Global Pollution Monitoring of Polychlorinated Dibenzo-p-Dioxins (PCDDs), Furans (PCDFs) and Coplanar Polychlorinated Biphenyls (coplanar PCBs) Using Skipjack Tuna as Bioindicator

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Introduction

Worldwide contamination by dioxins and related compounds, such as polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and coplanar polychlorinated biphenyls (coplanar PCBs) representing persistent organic pollutants (POPs) have been of great concern due to their persistency in the environment, highly bioaccumulative nature and adverse effects on wildlife and humans[1]. Several studies on air samples and marine organisms from open seas suggested a long range transport of these compounds through atomosphere[2-5]. Although several investigators have monitored dioxins pollution in localized areas, information on the global distribution of dioxins which can explain their atomospheric transport, behavior and fate are still limited.

Skipjack tuna is primarily distributed from offshore waters to open seas in tropical and temperate regions almost all over the world such as the Pacific, Atlantic and Indian Oceans[6]. This species is an important commercial fish and its ecology and biology has been well studied[6,7]. Moreover, suitability of skipjack tuna for global monitoring of organic pollutants (DDTs, HCHs PBDEs, organotins, etc.) has been established in our previous report[8-10], indicating that migration pattern, growth stage and sex of these animals have no or little effect on the variations of POPs residue levels in their bodies. Hence this species reflected POPs pollution levels in seawater when and where they were collected, caused by the rapid equilibrium partitioning between seawater and body lipid. These facts made skipjack tuna a suitable bioindicator for monitoring the contamination status of dioxins and related compounds. The objectives of this study are to elucidate the global distribution of dioxins (PCDD/Fs and coplanar PCBs) in offshore waters and open seas, and to understand the transport and behaviour of these chemicals using skipjack tuna as bioindicator.

Materials and Methods

The samples of skipjack tuna (*Katsuwonus pelamis*) were collected from offshore waters of various regions in the world (Japan, Taiwan, Philippines, Indonesia, Seychelles and Brazil, and the Japan Sea, East China Sea, South China Sea, Indian Ocean and North Pacific Ocean) during the years 1997-2001. Skipjack tuna were obtained from fish market and fisher village after confirming the fishing areas. The samples from the North Pacific Ocean and off-Brazil were caught by fishing during a research cruise. Muscles were taken from these individuals and kept in clean plastic bags stored at -20 °C until chemical analysis. The pooled muscles of five fish in each location were employed for chemical analysis. Chemical analysis of PCDDs, PCDFs (PCDD/Fs) and coplanar PCBs followed the method described in previous report[5].

Results and Discussion

Contamination Status

Dioxins and related compounds in the muscle of skipjack tuna were detected in almost all the specimens collected from the location surveyed, indicating widespread contamination of these compounds in the global marine environment. Concentrations of total PCDD/Fs and total coplanar PCBs ranged from less than detection limit to 79, and 4200 to 62000 pg/g lipid wt, respectively. Total concentrations of coplanar PCBs were 3 orders of magnitude higher than those of total PCDD/Fs. It may be due to the higher bioaccumulative properties of PCBs[11] and notable contamination of these compounds in offshore waters and open seas environment compared with PCDD/Fs[12]. Mean and range of PCDD/Fs and coplanar PCBs concentrations in skipjack tuna collected from offshore waters and open seas in this study were apparently lower than those in fish collected from coastal waters around Asian regions such as Japan[12-14]. It may be due to the fact that samples analyzed in this study were collected from warets far from anthropogenic activity.

