Doctoral Dissertation

Accumulation of plastic-derived polybrominated diphenyl ethers in tissues of seabirds ingesting marine plastics

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Abstract

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.

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Chapter 1.
General Introduction

1.1. Introduction

Problems of Marine Plastic Pollution

The consumption of plastics and the resultant generation of wastes have increased globally over the last several decades [1]. Some of the waste is released into the environment and ends up in the oceans [2]. Plastic debris is ubiquitous in the oceans around the world; it is estimated that at least 5.25 $\times 10^{12}$ plastic particles weighing 2.7×10^{5} t are currently floating at sea [3]. Plastics last a long time there because of their chemical stability. In particular, lighter polymers such as polyethylene and polypropylene float on seawater and are transported for long distances, and some of them accumulate in ocean gyres or stranded on beaches [4,5]. Plastics made of heavier polymers than seawater readily sink to the floor of the ocean [6].

Plastic debris is fragmented into smaller pieces, mainly by UV radiation, and results in the generation of a larger number of smaller particles [2]. In addition to the increment of the flow of the plastic into the ocean, their fragmentation makes this pollution more complicated and more difficult to recover. Even in the pelagic waters, increase in the amount of floating plastics is observed, e.g., the North Pacific Subtropical Gyre [7], North Pacific Central gyre [4] and the North Atlantic, during several decades [8]. The mass of plastic exceeds that of plankton at several points in the North Pacific Central gyre [4].

Marine plastics affect a wide range of species, from invertebrates to seabirds and whales [9]. The most visible and widely recognized problem is entanglement. Lost or discarded fishing gear such as nets, rope and lines may continue to catch and kill marine organisms over an extended period of time, which is called as “ghost fishing”. In addition to fishing gear, there are numerous reports of entanglement by the other anthropogenic material such as packing loops [9,10]. In the case of resident flora and fauna, smothering with plastic debris causes mechanical damage and disturbs their activity [9,10].

Ingestion of plastic debris is also found in a variety of marine organisms [10,11]. There are reports on ingested plastic in seabird and sea turtle from 1960s, and those in whale and fish from 1970s. Today, over two hundreds of species are known to ingest plastics [9]. Their motivation to ingest plastics is not clearly identified, however, in the case of turtles and birds, they are suspected to mistake plastic debris for their prey, e.g., jelly fish or crustaceans [12,13]. Ingested plastics may cause lethal damage such as penetration or blockage of the digestive tracts of marine organisms. They also cause wounds, ulcerating sores and clogging of the digestive tract and impairment of feeding [10]. In addition to these physical effects, ingested marine plastics also pose toxicological risks because of the chemicals present in them. Marine plastics retain chemicals applied during the manufacturing process, e.g., flame retardants, plasticizer and antioxidant, and also retain chemicals adsorbed from seawater, e.g., PCBs, DDTs and PAHs [14]. Some of these chemicals are known to have adverse effects, such as endocrine disruption, on organisms. There is great concern about how the mixture of toxic chemicals associated with marine plastics affects marine organisms which ingest plastics.

Assessing the ecological effects associated with chemicals in marine plastics depends on whether the chemicals are transferred from the ingested plastics into the organisms' tissues, but there is no clear answer [15,16]. To get evidence of the transfer of plastic-derived chemicals to the tissues of marine organisms is one of the most important keys to understand the ecological risk of plastics in the marine environment. In the present study, the author focused on seabirds and studied about the transfer and accumulation of plastic-derived chemicals into their tissue.

Chemicals in marine plastics and their effects on marine organisms.

Today, we can not construct our life without plastics because of their unique properties, i.e., mouldable, light weight, low cost, strong and tough [17]. The most widely used plastics are polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS) and polyethylene terephthalate (PET). They account for approximately 90 per cent of the total demand [17].

Various chemicals are added to plastics to improve their properties, e.g., plasticity, flame resistance, stability and resistance to oxidation. Although these applications append essential property to plastic products, there is considerable concern about their adverse effects on animals. For example, polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants, which have been added into plastics of products such as electrical appliances (5-30% of these products by weight [18]). They effect on neurobehavioural development, thyroid hormone homeostasis and carcinogenicity [19]. Phthalates may constitute up to 50 % of PVC products by weight, and cause developmental disorders and reproductive toxicity [20]. UV stabilizers, which are applied in plastics from 0.05% to 2% by weight [21], are suggested to have antiandrogenic activity [22], and agonistic activities toward aryl hydrocarbon receptor [23]. In addition, some polymers contain monomers and unreacted starting materials. Bisphenol A mainly used as a monomer for polycarbonate or epoxy resin, and effects estrogenic activity [24]. Styrene monomer is anticipated to be a carcinogen [25].

Marine plastics may retain many additives which are compounded into plastic products, and because of their highly hydrophobic nature, they adsorb and concentrate hydrophobic compounds such as polychlorinated biphenyls (PCBs) from sea water [26]. Hirai et al. (2011) analyzed polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), dichloro-diphenyl-trichloroethane (DDTs), PBDEs, alkylphenols and bisphenol A in marine plastic fragments (-10 mm) from the open ocean and remote and urban beaches [14]. Some of the fragments sporadically retained high concentration of chemicals such as PBDEs suggested to be derived from additives. They also detected hydrophobic organic compounds such as PCBs and PAHs, which were sorbed from seawater [14].

To assess the biological effects of plastic-derived chemicals, researchers have studied their transfer and accumulation in tissues of marine organisms [27,28]. There is increasing number of studies on transfer to invertebrate and fish by laboratory experiments, and some studies examined field caught samples of birds. All of these previous studies mainly focused on adsorbed hydrophobic chemicals.

In laboratory experiments, the accumulation of PCBs in lugworm (*Arenicola marina*) was facilitated by the existence of polystyrene in sediment [29], and another study observed the transfer of chemicals (nonylphenol, phenanthrene, PBDE-47 and triclosan) presorbed on plastic to the tissues of lugworm (*Arenicola marina*) [28]. Chua et al. (2014) exposed amphipods (*Allorchestes compressa*) to PBDEs in the presence or absence of plastics, and suggested the ability of plastic to transfer hydrophobic chemicals from water to biological tissue [30]. Rochman et al. (2013) exposed fish to plastics with chemicals presorbed in the marine environment, and found transfer of chrysene, PCB 28 and several congeners of PBDEs to fish [31]. There are also some laboratory studies about adverse effects of plastic ingestion on invertebrate [28,32] and fishes [31,33-37]. Some of these studies emerged the evidence of the impacts associated with chemicals derived from plastics [28,31,33-35], e.g., alteration of feeding behavior in lugworm [28], structural change of tissues and alteration of some biomarker responses in fish [31,34].

Study on seabirds in the field samples also showed some evidences of chemical transfer. A weak correlation between the mass of ingested plastics and the concentration of lower-chlorinated biphenyls in adipose tissue of twelve short-tailed shearwaters suggested the transfer of the chemicals from the plastics to the birds [38]. Great Shearwaters (*Puffinus gravis*) also showed the correlation between the mass of ingested plastics and PCBs concentration in the adipose tissue and eggs [27]. However, the correlation was weak, as explained by the exposure of the seabirds, as the top predators, to PCBs in their prey as well as by the ingestion of plastics. Marine plastics adsorb hydrophobic chemicals in the marine environment, and at the same time, marine organisms also concentrate them. Moreover, the chemicals can be biomagnified in food chain if the chemical does not have low bioavailability or high biodegradability. To get clearer evidence for the transfer of chemicals from ingested plastics to seabirds in the field, we must focus on chemicals which have larger exposure from ingested plastics than exposure from their preys. In this study, the author focused on PBDEs, which are applied in plastic products as additive, and the higher-brominated congeners of which are not biomagnified [39].

Plastic ingestion by seabirds and the risk of chemical exposure.

Plastic ingestion by seabirds is investigated for a long time, at least four decades [40]. Seabirds ingest mm- to cm-size plastics, and most of ingested plastics are accumulated in gizzard [41,42]. It is suggested that they mistake plastics for prey [43]. There is a relation between feeding ecology and frequency of plastic ingestion; Day (1980) detected plastics in 15 of 37 Alaskan seabird species and found more than 25% was feeding by pursuit-diving, 16% was feeding by surface-seizing, 9% was feeding by dipping, and none of those feeding by plunging or piracy contained plastic [44]. One study estimates that the frequency of plastic ingestion is increasing, and it will reach 99% of all seabird species by 2050 [45].

In this study, the author focused on Procellariiformes. Procellariiformes includes Diomedeidae and Procellariidae, and they show the highest frequency of plastic ingestion among seabirds [46]. In addition to the frequency, Procellariiformes are expected to be at high risk for chemical exposure from plastics because of their unique nature. Additives are compounded in a polymer matrix, and especially, hydrophobic chemicals are stable in plastics. Therefore, these chemicals are supposed to be difficult to leach out, and leaching of chemicals is the most important process in transfer of chemicals from plastic to biological tissue. First, Procellariiformes do not usually regurgitate ingested plastics and other indigestible items, therefore, they accumulate more plastics in the stomach than the other species such as gulls [47,48]. Accumulated plastics in gizzard are ground up and excreted after they wear down or fragment into sizes small enough to pass into the intestines [47]. Retention time of plastics in stomachs is estimated to be ranged from a half month to more than a year, by some field observation [44,47] and an experimental work [49]. The long retention time of plastics and grinding process in gizzards may cause high efficiency of extraction of chemicals retained in plastics. Next, the stomach oils of seabirds may act as an organic solvent and accelerate leaching. The stomachs of members of the order Procellariiformes hold oils derived from their diet, mainly fish. Stomach oils are composed mainly of wax esters or triacylglycerol (>70% of total lipids) [50,51] and could therefore facilitate the leaching of hydrophobic chemicals from ingested plastics. Thus, seabirds, especially in the order of Procellariiformes, may extract additives from plastics in their stomach and can be exposed to high concentration of the chemicals.

1.2. Purpose of this study

This study examined the accumulation of plastic-derived chemicals in tissue of seabirds, especially in the order Procellariiformes. The hypothesis is that plastics with applied chemical additives is transported in the marine environment retaining additives, and after ingestion by seabirds, chemicals in plastics are extracted in the stomach of seabirds. The author focused on PBDEs, which applied in plastic during the manufacture and can be contained in ingested plastic at much higher concentration than prey. The transport of PBDEs from plastics to seabirds was examined. Plastic debris distribute all over the ocean and many avian species are found to ingest plastics, therefore, the chemical pollution caused by plastics may occur in many species from different oceans.

The objectives of the present study are: 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.

Chapter 2.

Measurement of PBDEs in field-caught seabirds

2.1. Introduction

2.1.1. Objectives

There is great concern about the effect of toxic chemicals associated with marine plastics on marine organisms which ingested plastics. To get evidence of the transfer of plastic-derived chemicals to marine organisms is one of the most important keys to understand the ecological risks of plastics in the marine environment. Marine plastics retain additives that are compounded into plastic products [14]. Among the chemicals present in plastic fragments, the author focuses on polybrominated diphenyl ethers (PBDEs) as additives, because they are compounded in plastic with relatively high concentration [18]. The objective of the studies in this chapter is to get evidence of the transfer of PBDEs from ingested plastics to the tissues of seabirds through the analysis of PBDEs in seabird's tissue and ingested plastics.

2.1.2 Seabird samples

To examine the accumulation of plastic-derived chemicals in seabirds, the author analyzed PBDEs in tissue and ingested plastics of field caught short-tailed shearwaters (*Puffinus tenuirostris*) from North Pacific Ocean.

Short-tailed shearwaters (*Puffinus tenuirostris*)

Short-tailed shearwaters migrate annually from breeding grounds in southeastern Australia, including Tasmania (from October to March), to forage in the northern North Pacific (from May to September). In both regions, they feed on pelagic species such as crustacea and fish [52]. Short-tailed shearwaters are one of the species which ingest plastics the most frequently [12,53]. Ingestion of plastics by short-tailed shearwaters has been reported since the 1970s [44,54].

2.1.3. Target compounds

Polybrominated diphenyl ethers (PBDEs)

PBDEs are a class of brominated flame retardants, and compounded into various plastics at 5% to 30% by weight. Three commercial mixtures are used (penta-, octa-, and deca-BDEs), along with some minor technical mixtures such as hexa-BDEs. Penta- and octa-BDEs are now globally regulated by the 2009 Stockholm Convention because of their toxicity potential. Although deca-BDE is not yet banned globally, deca-BDE is now being phased out in many countries and the eleventh and twelfth meeting of the Persistent Organic Pollutants Review Committee (POPRC) recommended to list decabromo-diphenyl ether in Annex A. Large amounts of plastic products containing deca-BDE are still used or landfilled, and deca-BDE has been detected in marine plastic fragments in the open ocean [14].

