CONCENTRATION OF PERSISTENT ORGANIC POLLUTANTS (POPs) IN THREE SPECIES OF BALEEN WHALES IN THE WESTERN NORTH PACIFIC

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Abstract

Concentrations of PCB congeners and DDT, HCH, HCB and CHL isomers in the blubber of five mature males of each of common minke, sei and Bryde's whales taken by the Japanese Whale Research Program under Special Permit in the western North Pacific-Phase II (JARPNII) in 2011 were determined. For comparison, concentrations of these compounds in the blubber of five mature male Antarctic minke whales taken by the Japanese Whale Research Program under Special Permit in the Antarctic-Phase II (JARPAII) in 2010/11 in the Pacific sector of the Antarctic were also determined. Concentrations of PCBs were highest among organochlorines in the whales from the western North Pacific, and were much higher than PCBs concentrations in the Antarctic minke whales. Concentrations of HCB, DDTs and CHLs in Antarctic minke whales were higher or of the same order as North Pacific sei and Bryde's whales but much lower than those in North Pacific common minke whales. Differences are explained by the different trophic levels of the species and or the source of the pollutants. The accumulation of detectable 112 PCB congeners in the whale blubber samples was investigated by principal component analysis (PCA). Two significant factors, in which 72.5 % (PC1) and 8.5 % (PC2) of the total variance in the data were found. These were attributed to possible trophic level and pollution sources. The main component isomers from pesticide products originating in DDTs and HCHs were comparatively lower, although high levels of trans-chlordane contained in an insecticide were not detected in the whales from the western North Pacific. These results suggest that in the western North Pacific, a great deal of time has passed since the release of DDTs, HCHs and CHLs into the environment.

Key words: common minke whale; sei whale; Bryde's whale; North Pacific; POPs

Introduction

Persistent organic pollutants (POPs) are organic compounds that are resistant to environmental degradation through chemical and biological processes, therefore they have the potential to be transported over long distances. The manufacture, use and import/export of the specified compounds have been strictly restricted since the adoption of the Stockholm Convention on POPs by the United Nations Environment Programme in 2001 (Hagen and Walls, 2005). Among them, polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and chlordanes (CHLs) are commonly called 'legacy POPs' (French *et al.*, 2006), which had been produced and used in large quantities worldwide since the middle of last century. However, the legacy POPs remain a major environmental challenge for human health and wildlife risk.

In order to understand the fate of these compounds in the environment, it is necessary to directly monitor POPs levels in sea water and the atmosphere in the ocean areas which play a role as a final sink for POPs (Tanabe *et al.*, 1994). However, such monitoring generally produces unrealistic measures because of their extremely low concentrations and instability in the environment (Tanabe and Subramanian, 2006). Consequently, as a possible integrator of POPs, marine organisms such as mussels (Goldberg, 1986), squids (Yamada *et al.*, 1997) and skipjack tuna (Ueno *et al.*, 2003) have been investigated as bioindicators to monitor the fate and behavior of pollutants in the marine environment. Baleen whales have also been suggested as a suitable bioindicator for monitoring POPs levels in offshore waters, because they are mobile and long-lived animals, characteristics that mean that POPs can be monitored in wide sea areas and integrated in some way over time.

In the western North Pacific, yearly change of legacy POPs concentrations in marine mammals have been reported previously. Tanabe *et al.* (1994) reported temporal variation of legacy POPs in female northern fur seals (*Callorhinus ursinus*) from 1971 to 1988. The PCB and DDT concentrations showed a maximum around 1976 and then decreased until 1988. HCH concentrations decreased moderately in the research period. Aono *et al.* (1997) compared legacy POPs in blubber of common minke whales (*Balaenoptera acutorostrata*) from the western North Pacific in 1987 with those in 1994. Their results showed that the concentrations of DDTs, HCHs and HCB in 1987 were comparable or higher than those in 1994, whereas concentrations of PCBs and CHLs in 1987 were considerably higher those in 1994. On the other hand, Yasunaga Hakamada and Fujise (2016) reported that yearly changes of PCB concentrations were not observed in common minke whales from the offshore area in the period 2002-

2013. These results imply that PCB concentrations in baleen whales from the western North Pacific decreased until the end of the decade of the 1990'sand then stabilized since the 2000's. Marine mammals are good bioindicators of POPs in the marine environment, although there are limitations due to confounding factors such as sex, age, metabolism, and feeding (Tanabe and Subramanian, 2006).

