 (18–35)	0.037 (0.012–0.060)	1.0 (0.33–3.1)	1.6 (0.57–4.0)	0.003 (<0.002–0.012)	0.003 (<0.002–0.012)
India (dumping site)	120 (28–500)	0.12 (0.023–0.33)	1.1 (0.30–4.0)	1.6 (0.27–3.8)	0.024 (0.002–0.087)	0.003 (<0.002–0.009)
TDIs[c]	1–4	1.0	20	0.3	0.05	0.27

Note: Numbers in parentheses indicate range values.

[a] $pg\,kg^{-1}$ body wt. day^{-1}.

[b] $\mu\,kg^{-1}$ body wt. day^{-1}.

[c] Tolerable daily intakes (TDIs) were cited from Oostdam et al. (1999) and Van Leeuwen et al. (2000).

It is argued that DRCs induce various toxic effects such as cancer in animal bodies (Birnbaum, 1994). Furthermore, *p,p′*-DDE and β-HCH, the compounds that are there generally abundant in human breast milk, have been reported as an androgen antagonist and environmental estrogen, respectively (Keice et al., 1995; Willett et al., 1998). These observations imply that abundance of POPs in human breast milk may adversely affect development and reproductive systems of Asian children. However, it is difficult to draw any firm conclusion from Table 18.2 whether adverse effects by POPs have already occurred in Asian infants, because TDIs used here are estimated on the basis of life span exposure. Not only TDIs from life span but also TDIs of POPs estimated from breast-feeding period may be needed (LaKind et al., 2001).

If only the risk of PTS is considered, breast milk containing high levels of such contaminants should not be fed to infants. Feeding with formula milk diet not contaminated by PTS is one of the measures for protecting infants from the risk. In human breast milk, however, not only general nutrients but also essential components for infant's growth and development such as secretory IgA, oligosaccharides, lactoferrin and lysozyme, which can increase their resistance to common infections, are present (Lonnerdal, 2000). In addition, long-chain polyunsaturated fatty acids in human breast milk are indispensable for brain development of infants and it is reported that breast-feeding is associated with significantly higher scores for cognitive development than formula feeding (Uauy and Peirano, 1999). It is suspected that deficiency of breast-feeding negatively affects the growth and development of infants, while multiple and quantitative effects of these components are not completely elucidated. Furthermore, in some Asian developing countries, water used for many formula milk preparations may contain various infectious organisms and environmental contaminants, which may adversely affect infant health (Carpenter et al., 2000). Considering all these factors, breast-feeding is essential for the infants, as recommended elsewhere (LaKind et al., 2001; Longnecker and Rogan, 2001; Abelsohn et al., 2002). But it is necessary to reduce the levels of PTS in human breast milk, especially OC insecticides such as DDTs and HCHs in Asian developing countries and dioxins in Indian dumping site, to save the infants from possible toxic effects. In case of dioxins, however, it is estimated that 86% of dioxins in human breast milk comes through mobilization from adipose tissue and 14% from dietary sources, and so it may be difficult to reduce dioxin concentrations in human breast milk through short-term dietary control (Koppe, 1995). This means that the females have to lessen PTS exposure through dietary source from the early days of their life span. It is necessary to elucidate whether recent input and illegal use of OC insecticides are present and to continuously investigate temporal trends of POP pollution in Asian developing countries to remedy the situation. Furthermore, in the dumping sites of Asian developing countries, especially India, urgent control and regulation of pollution sources of DRCs are required.

Overall, our results suggest that in some Asian developing countries, OC insecticides such as DDTs and HCHs may be still in use and the residents are continuously exposed to these contaminants. Furthermore, we demonstrated that in India the concentrations of dioxins in human breast and bovine milk from the open dumping site were significantly higher than those from the reference sites and other Asian developing countries, indicating that significant pollution sources of dioxins are present in the Indian dumping sites and the residents around these have been exposed to relatively higher levels of these contaminants via bovine milk. Because of no control measure for dioxin emissions in dumping sites in Asian developing countries like India, it can be anticipated that the pollution by dioxins may increase further and, hence, the residue levels in human breast milk may also increase in future, although it is claimed that the residue levels of PTS in human breast milk decreased recently in developed countries like Japan. Further investigations on the pollution sources and human exposure to PTS in Asian developing countries, especially OC insecticides and dioxins in open dumping sites, are needed to elucidate future pollution trends and to assess infant health risk.