2.2. Materials and Methods

2.2.1. Sample collection and treatment

Thirty short-tailed shearwaters were caught as by-catch in experimental driftnets by the research vessel *Wakatake-maru* (Hokkaido Prefectural Government) in the northern North Pacific Ocean (56°30′–57°30′N, 179°00′E–178°00′W) during June to July 2008 to 2010, and the training ship *Osyoro-maru* (Hokkaido University) in the northern North Pacific Ocean (41°44′–43°57′N, 155°00′–165°00′E) during May to July 2010 and 2011. Collection on the *Wakatake-maru* was approved as a part of the Bering/Aleutian Salmon International Survey (North Pacific Anadromous Fish Commission 2001), but no permission was required for the collection on the *Oshoro-maru*.

The carcasses were stored at –30 °C until dissection. In the laboratory, the carcasses were dissected with a solvent-rinsed stainless steel knife. Sets of abdominal adipose, liver, and plastics were collected from short-tailed shearwaters sampled during 2008-2011. Abdominal adipose and plastics were collected from short-tailed shearwaters sampled in 2005.

The tissues were removed, put in solvent-rinsed glass vials, and stored in the freezer at –30 °C until analysis. The stomachs were removed to examine the contents. Plastic pieces found in the stomach were washed in distilled water, dried at room temperature, and weighed on an electronic balance (AB54, Shimadzu, Kyoto, Japan) with precision of 0.1 mg. The plastics were identified and sorted according to polymer type by near-infrared spectrometry (PlaScan-W, OPT Research Inc., Tokyo, Japan). Identification was done by comparison of infrared spectra with those in a library (e.g. [55]). After sorting, they were ultrasonicated in distilled water and dried at room temperature.

Specimens of lanternfish (Myctophidae) and squid (Gonatidae), prey species of short-tailed shearwaters, were collected in midwater trawl nets from the training ship *Oshoro-maru* (Hokkaido University) in the northern North Pacific Ocean (NNPO) as follows. Three individuals of lanternfish and one squid were collected in the eastern part of NNPO (39°300 –43°150 N, 155°000 E) in May 2012. Another three individuals of lanternfish were collected in the western part of NNPO (45°330 N, 159°210 W) in July 2012. The fishes and squid were analyzed whole.

2.2.2. Extraction and clean up of tissues and plastic samples

PBDEs in abdominal adipose tissue and ingested plastics of all samples were analyzed, and additionally PBDEs in liver were also analyzed for 18 of 30 short-tailed shearwaters.

All solvents were distilled in glass. All glass and stainless steel equipment were rinsed with the distilled solvents before use. Separate sets of equipment were used for the tissues and the plastic to avoid cross-contamination. If high concentrations of PBDEs were detected in a sample, the glassware used for the sample was not used again. Approximately 5 g (wet weight) of liver (from the right lobe), 1 g (wet weight) of abdominal adipose tissue and 10 g (wet weight) of lanternfish were first weighed and then extracted in a Polytron PT2000 homogenizer with dichloromethane (DCM), and the homogenate was then dried with anhydrous sodium sulfate. One gram of stomach oil was diluted with DCM to 2 mL. Plastics from each individual were analyzed together as a single sample. The samples were Soxhlet-extracted with 150 mL of DCM at a rate of 3–4 cycles per hour for 24h, the extract was concentrated to ~2 mL, and 3–4× volume of *n*-hexane was added to enhance the eduction of dissolved plastic. One-tenth of the supernatant was used for analysis.

The sample extracts were spiked with surrogate standards (^{13}C-labeled BDE3, 15, 28, 47, 99, 153, 154, and 183, and 4′-fluoro-2,2′,3,3′,4,5,5′,6,6′-nonabromo-diphenyl ether) and those of biological samples were purified by centrifugation (737× *g* for 30 min) and then, an aliquot (normally one-tenth) was subjected to determine lipid contents from the weight of the dried extracts. Another aliquot was rotary-evaporated just to dryness. The residue was dissolved in 1 mL of *n*-hexane/DCM (3:1, v/v) and transferred onto a 5%-H_2O-deactivated silica gel column (1 cm i.d. × 9 cm). PBDEs and PCBs were eluted with 20 mL *n*-hexane/DCM (3:1, v/v). Eluent was concentrated just to dryness in a rotary evaporator and topped up to ~2 mL with DCM, subjected to GPC (2 cm i.d. × 30 cm, CLNpak PAE-2000; Showa-denko, Tokyo, Japan) to separate target compounds from lipids in DCM at an eluent at flow rate of 4 mL/min. The fraction with a retention time of 15.5 to 25 min was collected for further purification. The GPC eluent was rotary-evaporated just to dryness, re-dissolved into 0.4 mL of *n*-hexane, and transferred onto fully activated silica gel column (0.45 cm i.d.× 18 cm). The first fraction containing alkanes and the second fraction containing PBDEs and PCBs were eluted with 4 mL of *n*-hexane and 10 mL of *n*-hexane/DCM (3:1, v/v), successively. Eluent in the second fraction was rotary-evaporated and transferred to a 1 mL glass ampoule. The solvent in the ampoule was evaporated just to dryness under gentle nitrogen stream, and the residue was re-dissolved into 100 µL of *iso*-octane containing 40 ppb of ^{13}C-labeled BDE77 and ^{13}C-labeled BDE139 as internal injection standards.

2.2.3. Instrumental analyses

One microlitre aliquots were analyzed by gas chromatography. Purified fractions were analyzed by GC/ion trap mass spectrometry (GC/IT/MS) for mono- to hepta-BDEs, and di- to deca-chlorinated biphenyls by GC-ECD for higher-brominated congeners. The author quantified 49 BDE congeners (BDE 1, 2, 3, 7, 8, 10, 11, 12/13, 15, 17/25, 30, 32, 33/28, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 116, 118, 119, 126, 138, 153, 154, 155, 166, 179, 181, 183, 188, 190, 196, 197, 202, 203, 206, 207, 208, and 209). Compounds were identified and quantified against native standards. Detailed operation conditions are listed in Table 2-1a,b.

Concentrations are expressed on a lipid weight basis for the tissue samples. In the case of ingested plastic samples, analytical results are expressed as amounts per individual (i.e., ng/individual), because all the plastic pieces from each individual were analyzed as a single sample to ensure enough for analysis. Concentrations in the samples were corrected against the recovery of the surrogates. The limit of detection (LOD) was set at 3 times the signal-to-noise ratio on the detector. LOD values for individual runs are listed in Table 2-2. The limit of quantification (LOQ) was set at 3 times the amount detected in the procedural blank. Typical blank values were 0.01 ng/g-lipid weight for BDE47, 0.01 ng/g for BDE183, and 0.09 ng/g for BDE209, though they were variable among the runs. Blank values for individual runs are listed in Table 2-3.

Reproducibility and recovery of the analytical procedures for the tissue samples were confirmed in advance through 4 replicate analyses of adipose tissue extracts with and without spiking of native PBDE standards. The relative standard deviations of concentrations of individual congeners were <10% and the recoveries were >87%. A procedural blank was run with every batch (7 samples).

2.2.4. Statistical analysis

The similarity of congener compositions of PBDEs among samples was compared by cluster analysis with the complete linkage method. The open-source software R was used.

2.3. Results and Discussions

2.3.1. Short-tailed shearwaters of North Pacific Ocean

2.3.1.1. PBDEs in ingested plastics and tissues

All 30 birds examined held plastics in their stomach (e.g., Fig.2-1), at 0.003–2.16 g per bird (Table 2-4). Fragments of end-products accounted for 63.6% of all pieces, followed by plastic sheets (18.9%), resin pellets (9.6%), and fibers (7.7%). The proportions of plastic types were similar to those found previously in short-tailed shearwaters [38,42]. On average, the author found 24.4 pieces and 0.28 g per bird. This mass was greater than those in previous studies of short-tailed shearwaters in the same region [38,42,53].

Although all 30 birds held plastics in their digestive tracts, only eleven (OS10-001, OS10-002, OS10-004, OS10-008, WK10-016, WK10-018, WK10-023, WK05-018, WK05-022, WK05-027, and WK05-030; Fig. 2-2, Table 2-5) had PBDEs higher than 0.1 ng/individual in the plastics, above which discussion about congener profiles would be meaningful. The congeners in the plastics were dominated by higher-brominated congeners, i.e., BDE209 and several octa- to nona-brominated congeners or hepta- and nona-brominated congeners in all birds except OS10-001, in which hexa- and hepta-BDEs were detected and BDE153 was predominant. These congener profiles are similar to that of deca-BDE technical product containing BDE209 as a major component and octa-BDE technical product containing BDE183 as a major component, respectively (Fig. 2-3) [56]. These results indicate that some of the plastics ingested by the short-tailed shearwaters were fragments of plastic products in which deca-BDE was industrially compounded. This sporadic occurrence of plastics containing deca-BDE technical product is consistent with that of plastics floating on the open ocean, including in the Central Pacific Gyre [14], and the fact that brominated flame retardants are added to specific plastic products.

PBDEs were detected in all birds in both the abdominal adipose (Fig. 2-2A) and liver (Fig. 2-2B). Some congeners (e.g., BDE154) were detected in all the samples, whereas the other congeners were not detected in some samples. Total PBDEs in the adipose and liver ranged from 0.28 to 186 ng/g-lipid weight with median of 1.90 ng/g-lipid (Table 2-6) and 0.07 to 65.9 ng/g-lipid weight with median of 0.71 ng/g-lipid weight (Table 2-7), respectively. The concentrations were comparable to those found in species living in pelagic waters and on remote islands (e.g. [57]), and much lower than those found in species living in human-affected areas (e.g. [58]). The lower contamination in short-tailed shearwaters was due to their pelagic behavior and their reliance on pelagic plankton and fishes that are distant from the sources of PBDEs.

In many of the birds, profiles were dominated by lower-brominated congeners (tetra- to hexa-brominated). These profiles, with a dominance of lower-brominated congeners, are similar to those found in pelagic fishes (Fig. 2-4, Table 2-8), the prey of shearwaters. These data indicate that these lower-brominated congeners are accumulated in the body of the seabird through the food web.

On the other hand, in the liver or abdominal adipose of six birds, higher-brominated congeners were dominant (BDE209 and octa- and nona-brominated congeners were dominant in five birds, OS10-008, WK10-019, WK10-023, WK05-018, WK05-022; and hepta- and nona-brominated congeners were dominant in one bird, WK05-027). In particular, the adipose of WK05-022 and the liver of OS10-008 showed the highest concentrations of PBDEs (186 ng /g-lipid and 65.9 ng/g-lipid, respectively), especially BDE209 (105 ng/g-lipid, and 32.1 ng/g-lipid, respectively).

The predominance of higher-brominated congeners in the tissues cannot be ascribed to prey, which lack those congeners. Their profiles, especially in the tissue of OS10-008, WK10-023, WK05-018, and WK05-022 are dominated by BDE209 and are similar to those in deca-BDE technical product, suggesting exposure from plastic additives. In fact, OS10-008, WK10-023, WK05-018, and WK05-022 had BDE209 in the ingested plastics and similar congener profiles. In the cluster dendrogram (Fig. 2-5), samples of tissue and plastic from OS10-008, WK10-023, WK05-018 and WK05-022 were clustered together. These similarities in congener profiles and the sporadic accumulation of high concentration of higher-brominated congeners in tissue indicate the transfer of PBDEs from the plastic to the tissues. The profile in adipose of WK05-027 was dominated by heptato nona-brominated congeners, especially BDE183. In addition, BDE183 were detected in the ingested plastics in WK05-027, of which profile is similar to that in octa-BDE technical product. This indicates that these congeners are derived from the plastics.

Plastics in the gizzard of OS10-001 showed a unique congener profile, i.e., a predominance of BDE153 with minor proportions of BDE154 and BDE183 (Fig. 2-6, Table 2-5). This profile is very similar to that of hexa-BDE in a unique commercial product manufactured by a Japanese company in the past [59]. The similarity indicates that the plastics contained the hexa-BDE product. The tissue of OS10-001 had the same profile with the predominance of BDE153, with the highest concentration among all birds examined. The concentrations of BDE153 were 1.4 ng/g-lipid in the liver and 12.7 ng/g-lipid in the adipose of OS10-001, each an order of magnitude higher than in the other birds (means of 0.14 and 0.50 ng/g-lipid, respectively). This correspondence of the profile and higher concentrations of BDE153 in OS10-001 is another indicator of the transfer of additive PBDEs from ingested plastics to tissues.

In the comparison of PBDE congener profiles, the author replaced values below LOQ with 0. This substitution (option 1) may have underestimated concentrations of some congeners in some samples and therefore affect the congener profiles and the comparison. So the author examined two other options. In option 2, the author used actual values between LOD and LOQ, and LOD for “not detected”. In option 3, the author used actual values between LOD and LOQ, and 0 for “not detected”. The four individuals (OS10-008, WK05-018, WK05-022, and OS10-001) showed similarities in PBDE congener profiles between tissues and plastics in all options (Figs 2-7 and 2-8). Option 1 would give lower limits, while option 2 would give higher limits. Real values must exist between option 1 and option 2. All the options quantitatively indicate the similarities in PBDE congener profiles between tissues and plastics for OS10-008, WK10-023, WK05-018, WK05-022, and OS10-001. This means that the similarities exist in the real world.

