学　位　論　文　要　旨

Accumulation of plastic-derived polybrominated diphenyl ethers in tissues of seabirds ingesting marine plastics
（海洋漂流プラスチック摂食による海鳥へのポリ臭素化ジフェニルエーテルの蓄積）

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Increasing amounts of plastics are entering the oceans on account of increases in production and poor waste management. Plastic debris is ubiquitous in the oceans around the world. Many species of marine-based organisms, such as seabirds, ingest these plastics. The plastics cause injury and inhibit the digestion of food. Further concerns arise from the toxic chemicals both contained in the plastics as additives and adsorbed from ambient seawater. Assessing the ecological effects associated with chemicals in marine plastics depends on whether the chemicals are transferred into organisms' tissues, but there has been no clear answer.

Several studies have examined the transfer of polychlorinated biphenyls (PCBs) from ingested plastics to seabirds, though the evidence was weak because seabirds intake PCBs not only from plastics but also from their preys which biomagnify PCBs through the food web. In the present study, the author focused on polybrominated diphenyl ethers (PBDEs), which are a class of brominated flame retardants. Additive PBDEs have been detected in marine plastic fragments in the open ocean. PBDEs are compounded in plastics with high concentration (5-30% by mass), and biomagnified less than PCBs. Therefore, contribution of transfer and accumulation from ingested plastics to seabirds could be more clearly seen for PBDEs. Study on the mechanisms of the transfer was also necessary. Because PBDEs are compounded in polymer matrix and highly hydrophobic, they were supposed to be difficult to leach out. It was assumed that the stomach oil, which was made in the stomach of members of the order Procellariiformes, might act as an organic solvent and accelerate leaching.

The objectives of the present study were: 1) to examine the transfer of additive PBDEs from ingested plastics to the tissue of short-tailed shearwaters (*Puffinus tenuirostris*) in North

Pacific Ocean; 2) to examine the role of stomach oil to facilitate the transfer of plastic-derived chemicals; 3) to study the controlling factors of the accumulation of plastic-derived chemicals through comparison among seabirds of various species from different areas.

The author analyzed PBDEs in short-tailed shearwaters, a pelagic bird species in the order Procellariiformes, from North Pacific Ocean. In 5 of 30 birds, BDE209 was detected in both tissue and ingested plastics. BDE209 is not present in their natural prey, but is a main congener of deca-BDE technical product. It was suggested that seabirds ingested plastic with deca-BDE and accumulated plastic-derived BDE209 into their tissue. In the other 2 birds, similarly, transfer of octa-BDE or hexa-BDE technical products from plastic to tissue was suggested.

To understand how the PBDEs are absorbed to the biota, leaching of PBDEs from plastics into digestive fluids was studied. Pieces of plastic compounded with deca-BDE were soaked in several leaching solutions. Trace amounts were leached into distilled water, seawater, and acidic pepsin solution. In contrast, over 20 times as much was leached into stomach oil. Model calculation of PBDE exposure to birds based on results of the leaching experiments suggested the dominance of plastic-mediated internal exposure to BDE209 over exposure via preys.

The author analyzed PBDEs in tissues and ingested plastics of three species of Procellariiformes from two oceans, i.e., northern fulmar (*Fulmarus glacialis*) from North Atlantic Ocean, white-chinned petrel (*Procellaria aequinoctialis*) and shy albatross (*Thalassarche cauta*) from South African waters. Three of twenty adult northern fulmars accumulated high concentration of BDE209 in their tissues. Although no plastics were observed in the digestive tracts or no PBDEs were detected in plastics, the sporadic detection of elevated concentrations of BDE209 in the tissue was corresponded to short-tailed shearwaters, and indicated plastic-derived accumulation. Among the birds of African waters, 2 of 23 white-chinned petrels accumulated hepta- to nona-brominated congeners, whose profiles were similar to that of octa-BDE, and 2 of 5 shy albatrosses accumulated nona- to deca-brominated congeners. PBDEs were not detected in plastics in their stomachs. The congener profiles which resembled that of octa-BDE could be considered to be specific to additives. However, low concentration of BDE209 in tissue as seen in shy albatross was difficult to distinguish from background accumulation of BDE209.

In several individuals of short-tailed shearwater and northern fulmar, the ratios of BDE209 concentration in liver to those in adipose (L/A ratio) were remarkably deviated from equilibrium. High or low L/A ratio was related to recent initiation or termination of exposure to high concentration of BDE209, respectively. The non-equilibrium state may be specific to plastic-derived BDE209 exposure. The profile of debromination products of BDE209 indicated difference of metabolic mechanism among species. Further study on the metabolism of PBDEs is important to understand behaviors and toxic risks of plastic-derived PBDEs in seabirds.

The conclusion is that plastics are transported in the ocean retaining additives, and after ingestion by seabirds, chemicals in plastics can be rapidly extracted by stomach oils and exposed to the birds. Marine plastics also contain many other chemicals than PBDEs. Bioaccumulation and toxicological risks of these chemicals should be studied in future.