The aim of the present study was to investigate the levels of concentration of legacy POPs in three species of baleen whales in the western North Pacific, common minke, sei (*B. borealis*) and Bryde's (*B. edeni brydei*) whales. Samples from the Antarctic minke whale (*B. bonaerensis*) were used for comparison. Potentially the outputs from this study can provide a better understanding of the current fate and behavior of legacy POPs in the marine environment.

Materials and Methods

Samples

Common minke, sei and Bryde's whales used in this study were sampled during the 2012 survey of the Japanese Whale Research Program under Special Permit in the western North Pacific-Phase II (JARPNII). Common minke whales were sampled in sub-area 7, sei whales in sub-area 9 and Bryde's whales in sub-areas 8 and 9, excluding the EEZ of foreign countries, which were established by the Scientific Committee of the International Whaling Commission (IWC) for management purposes (IWC, 1994) (Table 1, Fig. 1). For the analyses, only mature males were used in this study. The rational for this was as follows: (1) lipophilic POPs levels in whale body increase with age especially at young stage; (2) female whales were not used since accumulation depends on several reproductive processes such as parturition and lactation (Tanabe and Subramanian, 2006). Males of common minke, Bryde's and sei whales were defined as sexually mature by testis weight (larger side) of more than 290g, 560g and 1,090g, respectively (Bando *et al.*, unpublished data). Maturity of one common minke whale could not be identified by testis weight, however its body length (8.16 m) was much longer than the maximum body length of the males older than 14 years (mean 7.54 m; $n = 57$, Zenitani, Kato and Fujise, 2000). For the purpose of POPs analysis, blubber tissues were obtained from the central lateral part of each whale by researchers on board the research base ship. The blubber samples were stored at -20°C until chemical analyses. Chemical analyses were also conducted on samples from five mature male Antarctic minke whales sampled during the 2010/2011 survey of the Japanese Whale Research Program under Special Permit in the Antarctic-Phase II (JARPAII) in the Pacific sector of the Antarctic (Area V, 130°E-170°W),

which were used for comparative purposes (Table 1).

Laboratory analysis

Chemical analyses of the PCBs, DDTs, HCHs, HCB and CHLs were carried at the Miura Institute of Environmental Science using the standard method described by the Environmental Agency of Japan (Japan Environmental Agency, 1998), with some modifications.

Approximately 10 g of blubber were placed into a glass test tube, surrogate standard was added, and then homogenized with 20 mL of acetone and 40 mL of n-hexane. Then, the solution was filtered. This extraction procedure was repeated twice. All the solvent layers were combined and washed with 100 mL of 2% sodium chloride solution. The extracts were filtered through 20 g of anhydrous Na_2SO_4 and finally concentrated to 10 mL. Two milliliters of each of the extracts were purified using multilayer silica gel column chromatography composed of two components – 4 g of silica gel impregnated with sulfuric acid (44% mass fraction: 44% H_2SO_4–Si) and 3 g of silica gel impregnated with silver nitrate (10% mass fraction: 10%$AgNO_3$–Si). All the analytes were eluted with 200 mL of 5% dichloromethane/n-hexane. After that, the clean-up of the extract was performed by gel permeation chromatography (column: Shodex CLNpak PAE-2000AC) in order to remove remaining hydrocarbons. The mobile phase used acetone with a flow rate of 3.5 mL/min. The eluted solutions were then concentrated.

POPs were determined by a GC–MS (JEOL Ltd., JMS-700; JMS-SX102A). Concentrations of POPs were expressed on a lipid weight basis (ng/g lipid wt.). The quality control of the data was provided throughout the analyses by use of the certified reference material 'Organics in cod liver oil' (NIST 1588a). The results were 1626.4 ng/g fresh wt. for ΣPCBs, 596 ng/g for *p,p'*-DDE, 236 ng/g for pp-DDD, 417 ng/g for *p,p'*-DDT, 161.2 ng/g for cis-chlordane, 217.4 ng/g for trans-nonachlor, 67.0 ng/g for cis-nonachlor, 133 ng/g for HCB, and 78.7 ng/g forα-HCH. While the certified values by NIST were 1789.1 ng/g for fresh wt. ΣPCBs, 651 ± 11 ng/g/ for *p,p'*-DDE, 254 ± 11 ng/g for *p,p'*-DDD, 524 ± 12 ng/g for *p,p'*-DDT, 167.0 ± 5.0 ng/g for cis-chlordane, 214.6 ± 7.9 ng/g for trans-nonachlor, 94.8 ± 2.8 ng/g for cis-nonachlor, 157.8 ± 5.0 ng/g for HCB, and 85.3 ± 3.4 ng/g forα-HCH. Here, ΣPCBs includes PCB congener numbers 28, 31, 44, 49, 52, 66, 87, 95, 101, 105, 110, 118, 128, 132, 138, 149, 151, 153, 156, 159, 163, 164, 170, 180, 182, 183, 187, 190, 194 and 201 (IUPAC numbering). The detection limit was calculated in accordance with the standard method described by the