However, BDE209 was detected in the tissues of WK10-019 but not in the ingested plastics. A likely explanation is that plastics with BDE209 had been ingested and retained long enough for PBDEs to be leached and transferred to the tissues, before being broken up in the gizzard and excreted. The congener profile of PBDEs in the tissues of WK10-019 supports this process, as discussed in the next section.

On the other hands, BDE209 was not dominant in the tissues of four birds (OS10-002, OS10-004, WK10-018, and WK05-030) in which it was dominant in the ingested plastic. These inconsistencies may be explained that the plastic might not have been in the stomach long enough for the congener to be leached out and absorbed by birds. Because additives are compounded in a polymer matrix and PBDEs are highly hydrophobic, PBDEs are stable in plastics and supposed not to leach out easily. They can also be explained by the low availability of higher-brominated congeners of PBDEs.

Because of the high molecular weight, the adsorption of higher-brominated congeners in seabirds progress slowly. In addition, it takes time for the transfer of chemicals among organs. For example, although OS10-008 ingested plastic with the highest amount of PBDEs, its highest concentration of BDE209 in the liver but low concentration in the adipose tissue suggest that the transfer of BDE209 in the tissues was still under way. For more detailed discussion, the author additionally analyzed PBDEs in subcutaneous adipose tissue, heart, muscle, and gizzard of OS10-008 (Fig. 2-9). BDE209 and higher-brominated congeners are highly accumulated in liver, however, all of the other tissues accumulate lower than one tenth of PBDEs in OS10-008 on lipid weight basis, and especially low concentration and low proportion of BDE209 were observed in abdominal and subcutaneous adipose tissues (Fig. 2-9). This distribution of PBDEs indicates that BDE209 absorbed in intestine is first accumulated in liver, and then distributed to the other tissues and lastly accumulates in adipose tissues. Because adipose mainly works for lipid storage, the exchange of substances in adipose is less active than the other tissues.

2.4. Conclusion

The author detected sporadic accumulation of higher-brominated congeners of PBDEs in tissue of short-tailed shearwater. Seabirds which ingested plastics with deca-BDE accumulated BDE209 from plastics into their tissues, and seabirds which ingested plastics with octa-BDE (dominated by BDE183) or hexa-BDE (dominated by BDE153) also accumulated corresponding congeners. It is indicated that plastic-derived PBDEs were transferred and accumulated in seabirds' tissue.

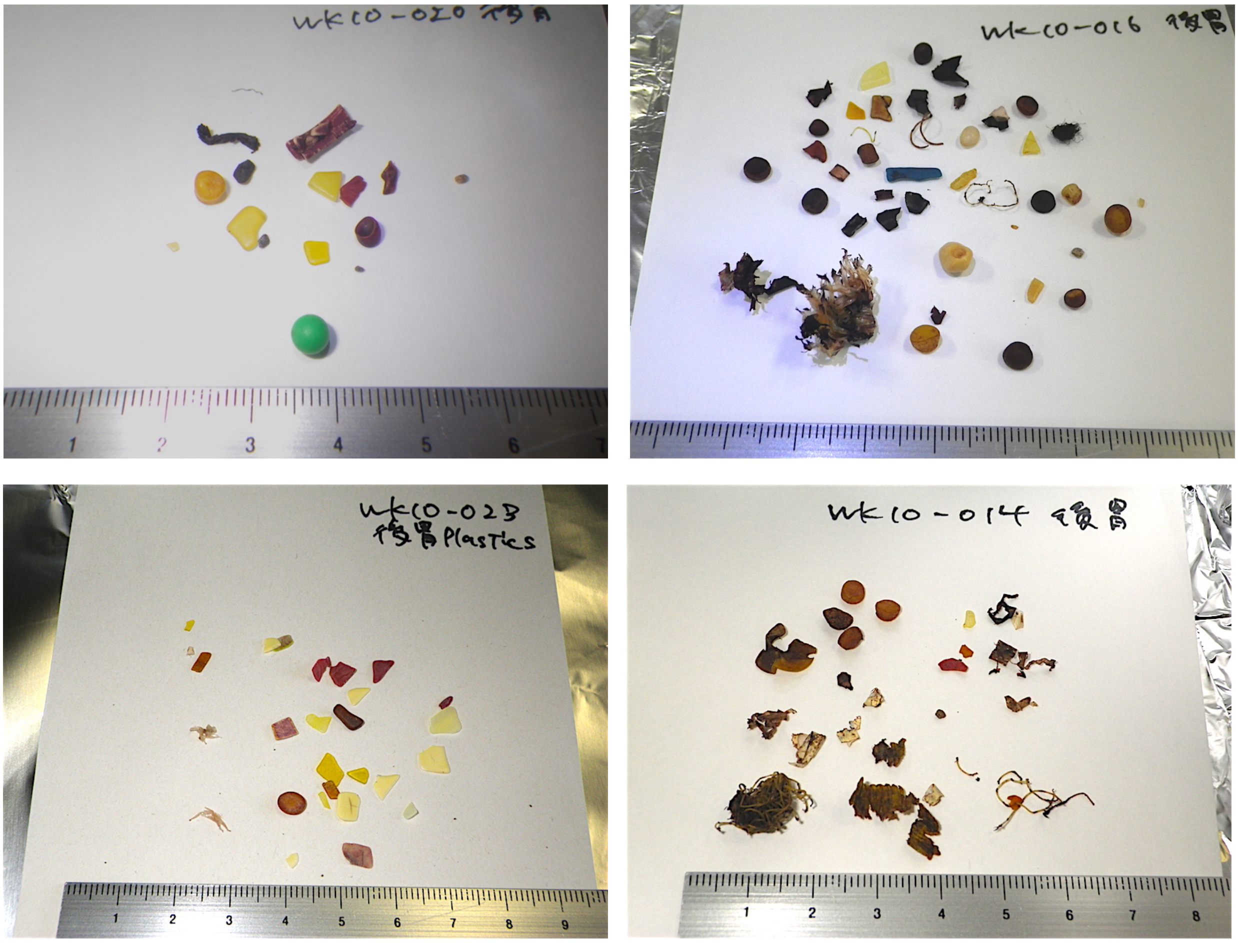


Figure 2-1. Examples of plastics in the gizzard of short-tailed shearwaters.

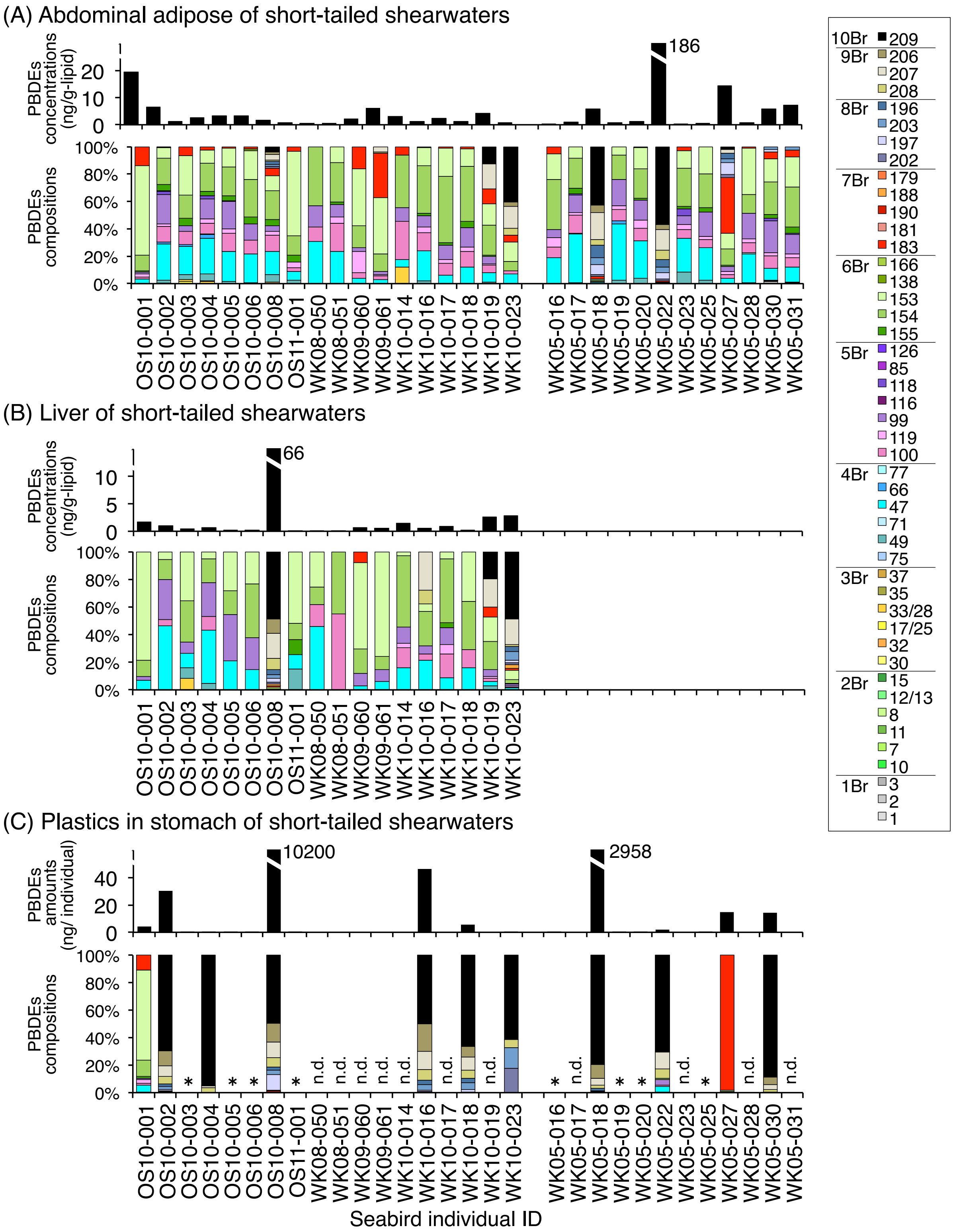


Figure 2-2. PBDE concentrations and compositions in the (A) abdominal adipose, (B) liver, (C) plastics in the stomach of short-tailed shearwaters. Asterisks indicate that the profile is not shown because only trace concentrations of one to three congeners (among BDE17/25, BDE47, BDE154, and BDE183) were detected; n.d. indicates not detected.

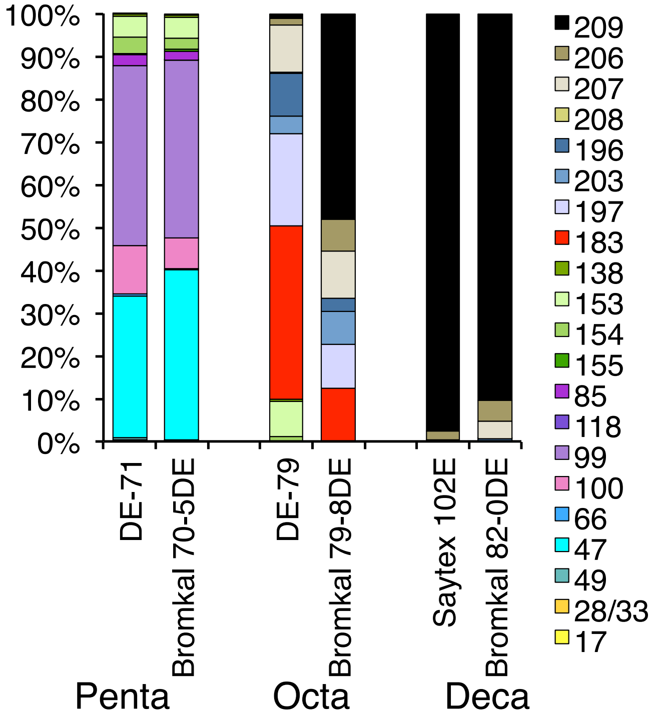


Figure 2-3. Congener profiles of PBDE technical products. (La Guraudia et al., 2006 [56])

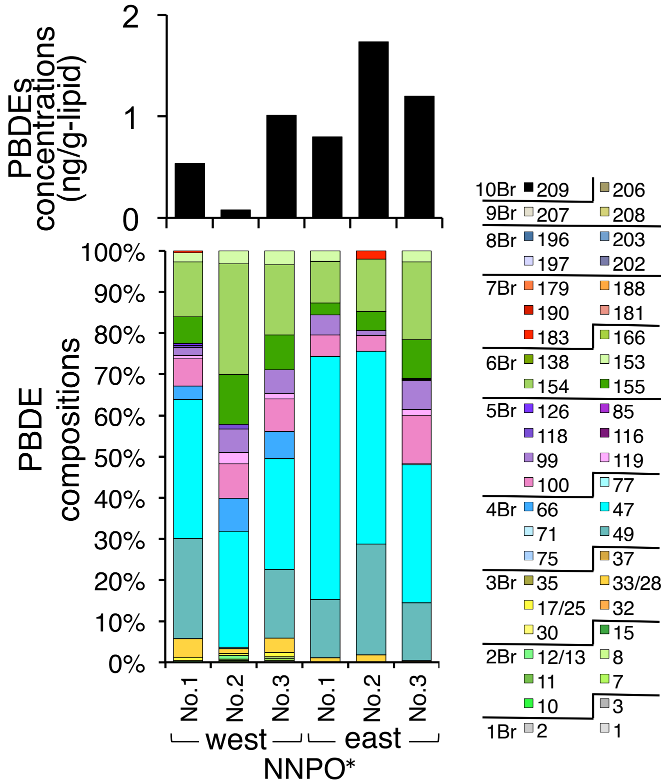


Figure 2-4. PBDE concentrations and compositions in lanternfish of Northern North Pacific Ocean. *NNPO: Northern North Pacific Ocean.