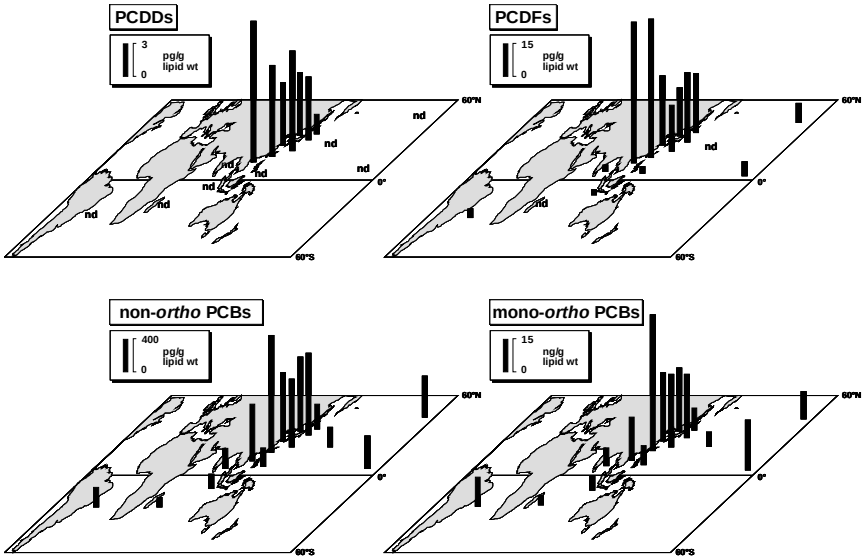

Fig. 1 Geographical distribution of dioxins and related compounds in the muscle of skipjack tuna

Geographical Distribution of Dioxins and Related Compounds

Fig. 1 shows concentrations of dioxins and related compounds in the muscle of skipjack tuna in this study. Although sampling locations in the southern hemisphere is small (off-Indonesia, off-Seychelles and off-Brazil), residue levels of these compounds in skipjack tuna from the northern hemisphere were found to be apparently higher than those from the southern hemisphere ($p<0.05$, Mann-Whitney *U* test) (Fig. 1). A similar type of distribution of these compounds was also reported in the air and avian samples from open sea[2,5]. It is plausibly due to larger usage and unintentional generation of dioxins and related compounds in the northern hemisphere.

Among the regions surveyed in this study, higher contamination levels of dioxins and related compounds were found in offshore waters around temperate Asian regions (Fig. 1). The highest

concentration of PCDDs was detected in S-China Sea (13 pg/g lipid wt), followed by E-China Sea-2 and off-Taiwan (9.2 and 8.4 pg/g lipid wt, respectively). The highest concentrations of PCDFs and coplanar PCBs were found in off-Taiwan (66 and 62000 pg/g lipid wt, respectively) followed by PCDFs in S-China Sea (65 pg/g lipid wt), and coplanar PCB in E-China Sea-2 (32000 pg/g lipid wt). These results suggest that these compounds have been used and generated in highly industrialized countries around the East China Sea and the South China Sea. However, recent study pointed out that source of dioxins is not only industrialized countries but also developing countries around Asian regions. Hileman[15] reported that massive amounts of electric products (television, computer, etc.) used in developed nations such as USA, Japan, Canada, Korea and Europe were exported as trash to Asian developing countries such as China, India or Pakistan, and these waste electric equipments were burned in open air in those countries. Combustion processes are believed to be the major source of PCDD/Fs to the environment[16]. Actually, notable contamination of dioxins and related compounds was found in soil from open dumping site in Asian developing countries, such as the Philippines, Vietnam, and Cambodia[17]. Minh et al.[17] and the present findings suggest recent formation of dioxins and related compounds in open dumping sites in Asian developing countries, and these areas can be potential sources for these chemicals to marine environment around Asian regions. Further investigations of marine environment along Asian developing countries are required. Another possible source of PCDD/Fs may be the technical PCBs used in industrialized countries around the East China Sea and the South China Sea, which can be inferred by the fact that the geographical distribution pattern of PCDFs was similar to those of coplanar PCBs (Fig. 1). It has also been reported that technical PCBs contained PCDFs as impurity[18].

Apparently lower residue levels of PCDDs (less than detection limit) were found in the samples from open ocean (N-Pacific-1 and -3) compared with those from waters around Asian regions (off-Japan, Japan Sea, E-China Sea and S-China Sea) (Fig. 1). On the other hand, comparable concentrations of PCDFs and coplanar PCBs were observed in samples from open seas (N-Pacific-1 and -3) and around Asian regions (off-Japan, Japan Sea, E-China Sea and S-China Sea). It was already shown that PCDFs and coplanar PCBs have higher vapour pressure and lower particle affinity than PCDDs[19]. These results indicated that PCDFs and coplanar PCBs may have higher transportability through atmosphere than PCDDs. Another possible reason of higher concentrations of PCDFs and coplanar PCBs than PCDDs in open seas is that these chemicals might have been released from international vessels passing through the North Pacific Ocean because technical PCBs were used on vessels as antifouling paint in the past[20].

In this context, among dioxins and related compounds, PCDFs and coplanar PCBs may have high potency to cause global pollution due to their highly transportable nature through atmosphere.

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