Environmental Agency of Japan (Japan Environmental Agency, 1998), which were 0.3 ng/g lipid wt. (H6CBs), 0.2 ng/g lipid wt. (α-HCH, β-HCH, oxychlordane, trans-chlordane, trans-nonachlor, cis-nonachlor, *p,p'*-DDE, *p,p'*-DDD, T4CBs, P5CBs and O8CBs), 0.1 ng/g lipid wt. (γ-HCH, cis-chlordane, *p,p'*-DDT and H7CBs), 0.08 ng/g lipid wt. (HCB, T3CBs and N9CBs), and 0.04 ng/g lipid wt. (M1CBs, D2CBs and D10CBs).

Statistical analysis

The concentrations of POPs among species were first analyzed by the Kruskal–Wallis test. Pairwise comparisons between species were then performed by the Steel–Dwass post hoc test for multiple comparisons. All differences with $p < 0.05$ were considered statistically significant. Principal component analysis (PCA) was used to assess the difference of composition of detectable PCB congeners in blubber among species caused by habitats, feeding habits and other biological factors. Principal components were derived from standardized data via the correlation matrix. PCA of all 112 PCB congeners were computed on the correlation matrix of the untransformed ng/g lipid wt. data and the projections of the factor scores of each sample along the first 2 principal component axes (PC1 and PC2) were computed. These statistical analyses were executed by SPSS ver.11 for Windows (SPSS Co. Ltd.).

Results and Discussion

Concentration of POPs

Table 2 shows the concentrations of POPs in blubber of common minke, sei, Bryde's and Antarctic minke whales. POPs were detected in all the blubber samples. Relative concentrations of POPs in the blubber of the whale species were significantly higher in the following orders, PCBs: common minke > sei; Bryde's > Antarctic minke whales; HCHs: common minke > sei > Bryde's > Antarctic minke whales; HCB: common minke; Antarctic minke > sei; Bryde's whales; CHLs and DDTs: common minke; sei; Bryde's > Antarctic minke whales ($p < 0.05$).

The POPs concentrations in the sample from the North Pacific whales were clearly higher than those in Antarctic minke whales except for HCB. Concentrations of most POPs such as PCBs levels in environmental samples were highest in northern hemisphere temperate locations (ca. 30-70 °N) where anthropogenic usage and atmospheric emissions have been concentrated, whereas relatively volatile chemicals such as HCB levels in environmental samples are related with absolute latitude in both

hemispheres due to cold condensation (Iwata *et al.*, 1993; Meijer *et al.*, 2003). The levels of PCBs were highest among POPs in the baleen whales from the northern Pacific, whereas they were comparatively lower than HCB, DDTs and CHLs in Antarctic minke whales. PCBs are still being released into the environment, whereas production and usage were banned in the 1970s in developed countries of the middle latitude of the northern hemisphere (Weber and Goerke, 2003). Furthermore, PCBs with higher lipophilicity are less transportable than the other OCs in the marine environment. Our finding indicates that PCB may still be of importance for health effects on aquatic organisms in the western North Pacific, even though PCB levels have been decreasing in marine mammals here (Tanabe *et al.*, 1994; Aono *et al.*, 1997; Yasunaga, Hakamada and Fujise, 2016).

In the western North Pacific in particular, PCBs, HCHs and HCB levels in common minke whales were higher than those in sei and Bryde's whales. This result is explained by the different feeding habit of those species. Common minke whales feed mainly on fishes such as Pacific saury and sardine while that sei and Bryde's whales feed mainly on zooplankton and smaller fishes (Konishi *et al.*, 2009). There were no differences in CHLs and DDTs levels among the three species. The effect of diet is particularly significant because the concentration of many persistent pollutants increases through the food web, and therefore tissue concentrations in marine mammals depend on concentrations of the POPs in the food organisms at the various trophic levels. The extent of biomagnification of each chemical is also influenced by other factors such as chemical and physical properties and metabolism (Aguilar, Borrell and Pastor, 1999).