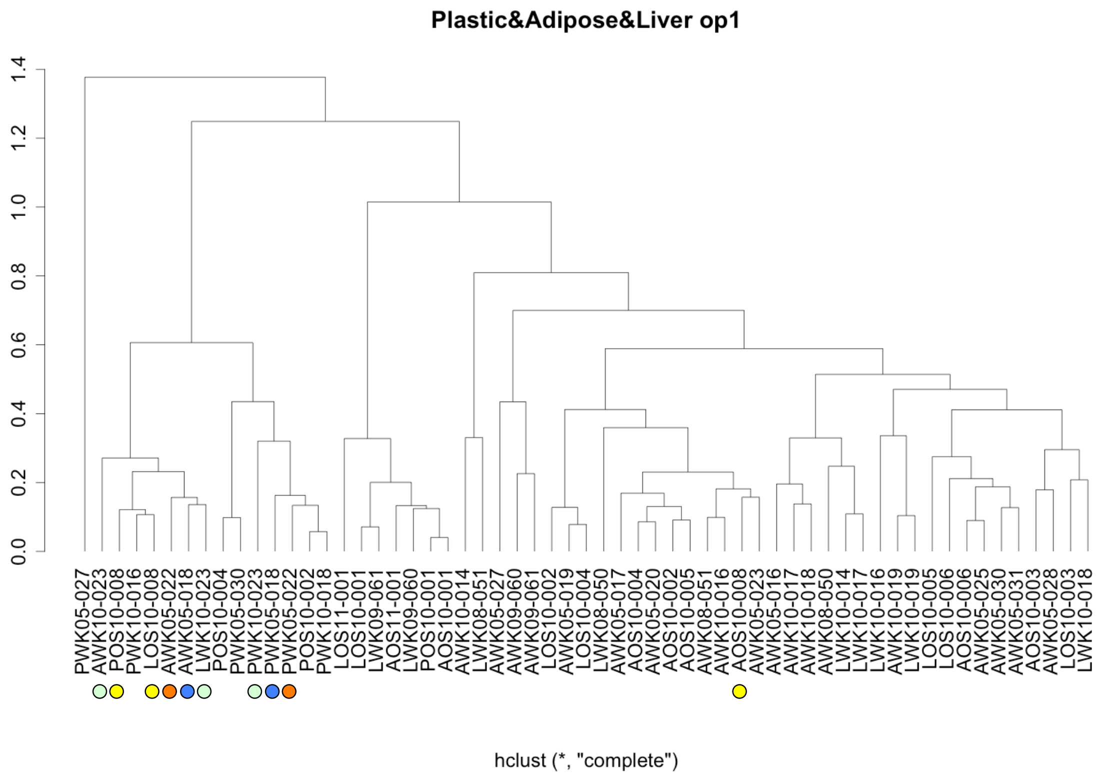


Figure 2-5. Cluster dendrogram of PBDE congener profiles in liver (L-), abdominal adipose (A-), and plastics (P-) in stomach of short-tailed shearwaters. Option 1: Values < LOQ were replaced with 0.

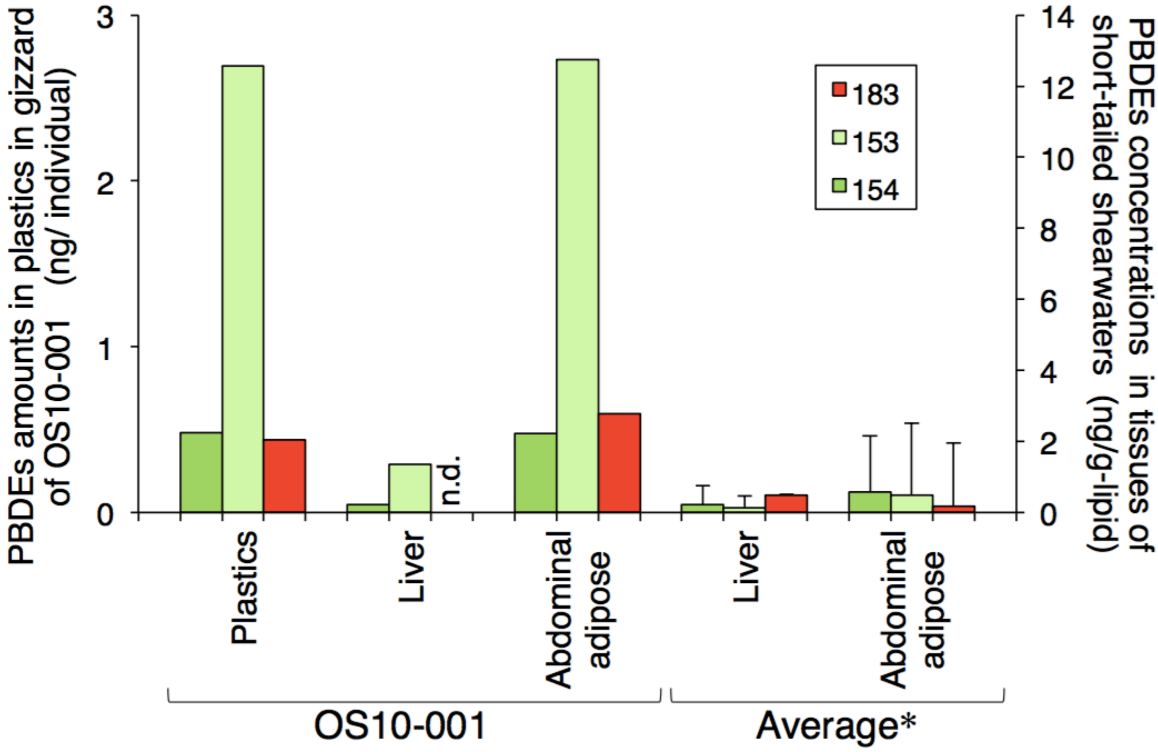


Figure 2-6. Concentrations of BDE183, BDE153, and BDE154 in plastic in the stomach, liver, and abdominal adipose tissue of short-tailed shearwaters. Asterisks indicate that the average of individuals other than OS10-001; upper bars represent the highest concentrations of each congener among the birds other than OS10-001; n.d. indicates not detected.

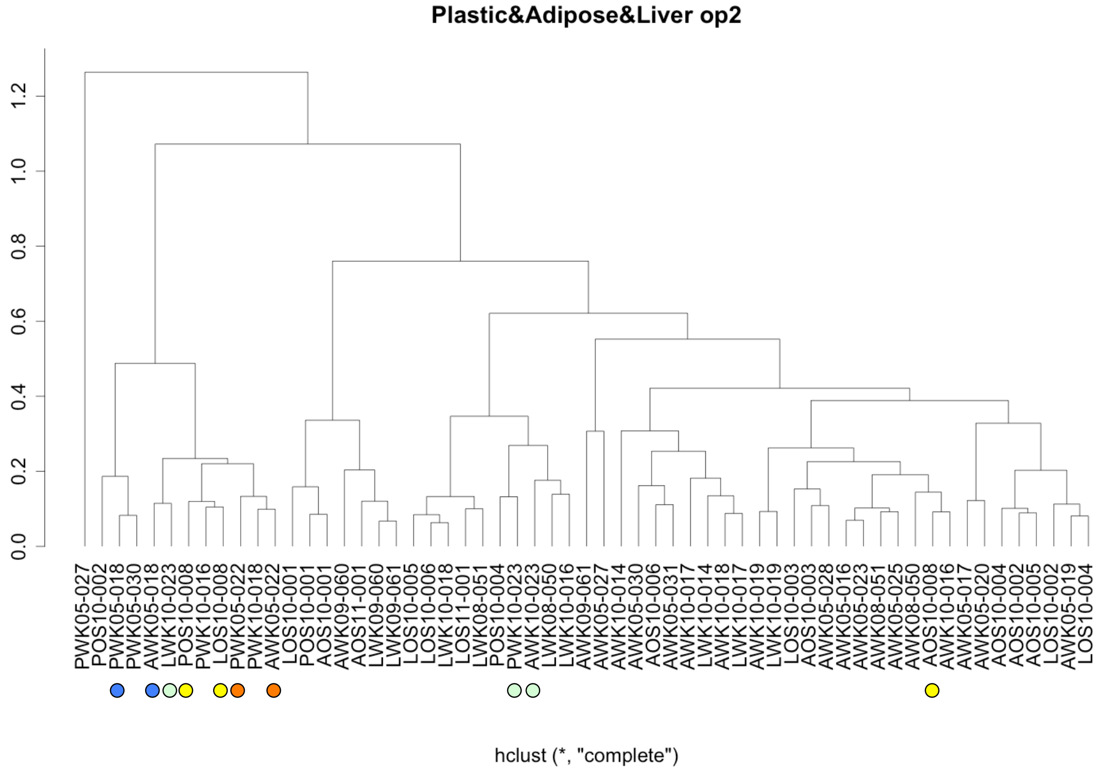


Figure 2-7. Cluster dendrogram of PBDE compositions in liver (L-), abdominal adipose (A-), and plastics (P-) in stomach of short-tailed shearwaters. Option 2: Actual values between LOD and LOQ were used, and LOD was used for “not detected”.

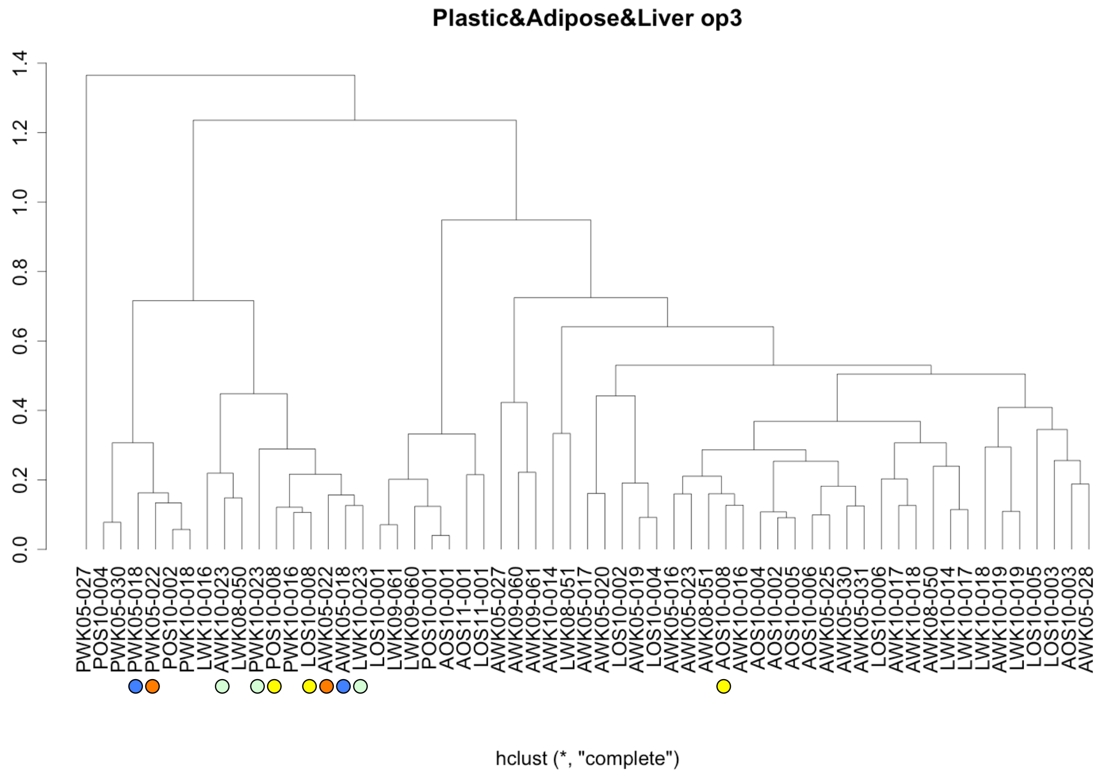


Figure 2-8. Cluster dendrogram of PBDE compositions in liver (L-), abdominal adipose (A-), and plastics (P-) in gizzard of short-tailed shearwaters. Option 3: Actual values between LOD and LOQ were used, and 0 was used for "not detected".