18.6. Conclusions and recommendations

Multi-media monitoring studies and surveys have made substantial contributions to the discovery, description and understanding of the environmental problems caused by persistent man-made chemicals. Over the last three decades, comprehensive monitoring surveys using the 80,000 specimens archived in *es-BANK* have been conducted in our laboratory to make clear various issues of PTS contamination including extent of pollution, transport behavior and fate, spatial and temporal trends and toxic impacts on environmental quality, wildlife and human health. With particular emphasis on Asian developing countries, the region that recently received considerable attention due to the potential emission sources of PTS, the following conclusions can be made on the basis of these monitoring studies:

- Despite considerable efforts to curtail and/or mitigate the pollution, PTS are still ubiquitous in both biotic and abiotic spheres, particularly in Asian developing countries, where some of these chemicals are still being used in considerable quantities. In general, the hot spots of PTS pollution were found in the areas where they have been heavily used; elevated HCH residues were found in many samples collected from India and South China, which are the two largest HCH users. DDT pollution was found to be high in China and Vietnam due to the large quantity used in China and very recent application in Vietnam. A recent survey conducted using mussels and skipjack tuna as bioindicators revealed that PBDEs, a new PTS, exhibited higher concentrations in Korea, Hong Kong coasts and nearby areas in East China Sea, suggesting the presence of significant sources of PBDEs in highly industrialized zones.

- As a result of monitoring surveys in different environmental media and biological samples, the transport behavior of PTS can be characterized as "local" and "global". DDTs and PCDD/Fs have less potency for long-range atmospheric transport, and their sources are therefore located in the proximity of areas where they were extensively used. On the other hand, HCHs and, to a lesser extent, PCBs and PBDEs exhibit relatively high ability for transport through atmosphere and their contamination may extend globally in future. From the human health point of view, the toxic impacts of "localized contaminants" such as DDTs and PCDD/Fs are of great concern in Asian developing countries. In addition, from the global point of view, the regulations on the "global contaminants" like PCBs and particularly the modern chemicals, PBDEs, should be implemented in order to mitigate their potential emission to the pristine areas.
- PTS pollution in Asian developing countries and nearby seas and oceans exhibited somewhat unique temporal trends as compared to developed nations. Seas and open oceans serve as final sink for such chemicals, and time trends of OC pesticides showed a slow decline; while those of PCBs seemed in a steady state or very slow decrease. With recent rapid growth of industry and human activities, the East Asian countries such as southern China, including Hong Kong, and Korea may be a hot spot of PBDE contamination due to the extensive manufacture and dumping of computer devices and electronic appliances. As a result, our recent study on temporal trends of PBDEs in this region clearly demonstrated a substantial increase in PBDE residue levels in small cetaceans collected from Hong Kong coastal waters. In any event, temporal trends of PTS in developing countries are among the most important environmental issues that certainly deserve systematic and long-term investigations in future.
- As far as toxic impacts of PTS are concerned, the open dumping sites for municipal wastes in Asian developing countries are potential sources of many chemical contaminants. The quality of environment in

these areas has been gradually degrading with severe disturbance to the ecosystem. More serious are the long-term impacts of PTS on health of humans living in and around these dumping sites. Detection of substantial residues of DRCs in breast milk of Indian residents at particularly elevated concentrations warrants long-term impacts of dioxins in the next generation—the young children.

– With rapid growth of industrial activities in Asia-Pacific region, the use of modern chemicals such as PBDEs as flame retardants is another issue that needs particular attention. In this context, the role of the e-waste dumping sites as substantial sources of toxic chemicals such as brominated flame retardants, including PBDEs, should be elucidated in future. The increasing trends of PBDE accumulation in marine mammals stranded along Hong Kong coasts and nearby areas in East China Sea indicated that southern China with considerable amounts of electronic wastes coming from various sources all over the world continues to be a hot spot of pollution by PBDEs and other related compounds.
– Considering these critical issues of PTS pollution, it is clear that continued and constant efforts should be made to deal with the environmental problems caused by PTS. Over more than five decades when persistent organic chemicals were first used, despite considerable attempts to reduce their emissions, PTS continue to pose challenges for mankind, and scientific community is still in the initial steps to mitigate the ill effects of these toxic chemicals on the environmental quality, biodiversity and humans. Comprehensive and long-term monitoring programs are urgently needed to be implemented with close collaboration and proper capacity building in the local areas where hot spots of pollution are conspicuous. In this context, well designed monitoring of temporal trends of PTS residues in developing countries over extended period of time is crucial for tracing the unrevealed sources and predicting future prospects of their pollution state. In addition, the issue of human exposure to toxic chemicals, particularly people who are currently residing in extremely poor living conditions such as open dumping sites for municipal waste in major urban areas in developing countries, should be the prime target for future research.

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