Composition of PCB congeners

Table 3 shows concentrations of PCB congeners in blubber samples of common minke, sei, Bryde's and Antarctic minke whales, and Fig. 2 shows their profiles. The most prevalent congeners in all whale species were hexachlorobiphenyl CB-153, 132 and 168. In order to clarify features of accumulation of PCB congeners, PCA was conducted using the detectable 112 PCB congeners in common minke, sei, Bryde's and Antarctic minke whales. The results showed that the two principal components (PCs) represented 72.5 % (PC1) and 8.5 % (PC2) of the variance (Table 4, Fig. 3a, 3b). Focusing on PCB congener with high correlation coefficients of PC1 and low correlation coefficients of PC2, PC1 was positively correlated with mainly higher-chlorinated PCB congeners (hexa- to deca-chlorinated) and part of tetra-chlorinated congeners (Fig. 3a). PC2 was positively correlated with part of di- and tri-chlorinated PCB congeners, although it was negatively correlated with CB-11, tetra- and penta- chlorinated congeners

(Fig. 3a).

With the exception of one common minke whale having higher PC1 and lower PC2, the coefficients of PC2 of three whale species in the western North Pacific were clearly higher than those of Antarctic minke whales (Fig 3b), which can be interpreted as geographical difference of their habitats. Then, PC2 might be associated with higher levels of di- (CB-4, 5, 8 and 10) and tri- (CB-16, 25 and 32) chlorinated PCB congeners in the three baleen whales in the western North Pacific whales and higher levels of di- (CB-11), tetra- (CB-46, 69 and 80) and penta- (CB-85, 115 and 117) chlorinated congeners in Antarctic minke whales. Although there was insufficient information given for some isomers, CB-11 was unintentionally produced in certain processes of the diarylide pigment production (Hu and Hornbuckle, 2010), whereas CB-4, 5 and 8 are slightly contained in technical PCB, Kanechlors having made in Japan before 1960's (Takasuga, Inoue and Ohi, 1995; Jarman, *et al.*, 1998). Furthermore, Vorkamp (2016) established that CB-11 was the most predominant PCB congeners in the atmosphere of the Antarctic region, because only this congener is ubiquitously present globally, although other lower chlorinated congeners are easily degraded in the atmosphere. Therefore, composition of lower chlorinated congeners in the whale body would be especially influenced by environmental factors such as the distance from anthropogenic sources and latitude.

With the exception of one common minke whale having higher PC1 and lower PC2, coefficients of PC1 were in the order of common minke > Bryde's = sei >Antarctic minke whales, which is consistent with trophic levels based on their feeding ecology reported in previous studies (Konishi *et al.*, 2009; Tamura and Konishi, 2009). In the western North Pacific baleen whales, the second most prevalent congeners in common minke, sei and Bryde's whales were hexachlorobiphenyl CB-138, 163 and 160. This was negatively correlated with lower-chlorinated PCB congeners (di- to penta-chlorinated) which are rapidly metabolized and eliminated in the marine environment although they have higher mobility and therefore more available to aquatic organisms (de Boer *et al.*, 2001). This suggests that PC1 can explain the persistence of PCB congeners through the food chain in the marine environment.

In previous studies, hexa-chlorinated CB-153 was the most prevalent congener, followed by CB-138 in small cetaceans such as Dall's porpoise, striped dolphin and finless porpoise from the western North Pacific (Minh *et al.*, 2000), whereas CB-153 was also the most prevalent congener, followed by CB-128 in common minke whales from the Northeast Atlantic (Kleivane and Skaare, 1998). The difference in the secondary dominant congeners is mainly because CB-128 is partly contained in Aroclors which had been

used as PCB products in Europe, whereas it is contained in lesser amounts in Kanechlors which had been used in Japan (Kannan, Maruya and Tanabe, 1997). This suggests that levels of CB-128 in baleen whales would reflect their habitat.