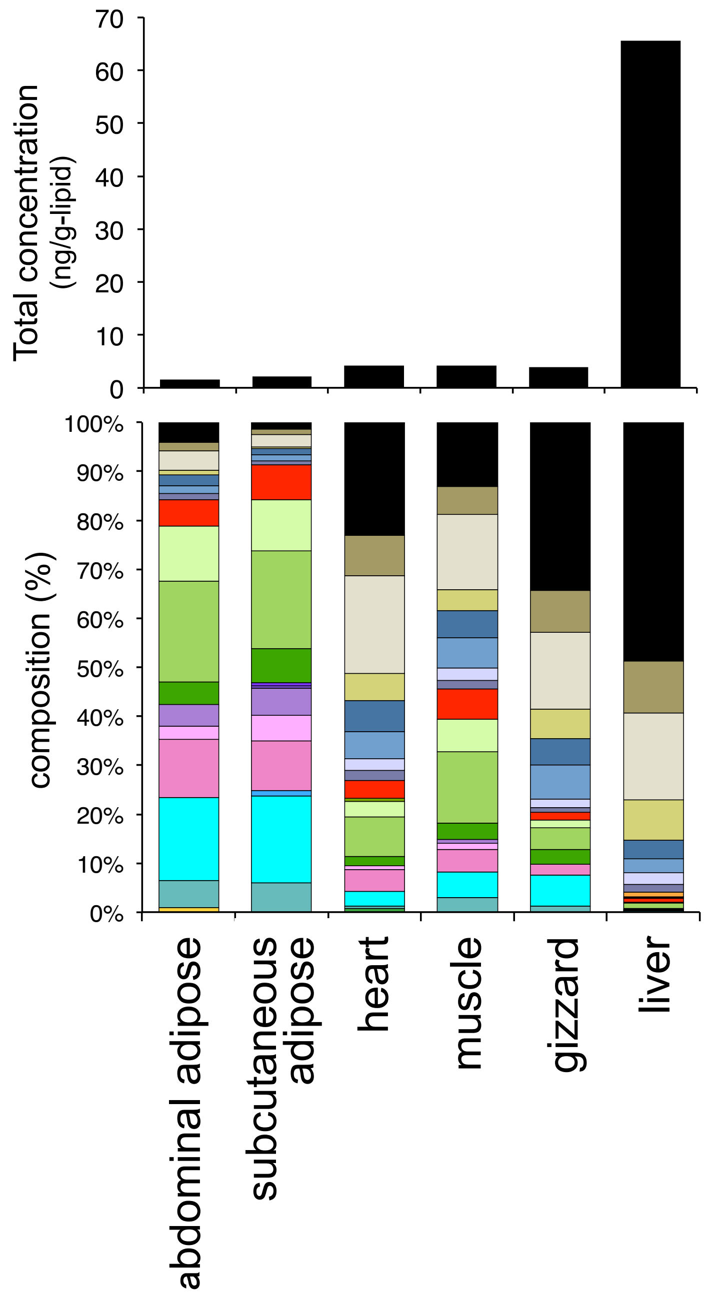


Figure 2-9. PBDEs concentration and composition in abdominal adipose, subcutaneous adipose, heart, muscle, gizzard, and liver of OS10-008.

Table 2-1a. Instrumental conditions for hepta- to deca- brominated congeners.

GC: Agilent Technologies 7890A GC system	
Column	J&WScience DB-5 (0.25mm i.d. x 15m)
injection temp.	250C
carrier gas	Helium
detector temp	300C
initial temp	80C
initial time	2 min
purge	2 min
rate	15 C/min
final temp	310 C
final time	10 min
detector	ECD

Table 2-1b. Instrumental conditions for mono- to hepta- brominated congeners.

GC: TRACE GC ULTRA, injector: AS2000(Thermo fisher scientific)	
column	J&WScience DB-5MS (0.25mm i.d. x 30m)
carrier gas	Helium
mode	constant flow
initial value	on
initial value	1.00 mL/min
initial time	1 min
gas saver	on
gas saver flow	20mL/min
gas saver time	2 min
vaccume compensation	on
transfer line temp.	250C
injection temp.	250C
mode	splitless
split flow	10 ml/min
splittless time	1 min
surge pressure	off
MS:POLARIS Q (Thermo fisher scientific)	
acquisition time	GC run time
cal. gas	off
reagent gas	off
acquire profile	no
source temp.	250C

Table 2-2. LOD values for individual runs.

Batch code	A	B, C	D	E
	Liver (ng/g-lipid)	Adipose (ng/g-lipid)	Plastic (ng/individual)	Plastic (ng/individual)
BDE-1	0.12	0.09	0.03	0.31
BDE-2	0.01	0.01	0.003	0.05
BDE-3	0.01	0.01	0.003	0.04
BDE-10	0.01	0.005	0.002	0.01
BDE-7	0.01	0.01	0.002	0.01
BDE-11	0.01	0.005	0.002	0.01
BDE-8	0.01	0.01	0.002	0.01
BDE-12/13	0.01	0.01	0.002	0.02
BDE-15	0.01	0.005	0.002	0.01
BDE-30	0.01	0.01	0.004	0.02
BDE-32	0.01	0.01	0.003	0.02
BDE-17/25	0.01	0.01	0.01	0.03
BDE-33/28	0.01	0.01	0.004	0.02
BDE-35	0.02	0.02	0.01	0.07
BDE-37	0.02	0.02	0.01	0.05
BDE-75	0.01	0.01	0.002	0.01
BDE-49	0.01	0.01	0.003	0.02
BDE-71	0.01	0.01	0.004	0.02
BDE-47	0.01	0.01	0.002	0.01
BDE-66	0.01	0.01	0.004	0.02
BDE-77	0.02	0.01	0.005	0.03
BDE-100	0.01	0.01	0.002	0.01
BDE-119	0.01	0.01	0.003	0.02
BDE-99	0.01	0.01	0.002	0.02
BDE-116	0.03	0.02	0.01	0.04
BDE-118	0.02	0.01	0.004	0.02
BDE-85	0.01	0.01	0.003	0.02
BDE-126	0.02	0.01	0.004	0.02
BDE-155	0.01	0.005	0.001	0.01
BDE-154	0.01	0.004	0.001	0.01
BDE-153	0.01	0.01	0.002	0.01
BDE-138	0.02	0.01	0.003	0.02
BDE-166	0.01	0.01	0.002	0.01
BDE-183	0.01	0.01	0.002	0.01
BDE-181	0.01	0.01	0.002	0.01
BDE-190	0.01	0.01	0.002	0.02
BDE-188	0.01	0.01	0.001	0.01
BDE-179	0.01	0.01	0.001	0.01
BDE-202	0.004	0.002	0.001	0.01
BDE-197	0.01	0.01	0.01	0.01
BDE-203	0.01	0.01	0.001	0.01
BDE-196	0.004	0.01	0.002	0.01
BDE-208	0.004	0.01	0.002	0.01
BDE-207	0.01	0.01	0.002	0.01
BDE-206	0.01	0.01	0.002	0.01
BDE-209	0.01	0.01	0.003	0.01

*Batch codes (A–E) for the samples correspond to those in Tables S6, S7, S9, and S10, respectively.

Table 2-3. Blank values for individual runs.

Batch code	**A**	**B**	**C**	**D**	**E**
	Liver (ng/g-lipid)	Adipose (ng/g-lipid)	Adipose (ng/g-lipid)	Plastic (ng/individual)	Plastic (ng/individual)
BDE-47	0.01	0.01	0.01	0.002	0.01
BDE-100	0.01	0.01	0.01	0.002	0.01
BDE-99	0.01	0.01	0.01	0.002	0.02
BDE-154	0.01	0.004	0.004	0.001	0.01
BDE-153	0.01	0.01	0.01	0.002	0.01
BDE-183	0.01	0.01	0.01	0.002	0.01
BDE-202	0.004	0.002	0.004	0.001	0.01
BDE-197	0.01	0.01	0.01	0.01	0.01
BDE-203	0.01	0.02	0.01	0.01	0.02
BDE-196	0.004	0.01	0.01	0.002	0.01
BDE-208	0.004	0.03	0.01	0.004	0.01
BDE-207	0.01	0.04	0.01	0.01	0.03
BDE-206	0.01	0.05	0.01	0.004	0.02
BDE-209	0.04	0.09	0.02	0.01	0.08

*Batch codes (A–E) for the samples correspond to those in Tables 2-5, 2-6, and 2-7, respectively.

Table 2-4. Number (pieces) and mass (g) of plastic found in short-tailed shearwaters.

	Total number	Total mass (g)		Total number	Total mass (g)
OS10-001	5	0.01	WK05-016	16	0.25
OS10-002	13	0.27	WK05-017	26	0.12
OS10-003	30	0.21	WK05-018	27	0.18
OS10-004	3	0.09	WK05-019	25	0.39
OS10-005	8	0.06	WK05-020	11	0.04
OS10-006	7	0.01	WK05-022	12	0.29
OS10-008	98	2.16	WK05-023	39	0.25
OS11-001	52	0.46	WK05-025	24	0.09
WK08-050	17	0.28	WK05-027	101	0.59
WK08-051	24	0.14	WK05-028	4	0.21
WK09-060	7	0.02	WK05-030	15	0.29
WK09-061	1	0.003	WK05-031	26	0.19
WK10-014	29	0.31			
WK10-016	44	0.71			
WK10-017	3	0.05			
WK10-018	25	0.38			
WK10-019	10	0.21			
WK10-023	29	0.21			
			Average	24.4	0.3

– : no item found.

Table 2-5. PBDE concentration in plastics in the stomach of short-tailed shearwaters (ng/individual).

		OS10-001	OS10-002	OS10-003	OS10-004	OS10-005	OS10-006	OS10-008	OS11-001	WK08-050	WK08-051	WK09-060	WK09-061	WK10-014	WK10-016	WK10-017	WK10-018	WK10-019	WK10-023
1Br	1																		
	2																		
	3																		
2Br	10																		
	7																		
	11																		
	8																		
	12/13							0.01											
	15							0.02											
3Br	30																		
	32							0.02											
	17/25							0.09	0.02										
	33/28							0.10											
	35																		
	37							0.04											
4Br	75																		
	49	0.01	0.01					8.06											
	71							0.33											
	47	0.22	0.06	0.02		0.004	0.01	0.98	0.01						0.15				
	66							1.57											
	77																		
5Br	100	0.06	0.02					5.03											
	119	0.11	0.01					4.72											
	99	0.04	0.01					2.86							0.05				
	116																		
	118							2.64											
	85							0.49											
	126							0.28											
6Br	155	0.05						1.31											
	154	0.48	0.03	0.003				15.3											
	153	2.69						9.15											
	138							3.71											
	166							0.29											
7Br	183	0.44	0.19	0.01				65.2	0.004										
	181							10.8											
	190							14.9											
	188							8.63											
	179																		
8Br	202		0.14			*0.001*		49.5							0.22		0.02		0.13
	197		0.21			*0.03*		1150							0.50		0.11		*0.01*
	203		0.74			*0.002*		331							1.77		0.27		0.11
	196		0.45			*0.005*		234							1.45		0.18		0.03
9Br	208		1.66		0.02	*0.01*		689							3.66		0.33		0.04
	207		2.34		*0.004*	*0.01*		1140							6.14		0.53		*0.02*
	206		3.26		*0.003*	*0.01*		1400							9.11		0.44		*0.01*
10Br	209	*0.02*	21.0	*0.004*	0.39	*0.03*		5080			*0.03*				23.2	*0.02*	3.73		0.45
total		4.12	30.2	0.03	0.41	0.10	0.01	10200	0.04		*0.03*				46.2	*0.02*	5.61		0.81
batch code		D	D	D	D	D	D	D	D	E	E	E	E	E	E	E	E	E	E

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

(continued)

		WK05-016	WK05-017	WK05-018	WK05-019	WK05-020	WK05-022	WK05-023	WK05-025	WK05-027	WK05-028	WK05-030	WK05-031
1Br	1												
	2												
	3												
2Br	10												
	7												
	11												
	8												
	12/13												
	15			0.01									
3Br	30												
	32												
	17/25												
	33/28												
	35												
	37												
4Br	75												
	49												
	71				0.01		0.01						
	47	0.01				0.01	0.07		0.01	0.01		0.01	
	66												
	77												
5Br	100						0.01						
	119												
	99						0.07						
	116												
	118												
	85												
	126												
6Br	155												
	154						0.01			0.09			
	153									0.08		0.01	
	138												
	166												
7Br	183									12.2		0.03	
	181												
	190												
	188												
	179												
8Br	202			21.8									
	197			7.5									
	203			11.7									
	196			53.5									
9Br	208			60.9			0.13					0.28	
	207			152			0.24					0.49	
	206			303								0.76	
10Br	209			2350			1.38					12.7	
total		0.01		2960	0.01	0.01	1.92		0.01	12.35		14.2	
batch code		D	D	D	D	D	D	D	D	D	D	D	D

Table 2-6. PBDE concentration in abdominal adipose of short-tailed shearwaters (ng/g-lipid weight).

		OS10-001	OS10-002	OS10-003	OS10-004	OS10-005	OS10-006	OS10-008	OS11-001	WK08-050	WK08-051	WK09-060	WK09-061	WK10-014	WK10-016	WK10-017	WK10-018	WK10-019	WK10-023
1Br	1																		
	2																		
	3																		
2Br	10																		
	7																		
	11				0.005														
	8																		
	12/13																		
	15				0.004														
3Br	30																		
	32																		
	17/25			0.02															
	33/28		0.01	0.02	0.04			0.01						0.38					
	35																		
	37																		
4Br	75				0.003														
	49	0.04	0.15	0.04	0.14	0.06	0.02	0.09	0.02						0.02			0.04	
	71																		
	47	0.65	1.75	0.23	0.72	0.74	0.73	0.28	0.06	0.16	0.11	0.08	0.19	0.17	0.26	0.15	0.14	0.29	0.05
	66		0.10	0.02	0.08	0.01												0.01	
	77		0.01		0.02														
5Br	100	0.27	0.74	0.11	0.21	0.44	0.34	0.19	0.02	0.06	0.10	0.09	0.16	0.88	0.16	0.21	0.13	0.23	0.02
	119	0.45	0.12		0.09	0.11		0.04	0.04		0.02	0.33	0.06		0.05	0.06	0.04	0.03	
	99	0.22	1.43	0.05	0.41	0.69	0.42	0.07		0.08	0.04	0.06	0.13	0.32	0.10	0.26	0.16	0.24	
	116																		
	118		0.15		0.05														
	85																		
	126		0.01		0.01	0.02													
6Br	155	0.24	0.33	0.06	0.08	0.14	0.18	0.08	0.04		0.01				0.02	0.05	0.05	0.04	
	154	2.22	1.27	0.19	0.57	0.71	0.93	0.34	0.13	0.23	0.14	0.33	0.78	1.22	0.41	1.17	0.47	0.93	0.05
	153	12.7	0.50	0.34	0.27	0.48	0.73	0.18	0.55		0.05	0.88	2.50		0.16	0.49	0.15	0.66	0.11
	138		0.01				0.04												
	166																		
7Br	183	2.77	0.04	0.07	0.06	0.03	0.06	0.09	0.03			0.34	1.97	0.19	0.01	0.03	0.02	0.45	0.04
	181																		
	190																		
	188																		
	179																		
8Br	202							0.02											
	197																		
	203							0.03											
	196							0.04											
9Br	208		*0.02*	*0.03*		*0.02*	*0.06*	*0.02*	*0.04*	*0.02*	*0.003*		0.05		*0.04*	*0.04*		*0.08*	0.04
	207		*0.01*	*0.01*	*0.005*	*0.02*	*0.11*	0.06	*0.05*	*0.02*		*0.03*	0.24		*0.05*	*0.04*	*0.01*	0.79	0.12
	206	*0.01*	*0.04*	*0.04*		*0.06*	*0.06*	0.03	*0.14*				*0.07*		*0.08*	*0.13*		*0.11*	0.02
10Br	209	*0.07*	*0.01*			*0.03*		0.07		*0.05*	*0.02*	*0.06*	*0.02*		*0.16*	*0.04*		0.53	0.32
total		19.7	6.69	1.23	2.77	3.56	3.69	1.63	1.12	0.64	0.50	2.19	6.16	3.16	1.52	2.67	1.17	4.45	0.78
batch code		B	B	B	B	B	B	C	B	C	C	C	C	C	B	B	C	B	C

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

(continued)

		WK05-016	WK05-017	WK05-018	WK05-019	WK05-020	WK05-022	WK05-023	WK05-025	WK05-027	WK05-028	WK05-030	WK05-031
1Br	1												
	2												
	3											0.10	
2Br	10												
	7												0.03
	11											0.01	
	8											0.004	
	12/13												
	15												
3Br	30												
	32			0.02									
	17/25												
	33/28									0.02		0.01	
	35												
	37												
4Br	75						0.005						0.01
	49		0.01		0.02	0.04	0.01	0.02	0.01	0.03	0.004	0.04	0.03
	71												
	47	0.07	0.36	0.04	0.30	0.32	0.14	0.07	0.13	0.49	0.15	0.49	0.83
	66		0.01		0.01					0.02		0.01	
	77										0.01		
5Br	100	0.03	0.13	0.02	0.06	0.11	0.09	0.02	0.03	0.41	0.05	0.53	0.49
	119	0.03	0.02		0.02	0.07	0.10	0.01	0.01	0.29	0.02	0.15	0.21
	99	0.02	0.13	0.02	0.14	0.17	0.15	0.02	0.09	0.56	0.13	1.34	1.04
	116												
	118		0.01				0.01	0.01		0.03		0.11	0.08
	85											0.01	
	126												
6Br	155		0.04			0.01	0.01	0.005	0.02	0.12		0.14	0.32
	154	0.14	0.22	0.05	0.13	0.25	0.55	0.08	0.13	1.71	0.10	1.38	2.16
	153	0.07	0.09	0.04	0.04	0.18	0.53	0.04	0.11	1.59	0.24	1.00	1.62
	138			0.01			0.31						
	166						0.01						
7Br	183	0.02		0.11		0.01	2.11	0.01		5.88	0.01	0.28	0.35
	181												
	190									0.02			
	188												
	179												
8Br	202			0.08			2.44			0.33			
	197			0.43			8.64			1.24		0.10	*0.06*
	203		*0.57*	0.29		*0.34*	7.60			0.38		0.13	0.19
	196			0.53			9.98			0.62			
9Br	208			0.26			12.7						
	207			1.17			28.6			0.35			
	206			0.31			6.58			0.08			
10Br	209			2.54			106			0.23			
total		0.38	1.59	5.94	0.73	1.50	186	0.28	0.53	14.38	0.72	5.83	7.44
batch code		C	C	C	C	C	C	C	C	C	C	C	C

Table 2-7. PBDE concentration in liver of short-tailed shearwaters (ng/g-lipid weight).

		OS10-001	OS10-002	OS10-003	OS10-004	OS10-005	OS10-006	OS10-008	OS11-001	WK08-050	WK08-051	WK09-060	WK09-061	WK10-014	WK10-016	WK10-017	WK10-018	WK10-019	WK10-023
1Br	1																		
	2																		
	3																		
2Br	10																		
	7																		
	11																		
	8																		
	12/13																		
	15																		
3Br	30																		
	32																		
	17/25																		
	33/28			0.04															
	35																		
	37																		
4Br	75																		
	49			0.03	0.03				0.02									0.08	
	71																		
	47	0.12	0.49	0.05	0.27	0.04	0.03	0.19	0.01	0.05		0.02	0.03	0.23	0.13	0.08	0.03	0.08	0.04
	66																		
	77																		
5Br	100		0.05		0.07			0.04		0.02	0.03			0.21	0.03	0.15	0.02	0.05	0.02
	119							0.07						0.05		0.06		0.04	0.03
	99	0.05	0.31	0.04	0.17	0.07	0.05					0.06	0.05	0.17	0.04	0.11		0.13	0.04
	116																		
	118							0.09											
	85																		
	126																		
6Br	155							0.12	0.02							0.03			0.01
	154	0.21	0.15	0.13	0.12	0.04	0.09	0.74	0.02	0.02	0.02	0.12	0.05	0.75	0.15	0.42	0.07	0.54	0.08
	153	1.37	0.06	0.16	0.04	0.06	0.05	0.14	0.07	0.03		0.43	0.42	0.04	0.03	0.05	0.07	0.47	0.20
	138																		
	166																		
7Br	183							0.51				0.05						0.19	0.03
	181							0.07											
	190							0.06											
	188							0.66											0.09
	179																		
8Br	202							1.02											0.04
	197							1.63											0.05
	203							1.91											0.18
	196							2.50											0.11
9Br	208							5.38		*0.03*					0.06			*0.02*	0.04
	207		*0.12*		*0.02*			11.7		*0.06*					0.17			0.54	0.53
	206							6.95											*0.05*
10Br	209		*0.17*		*0.03*	*0.04*		32.1		*0.12*	*0.02*				*0.26*	*0.02*	*0.04*	0.53	1.40
total		1.74	1.36	0.45	0.73	0.25	0.23	65.9	0.11	0.32	0.07	0.69	0.56	1.46	0.87	0.92	0.23	2.68	2.94
batch code		A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 2-8. PBDE concentration in prey species (ng/g-lipid weight).

		east NNPO*			west NNPO*			
		lanternfish			lanternfish			squid
congener		No.1	No.2	No.3	No.1	No.2	No.3	
1Br	1							
	2							
	3						0.01	
2Br	10							
	7					0.01	0.005	
	11					0.01		
	8						0.01	
	12/13				0.00	0.02		
	15							
3Br	30							
	32			0.002				
	17/25				0.01	0.01	0.01	
	33/28	0.01	0.001	0.003	0.04	0.02	0.04	
	35							
	37							
4Br	75							
	49	0.08	0.02	0.14	0.20	0.01	0.20	
	71							
	47	0.32	0.04	0.34	0.27	0.49	0.32	1.55
	66				0.03	0.14	0.08	
	77			0.002				
5Br	100	0.03	0.003	0.12	0.05	0.14	0.09	
	119			0.01	0.01	0.05	0.02	
	99	0.03	0.001	0.07	0.02	0.10	0.07	
	116							
	118			0.003	0.004	0.02		
	85							
	126			0.002	0.004			
6Br	155	0.02	0.004	0.09	0.05	0.21	0.10	
	154	0.05	0.01	0.19	0.11	0.47	0.20	1.38
	153	0.01		0.03	0.02	0.05	0.04	
	138							
	166							
7Br	183		0.002		0.004			
	181							
	190							
	188							
	179							
8Br	202							
	197							
	203							
	196							
9Br	208							
	207							
	206							
10Br	209							
total		0.5	0.08	1.01	0.80	1.73	1.20	2.93

Blank cells mean "not detected" (no peak was detected on chromatogram).

*NNPO : Northern North Pacific Ocean.

Chapter 3.

Leaching experiment

3.1. Introduction

Chapter 2 did not propose a mechanism of transfer. So here the author conducted a leaching experiment using stomach oil. Because additives are compounded in a polymer matrix and PBDEs are highly hydrophobic, PBDEs are supposed to be difficult to leach out. However, the stomach oil of seabirds may act as an organic solvent and accelerate leaching. The stomachs of members of the order Procellariiformes, including short-tailed shearwaters, hold oil derived from their diet, mainly fish. Stomach oils are composed mainly of wax esters or triacylglycerol (>70% of total lipids) [50,51] and could therefore facilitate the leaching of PBDEs from ingested plastics.

Stomach oils are found in chicks and adults (male and female), breeders and non-breeders [60]. The main function of stomach oils has been thought as an energy and water reserve [61]. Stomach oils contain from 5 to 35 times the energy content of the prey, and help adults to trip over patchy food sources and also help the chicks to go for days without meals [61]. Roby et al. (1989) proposed that metabolizing stomach oils during fast periods was more energetically efficient than metabolizing adipose tissue because to synthesize and mobilize the adipose tissue consumed more energy [62]. Stomach oils are slowly digested and efficiently absorbed by seabirds. This means that the plastics in stomach have many chances to contact with the oil, and the extracted chemicals can be efficiently absorbed. The objective of this study was to understand how the PBDEs are transferred from the plastics to the tissues, with a focus on the facilitation of leaching by stomach oils. To investigate the leaching of PBDEs, leaching experiments were conducted using pieces of plastic compounded with deca-BDE.

3.2. Materials and methods

Leaching experiments were conducted using pieces of plastic compounded with deca-BDE and five types of leaching solution. High-density polyethylene plate (2 mm × 25 mm × 39.5 mm) containing deca-BDE was purchased from Dokuritsu Tensyoku Kankyo Hozen Kenkyujo (Tokyo, Japan). The plastic was cut into pieces 2 mm × 3 mm × 3 mm, similar to the size of the plastics found in the stomach of short-tailed shearwaters [38]. To determine the PBDE concentration in the plastic, the plastic plate were dissolved in toluene, the solution was purified by 5% H_2O-deactivated silica gel column chromatography and then by fully activated silica gel column chromatography, and the PBDEs in the toluene solution were measured by a gas chromatograph – electron capture detector (GC-ECD). The average concentration of PBDEs in the plate (Table 3-1) was used to calculate the percent leaching. Duplicate analyses measured BDE209 in the plastic plate at 697 μg/g (Table 3-1).

Leaching experiments were performed with distilled water, seawater, acidic pepsin aqueous solution, fish oil (a major component of stomach oil), and actual stomach oil taken from wild seabirds. Seawater was collected in Tokyo Bay in June 2013 and autoclaved before experiments. Acidic pepsin aqueous solution was prepared from commercial pepsin (Sigma-Aldrich Co. U.S.A.) at 10 g/L, and the pH was adjusted to 3 with HCl to simulate a seabird's gastric juice [63,64]. Fish oil, made from walleye pollack (*Theragra chalcogramma*), was obtained from Hokkaido Fine Chemical (Hokkaido, Japan). Stomach oil was taken from adult streaked shearwaters (*Calonectris leucomelas*) in the breeding season (July) on Awa Island (Niigata, Japan) by a non-lethal technique [65]: After the injection of water into the proventriculus via the mouth through an elastic tube, the stomach contents were flushed into a bucket by gently pushing the belly of the birds. The pH of the leaching solutions was 3.0–3.8 in the pepsin solution, 8.5–9.0 in seawater, and 7.2–10.6 in distilled water. The fish oil and stomach oil were analyzed in advance of the experiments to confirm no detectable concentrations of BDE209.