Composition of DDT, HCH and CHL isomers

The averages of percentage of *p,p'*-DDT in the whale species in this study were in the order of Antarctic minke whale (20.0 %) > Bryde's whale (8.7 %) ≈ common minke whale (7.4 %) > sei whale (3.3 %) (Table 5; Fig. 4). The *p,p'*-DDE/total DDT ratio is widely used as an indicator of the time elapsed since releasing technical DDT into the environment (Borrell and Aguilar, 1987), because technical DDTs which have been used to control malaria is composed of almost all *p,p'*-DDT (De Jager *et al.*, 2006), and *p,p'*-DDT released in the environment is changed to *p,p'*-DDE in the animal body and the environment (Okonkwo *et al.*, 2008). Our results are consistent with previous reports that *p,p'*-DDE/total DDT ratios in male Antarctic minke whales had decreased for 1984/1985 (ca. 40 %) and 1992/1993 (ca. 25 %) (Aono *et al.*, 1997), and that of mature male common minke whales sampled in sub-area 7 for 1996 and 1998 was 8.9% (calculated by mean of *p,p'*-DDT 184 ng/g lipid wt. and total DDT 2,060 ng/g lipid wt.) (Fujise *et al.*, 2000). This suggests that a technical DDT might have been slightly used in the southern hemisphere.

In the ratios of the three baleen whale species in the western North Pacific, that of sei whale was higher than those of the other whales. Notably, that of sei whale was higher than that of Bryde's whale in spite of the same levels of DDT (Table 2) and food position. The production and usage of DDTs had been discontinued in the early 1970s in most developed countries, whereas they have been continued until now for malaria and leishmaniasis control in the tropical developing countries (De Jager *et al.*, 2006). Moreover, van den Berg, Manuweera and Konradsen (2017) estimated DDT usage globally declined by 30 percent from 2001 to 2014, mainly because of the reduction in India which holds 84% of the total global stockpile. Our result suggests that the lower *p,p'*-DDT ratio of sei whales compared to common minke and Bryde's whales in the northern hemisphere might be the distance of their habitat to the major technical DDT sources. This is also consistent with the fact that the ratios of common minke whale in this study was lower than that of common minke whale collected in 2006 from the Korean coast (ca. 35-40 %) (Moon *et al.*, 2010).

The averages of percentage of β-HCH accounted for over 90% of total HCHs in common minke, sei and

Bryde's whales from the western North Pacific, whereas the percentage of γ-HCH was the dominant isomer of HCHs in Antarctic minke whales (Table 6; Fig. 5). An odorless γ-HCH purified from technical HCH has been produced and used worldwide since the 1950s, although the technical HCH (α-HCH: 53–70%, β-HCH: 3–14%, γ-HCH: 10–18% and others) having a persistent bad smell and taste had been produced as a pesticide in Europe and other countries in the late 1940s and 50s (Vijgen *et al.*, 2011). After then, the mass-production base of γ-HCH moved to China, Russia and India, now leaving only a small number of producing countries (India and Romania) (Vijgen *et al.*, 2011). It is noted that γ-HCH is produced from 8 times the amount of technical HCH to it and that the residue was not properly treated in the producing countries. Therefore, HCH emissions have been almost all only γ-HCH in the southern hemisphere, suggesting that this may be reflected in the HCH levels in Antarctic minke whales.

In the three whale species in the western North Pacific, β -HCH is much higher than γ-HCH, whereas β-HCH is a minor component in a technical HCH. Because γ-HCH is rapidly metabolized, the β-HCH is consistently found in higher concentrations in mammals (Willett, Ulrich and Hites, 1988).

The percentages of CHL isomers in common minke, sei and Bryde's whales from the western North Pacific were in the order of trans-nanochlor > cis-nonachlor, whereas those in Antarctic minke whales were in the order of trans-nanochlor > oxychlordane (Table 7; Fig. 6). Technical CHLs were used for insecticide in Japan until 1986, of which 60-70% is consisted of cis and trans-chlordane (Loganathan *et al.*, 1993). Trans-chlordane was not detected in all four whale species in this study, whereas cis-chlordane was detected at relatively low levels. Ueno *et al.* (2003) reported that their trans-chlordane / trans-nanochlor ratios are in the range of 2.9-3.4, and decreasing ratio in the environment means that time has passed after technical CHLs release. This implies that the four baleen whale species from the western North Pacific and Antarctic would not have been recently exposed to new technical CHLs.

Conclusion

To examine the accumulation features and current status of POPs in the western North Pacific, the present study determined the concentrations of these compounds in the blubber of common minke, sei and Bryde's whales. The study found that there have been no recent anthropogenic inputs of legacy POPs in the western North Pacific, however, the POPs compositions in the whales might reflect their usage and fate. Understanding the historical usage and fate of these compounds may help predict their future behavior in the marine environment.

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