Both the fish oil and the stomach oil are composed mainly of triacylglycerol and are chemically similar. The leaching treatments are listed in Table 3-2. In each experiment, one piece of plastic (0.015–0.023 g) and leaching solution (15–23 mL) were placed in an amber glass centrifuge tube (50 mL) at a liquid-to-solid ratio of 1000 mL : 1 g. Experiments using stomach oil were performed in a quarter of the size of the other experiments (2 mm × 1.5 mm × 1.5 mm and 5 mL) because only a limited amount of stomach oil was available. During the experiments, the centrifuge tube was continuously shaken (200 rpm) at 20 °C to simulate the temperature of seawater and cold-blooded animals or at 38 °C to simulate the temperature of a seabird's stomach [66]. All experiments were conducted in duplicate.

The amount of PBDEs leached out was calculated from the concentrations in the leaching solution at 5 and 15 days (and at 0.5 and 2 days in fish oil). The water-based solutions were liquid–liquid extracted with the same volume of dichloromethane (DCM) in the centrifuge tube after the plastic was removed. After spiking with surrogate standard (4′-fluoro-2,2′,3,3′,4,5,5′,6,6′-nonabromo-diphenylether) and shaking (1 min), the organic phase was taken and dehydrated with anhydrous sodium sulfate. The oil-based leaching solutions were spiked with the surrogate standard, diluted with DCM, and subjected to gel permeation chromatography to remove the oils and obtain the DCM eluent containing PBDEs.

The extracts or DCM eluents were purified by 5% H_2O-deactivated silica gel column chromatography and then by fully activated silica gel column chromatography as described in Chapter 2. Purified samples were concentrated and analyzed by GC with electron capture detector (GC-ECD) for 10 BDE congeners (hepta- to deca-brominated; BDE179, 188, 196, 197, 202, 203, 206, 207, 208, 209).

3.3. Results and discussions

3.3.1. Leaching of deca-BDE from plastic

The water-based solutions (distilled water, seawater, and pepsin solution) leached <1% of the PBDEs in the plastic, except for 1.09% by pepsin solution on day 15 (Fig. 3-1). On the other hand, fish oil leached more than 50 times as much: around 45% by day 2 and thereafter. The stomach oil leached 12.6%–14.5% of the PBDEs in the plastic by day 5 and 12.6%–15.6% by day 15 (Fig. 3-1), more than 20 times the proportion by water-based leaching solution. These results indicate that the contact of oil with plastics in a bird's stomach greatly facilitates the leaching of hydrophobic chemicals.

Minimal leaching (<1%) by the water-based solutions is consistent with the highly hydrophobic nature of PBDEs (Fig. 3-1). Water is not efficient at extracting hydrophobic additives from plastic matrix. On the other hand, the hydrophobic nature of the fish oil greatly facilitated leaching. However, if fish oil were as highly hydrophobic as non-polar organic solvent (e.g., *n*-hexane, octanol), it would leach 99.9% (i.e., 1 – 1/1000) at an equilibrium based on the liquid-to-solid (plastic) ratio of 1000:1 in the experiment. Instead, it reached an equilibrium of ~40% (Fig. 3-1), because fish oil is not as hydrophobic as non-polar organic solvents. The fish oil consists of a neutral lipid fraction (90%–95% by weight) which is dominated by triacylglycerol and polar lipids (5%–10%) such as phospholipids [67,68], which, although hydrophobic, have a considerable polar moiety, which may decrease the hydrophobicity of the fish oil. Similarly, stomach oil enhanced the leaching of PBDEs (Fig. 3-1), reaching an equilibrium of ~15%. This lower proportion is probably due to the presence of water and polar organic matter derived from prey in the stomach oil, which lowered the partitioning of hydrophobic components into the stomach oil.

The obtained results are consistent with the finding that dissolved humic matter and methanol can each facilitate the leaching of PBDEs from plastics [69,70]. Sodium taurocholate, a bile salt, increased the desorption of adsorbed persistent organic pollutants from microplastics, especially under conditions simulating warm-blooded organisms: up to 30 times that in seawater [71]. In the present study, however, pepsin solution only slightly enhanced leaching compared with seawater, probably because pepsin is not hydrophobic enough to extract hydrophobic additives mixed within plastic matrix instead of adsorbed to the surface.

In the leaching experiment, plastic plate with BDE209 at 697 μg/g was utilized, two orders of magnitude higher than observed in plastics floating on the open ocean [14]. If leaching were concentration-dependent, PBDEs would be leached more slowly than the measurement of this study. However, few surveys of PBDEs in plastics in the open ocean have been conducted, and seabirds may encounter plastic debris with similar concentrations of PBDEs to those used in the leaching experiment. The highest level of BDE209 found in plastic was 5080 ng in the bird OS10-008, which held 0.452 g of plastic fragments in the gizzard. Therefore, the "average" concentration of BDE209 in the ingested plastics was 11 μg/g. However, the additives are not present uniformly in all plastics but are compounded into only some plastics, and therefore this "average" value would be an underestimate. For example, if BDE209 was present only in the largest piece of plastic fragment in OS10-008 (0.0487 g), its concentration would be 103 μg/g, of the same magnitude as in the leaching experiment. A more extensive survey of PBDEs in marine plastic fragments is necessary. In addition, the concentration dependence of the leaching of plastic additives should be examined in future studies.

In members of the order Procellariiformes, stomach oils are generated from prey items in the proventriculus, where easily digestible organic matter such as proteins and carbohydrates are rapidly broken down, and the remaining oily components are separated from the aqueous fraction [50,62,72]. The aqueous (lower) phase is emptied first [62,72]. Then the stomach oil is slowly emptied into the gizzard and thence into the intestine, where it is gradually digested by pancreatic lipases and bile. Plastic fragments become trapped mainly in the gizzard [41], where they come into contact with the stomach oil. This contact could be facilitated by intestinal reflux, which moves the contents of the intestine back into the stomach. Birds that eat oily prey have a high rate of intestinal reflux, and repeatedly shuttle the lipid contents between stomach and intestines to improve digestion [73]. These processes would therefore permit intermittent contact between stomach oil and plastics in the gizzard. This intermittent contact may explain the inconsistencies in the occurrence of higher-brominated congeners between plastics in gizzard and tissues, which observed in field sample analysis in Chapter 2.

3.3.2. Model calculation of plastic-derived exposure of PBDEs relative to dietary exposure

By using PBDE concentrations in plastics, their amounts in the digestive tracts and rate of leaching in the experiment, plastic-derived exposure of PBDEs were calculated for comparison with exposure through natural foods as below: OS10-008 is used as a model individual. The plastics in OS10-008 contained 5080 ng of BDE209. The rate of leaching of BDE209 by stomach oil in the leaching experiment was 15%. Therefore, 15% of 5080 ng-BDE209 (i.e., 762 ng) would be leached into the digestive fluid within the bird during 15 days, the duration of the leaching experiment. Similarly, 0.15 ng of BDE47 would be leached. From the energy requirement (929 kJ/day), assimilation efficiency (0.75), and prey composition [74], the consumption of 364 g/day of prey was calculated. Assuming that all preys contain the same concentrations of BDEs as those of lantern fish (BDE209, 0.002 ng/g; BDE47, 0.03 ng/g) (Table 2-8, Fig. 2-4) an exposure to 11 ng of BDE209 and 164 ng of BDE47 was calculated through prey over 15 days. The resultant dominance of plastic-mediated exposure of BDE209 (762 ng) over natural prey (11 ng) is consistent with the result of fingerprinting of the PBDE profiles. In contrast, the case of BDE47, the model calculates the dominance of exposure through prey (164 ng) over ingested plastic (0.15 ng). The model assumes that PBDE exposure is saturated at 15% by 15 days. After this time, only dietary exposure of PBDEs would continue. However, even after a couple of years, plastic-mediated exposure (762 ng) would dominate cumulative dietary exposure (0.002 ng/g × 364 g/day × 365 days × 2 = 531 ng). Meanwhile, the bird may continue to ingest plastics with additive deca-BDEs. Although no data are available on the exact frequency of plastic ingestion by birds and the occurrence of plastic fragments containing deca-BDE in the open ocean, this scenario is likely because of the consistency with environmental observations (i.e., similarity of PBDE profiles). These comparisons highlight the important contribution of additives in marine plastic debris to chemical exposure to marine organisms.

3.4. Conclusion

The obtained data in the present study indicate that, in members of the order Procellariiformes, stomach oil acts as an organic solvent, facilitating the leaching of hydrophobic chemicals from ingested plastics. Up to now, hydrophobic chemicals in plastics were expected that they are not likely to leach out in marine environment or in digestive tract of marine organisms. It is concluded that in case of seabirds, especially in the order Procellariiformes, the leaching of chemicals from plastics can be greatly facilitated in their stomach by stomach oil, and the exposure to additive chemicals may be far larger than dietary exposure.

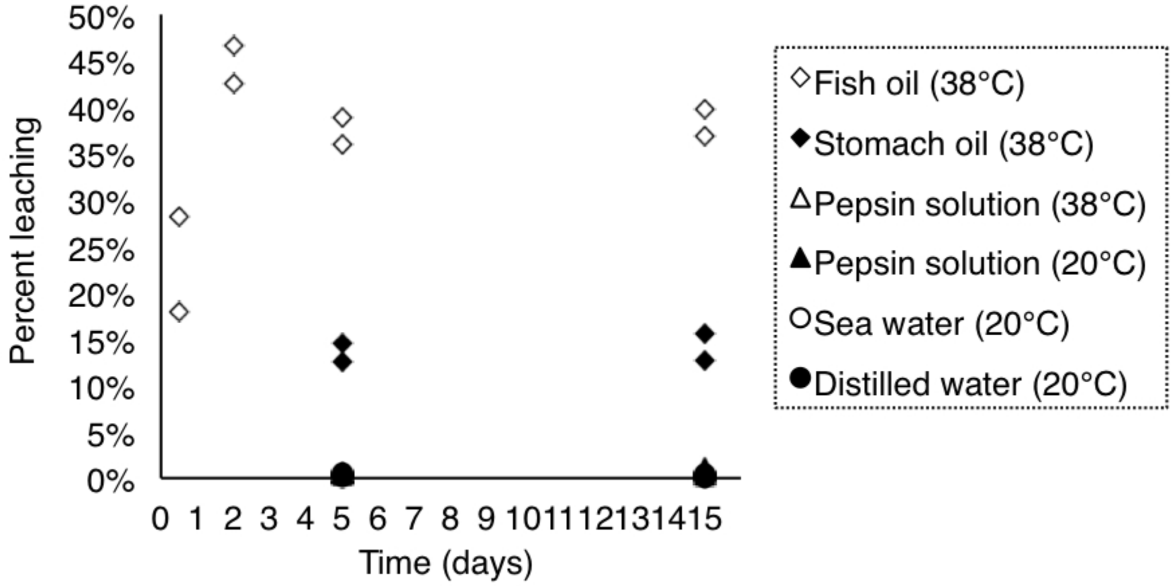


Figure 3-1. Proportions of total PBDEs in the leaching solutions relative to total PBDEs originally present in plastic plate.

Table 3-1. PBDE concentrations (μg/g) in plastic plate used in the leaching experiment.

	No. 1	No. 2	Average
BDE-188	0.1	0.1	0.1
BDE-179	0.1	0.2	0.1
BDE-202	0.4	0.4	0.4
BDE-197	1	1	1
BDE-203	4	3	3
BDE-196	3	2	2
BDE-208	15	16	16
BDE-207	25	28	26
BDE-206	17	20	19
BDE-209	648	746	697
Total	713	817	765

Table 3-2. Treatments used in leaching experiment.

	Period (days)	Temperature (°C)	Details
Distilled water	5, 15	20	
Sea water	5, 15	20	Tokyo Bay
Pepsin solution	5, 15	20	Pepsin 1%, pH 3, HCl
Pepsin solution	5, 15	38	Pepsin 1%, pH 3, HCl
Fish oil	0.5, 2, 5, 15	38	Walleye pollack (*Theragra chalcogramma*)
Stomach oil	5, 15	38	Streaked shearwater (*Calonectris leucomelas*)

Chapter 4.

Measurement of plastic-derived contaminants in seabirds of various species from different areas

4.1. Introduction

4.1.1. Objectives

In chapter 2, PBDEs in short-tailed shearwater were analyzed and higher-brominated congeners were sporadically detected in both of ingested plastics and tissue. It is indicated that additive PBDEs retained in ingested plastics were reached out from plastics, and were transferred and accumulated into seabirds' tissue. The reaching process can be greatly facilitated by stomach oil, which is present in the digestive tract of birds in the order Procellariiformes (Chapter 3). Therefore, Procellariiformes seems to be at high risk of exposure to additive PBDEs retained in marine plastics. There are many species in the order Procellariiformes, and they are distributed all around the ocean. The objective of the study in this chapter is to investigate the accumulation of plastic derived chemicals across seabird species and the oceans, with a focus on Procellariiformes.

4.1.2 Seabird samples

The author analyzed PBDEs in tissue and ingested plastics of field caught samples. Three species of Procellariiformes from different oceans were sampled: northern fulmar (*Fulmarus glacialis*) from North Atlantic Ocean, and white-chinned petrel (*Procellaria aequinoctialis*) and shy albatross (*Thalassarche cauta*) from South African waters.

Northern fulmar (*Fulmarus glacialis*)

Northern fulmar has a circumpolar range in the northern hemisphere occurring in the pack ice and in waters further south in coastal, offshore, and pelagic zones [52]. They mostly feed by surface-seizing while swimming buoyantly. The main components of the diet are fish, crustacea, cephalopods, and carrion [52]. Plastic ingestion of northern fulmars in North Sea has been monitored for long term from early 1980s [75].

White-chinned Petrel (*Procellaria aequinoctialis*)

White-chinned Petrel is pelagic and widely distributes in the subantarctic zone. They travel a long distance up to several thousand kilometers when breeding. Their major foods are fish, cephalopods, and krill, and they catch the preys mainly by surface-seizing or surface-diving [52].

Shy albatross (*Thalassarche cauta*)

Shy albatross breeds on several islands around Tasmania, and post-breeding adults tend to remain over the continental shelf and slope of south-east Australia. However, young birds are more mobile and are known to leach South Africa [52]. They mostly feed on fish by surface-seizing or diving in the shallow water.

4.1.3. Target compounds

Although PBDEs were the main focus in this study, polychlorinated biphenyls (PCBs) in adult and chick northern fulmar was also analyzed. PCBs are adsorbed on plastics in marine environment. In previous study, the correlation between PCBs concentration in tissue and the mass of plastics in stomach were investigated, however, only weak correlation have been observed [38]. The author focused on chick samples. Because plastics generally retain in the stomach of seabirds for more than few months [53], the chick samples, of which were approximately 2-month-old, were expected to hold most of ingested plastics without regurgitating. Therefore, the mass of plastics in stomach can be assumed to be more precisely correlated with the exposed amount of PCBs from plastics than adults.

Polychlorinated biphenyls (PCBs)

PCBs were widely manufactured throughout 40 years, from 1930 to the 1970s, being used as stable fluid insulators in high-voltage electric transformers, in high-capacity condensers, as heat exchangers, pesticide extenders, adhesives, dedusting agents, components of cutting oils, flame retardants, hydraulic lubricants, and components of plasticizers in paints, inks, toners, and printing inks. By the late 1970s most governments banned PCB production but PCBs still persist and are ubiquitous in the environment [76]. Moreover, because PCBs have high bioaccumulation capacity and are persistent, they are biomagnified in the food web and found to be highly concentrated in top predators, such as seabirds.

4.2. Materials and Methods

4.2.1. Sample collection and treatment

Adult northern fulmars (*n* = 20) killed accidentally in long-line fisheries have been used. They were sampled at the ocean around Faroe Islands (61°36'- 62°52'N, 06°25'-08°00'W) during June to August 2011 to 2012, and three birds, i.e., FAE-2012-024, FAE-2011-687, and FAE-2011-688 were collected on January 2012, November 2011, and May 2011, respectively. Chick fulmar (n = 18) were hunted for human consumption on the ocean at north of Vágoy, which is an island placed in west of Faroe Islands, in September 2013. The samples of white-chinned petrels (*n* = 23) and shy albatrosses (*n* = 5) were killed accidentally on long-lines in South African waters (29°33′–37°40′N, 21°49′E–32°29′W) during June to October 2012 to 2013.

The carcasses were stored at –30 °C until dissection. In the laboratory, the carcasses were dissected with a solvent-rinsed stainless steel knife. Sets of abdominal adipose, liver, and plastics were collected from adult northern fulmars. Abdominal adipose, plastics, and stomach oil were collected from chicks of northern fulmar. Abdominal adipose and plastics were collected from white-chinned petrels and shy albatrosses.

The tissues were removed, put in solvent-rinsed glass vials, and stored in the freezer at –30 °C until analysis. Plastic pieces found in the stomach were washed in distilled water, dried at room temperature, and weighed on an electronic balance (AB54, Shimadzu, Kyoto, Japan) with precision of 0.1 mg. The plastics were identified and sorted according to polymer type by near-infrared spectrometry (PlaScan-W, OPT Research Inc., Tokyo, Japan). Identification was done by comparison of infrared spectra with those in a library (e.g. [55]). After sorting, they were ultrasonicated in distilled water and dried at room temperature.

4.2.2. Extraction and clean up of tissues and plastic samples

The method for extraction and clean up processes were fully the same as those for short-tailed shearwater, which detailed in Chapter 2. PBDEs and PCBs in abdominal adipose tissue, liver, and ingested plastics were analyzed for adult northern fulmars, and those in abdominal adipose tissue, ingested plastics, and stomach oils in the proventriculus were analyzed for chicks of northern fulmar. The African birds, white-chinned petrels and shy albatrosses were analyzed for PBDEs in abdominal adipose tissue and ingested plastics.

4.2.3. Instrumental analyses

The analytical settings and methods for instrumental analyses were the same as those for short-tailed shearwater, which detailed in Chapter 2, but four PBDE congeners, BDE-188, 179, 202, 197 were excluded. These 4 congeners could not be identified clearly in many of biological samples, because of disturbance of ECD chromatogram by some unidentified peaks. In the analysis of northern fulmars, 39 PCB congeners (CB-8, 18, 28, 52, 49, 44, 74, 66, 101, 99, 87, 110, 118, 105, 151, 149, 146, 153, 138, 158, 128, 167, 156, 157, 178, 187, 183, 177, 172, 180, 170, 189, 199, 196, 206, 195, 194, 206, and 209) were identified. Compounds were identified and quantified against native standards.

4.3. Results and Discussions

4.3.1. Northern Fulmars of North Atlantic Ocean

4.3.1.1. PBDEs and PCBs in ingested plastics and tissues

Ten of 20 adult fulmars and all of 18 chicks held plastics in their stomachs, at n.d.–0.18 g per bird in adults (Table 4-1) and 0.02–0.62 g per bird in chicks (Table 4-2, Fig.4-1). The numerical compositions of ingested plastics in adults and chicks were as follows: fragments, 68% and 77%; plastic sheets, 5% and 6%; resin pellets, 1% and 6%; and fibers, 23% and 5%, respectively. In the adult birds, the author found 3.9 pieces and 0.03 g per bird on average, and were much less than previous investigation of stomach contents of fulmars from the Faroe Islands, which has reported 88% of incidence, 14.2 pieces and 0.17 g per bird on average [75]. This difference attributes to that most of the adult samples in this study was successful breeders, which are known to regurgitate plastics in the stomach during feeding to their chick with food [75]. According to the methods described in [77], 18 of 20 adult birds were identified as those which had just finished breeding period (Table 4-1).

Total PBDEs in the adipose of the adults ranged from 12.9 to 386 ng/g-lipid weight with median of 40.6 ng/g-lipid (Table 4-3, Fig. 4-2A) and those in liver of adults ranged from 6.59 to 175 ng/g-lipid weight with median of 20.3 ng/g-lipid weight (Table 4-4, Fig. 4-2B). Total PCBs in the adipose of the adults ranged from 1890 to 77400 ng/g-lipid weight with median of 20700 ng/g-lipid (Table 4-5) and those in liver of the adults ranged from 3400 to 42800 ng/g-lipid weight with median of 11800 ng/g-lipid weight (Table 4-6).

Total PBDEs in the adipose of the chicks ranged from 1.46 to 26.3 ng/g-lipid weight with median of 2.44 ng/g-lipid weight (Table 4-7, Fig. 4-3A). Total PBDEs in stomach oil ranged from 2.86 to 9.49 ng/g-lipid weight with median of 4.13 ng/g-lipid (Table 4-8, Fig. 4-3C). Total PCBs in the adipose of the chicks ranged from 112 to 1860 ng/g-lipid weight with median of 254 ng/g-lipid weight (Table 4-9). Total PCBs in stomach oil ranged from 78.3 to 394 ng/g-lipid weight with median of 122 ng/g-lipid (Table 4-10). In one chick (FAE2013-003), apparently higher concentration of both PBDEs and PCBs were detected. The reasons for this accumulation in FAE2013-003 are discussed later.

PBDEs and PCBs in fulmar of Faroe Islands have been analyzed in a previous study [57]. Comparing geometric mean of Σ (BDE-47, -99, -100, -153, and -154) and Σ (PCB-105, -118, -128, -167, -138, -153, -156, -170, -180, and -183) in adipose of chicks, 19 ng/g-lipid and 880 ng/g-lipid, respectively, were reported in the previous study [57], which were higher than the observation in the present study (3.6 ng/g-lipid and 276 ng/g-lipid, respectively). This difference may be caused by the difference of sampling year, i.e., the birds in Fängström (2005) were collected during 2000-2001 [57], and the samples in this study were collected during 2011-2013. PCBs and PBDEs have been phased out before the year 2000 in EU, therefore, those in fulmars may reflect the decreasing trends of environmental medias. The similar decreasing trends were also found in herring gull of European waters [78].

In many adult and chick fulmars, PBDEs profiles were dominated by lower-brominated congeners (tetra- to hexa-brominated) (Tables 4-3, 4-4 and 4-7, Figs 4-2A,B and 4-3A). Stomach oil showed similar PBDEs profiles, which were made from lipids in prey fish. The tetra- to hexa-brominated congeners in birds' tissue were indicated to be concentrated from their food. On the other hand, in the both liver and abdominal adipose of three adults, i.e., FAE-2012-015, FAE-2012-019 and FAE-2012-027, high concentration of higher-brominated congeners (BDE209 and nona-brominated congeners) were detected and they were dominant in the profile (Tables 4-3 and 4-4, Fig. 4-2A,B). This accumulation pattern of PBDEs, i.e., sporadic accumulation of higher-brominated congeners, corresponded to that observed in short-tailed shearwater. Exposure to PBDEs derived from plastics with deca-BDE in their stomachs is indicated. However, in the stomach of these three birds, no plastic was found or few plastics with no PBDEs were detected (Table 4-11, Fig. 4-2C). A likely explanation is that they had ingested plastics with deca-BDE and accumulated BDE209 in their tissue, before excreting the plastics.

In some of the chicks, low concentration of BDE209 was detected in the adipose (Table 4-7, Fig. 4-3A), but not in ingested plastics (e.g. FAE-2013-018) (Table 4-12, Fig. 4-3B). This inconsistency is hard to be explained by excretion of plastics, because the chicks were approximately 2-month-old and seabirds are generally retain plastics in the stomach for more than few months [53]. Chicks, which accumulated plastics fed by their parents, are likely to hold most of ingested plastics. One likely explanation is that distribution of BDE209 in maternal tissue to egg. In fact, Karlsson et al (2006) reported sporadic detection of relatively high levels of BDE209 among egg samples of northern fulmar from Faroe islands [79].

Plastics in the stomach of one chick, FAE-2013-008, contained more than three orders of total PBDEs than the other birds (Table 4-12, Fig. 4-3B). The congeners in the plastics were dominated by BDE47 and BDE99. This congener profile is similar to that of penta-BDE technical product containing BDE47 and BDE99 as a major component [56]. These results indicate that FAE-2013-008 ingested plastic in which penta-BDE was industrially compounded. However, the concentrations of BDE47 and BDE99 were not significantly higher in the adipose of FAE-2013-008 than the others (p-value > 0.05). This indicates that these congeners were not transferred and accumulated from plastics to the birds' tissue, because the plastic might not have been in the stomach long enough for PBDEs to be leached out and absorbed by birds.

The lowest total PCBs were observed in liver and adipose of FAE-2011-687, and different congener profile of PCBs than the other birds were detected, i.e., relatively higher proportion of CB138 and lower proportion of CB153 (Tables 4-5, and 4-6, Fig. 4-4A,B). In addition, congener profiles of PCBs in adipose and liver of FAE-2011-687 were placed closely and separated by the others on the dendrogram and Nonmetric multidimensional scaling (NMDS) ordination (Fig. 4-5). FAE-2011-687 was the only bird of which color phase was classified double dark (DD), according to van Franeker (2004)[77], however, all of the other birds were classified as double light (LL). van Franeker (2004) described as follows: colored individuals which include double dark (DD) are virtually limited to arctic populations (subspecies F.g.glacialis), and double light (LL) individuals make up near 100% of the southern subspecies populations (F.g.auduboni)[77]. The difference of congener profile between FAE-2011-687 and the others may be caused by the difference of regional pollution or food.

4.3.1.2. Correlation between PCBs in tissue and mass of plastics

To assess the accumulation of plastic derived PCBs, the author investigated the correlation between PCBs concentration in tissue and the mass of ingested plastics, using chicks (Fig. 4-6). This calculation was not conducted on adult samples because most of the adult samples raised chicks and regurgitated ingested plastics (Table 4-1). As a result, there are no positive correlations between the concentrations of each PCB homologues in tissue and the mass of ingested plastics. Comparing the exposure from plastics and that from natural diet, an estimation was made in Herzke et al. (2016), ingesting rate (mass) of plastics by northern fulmar was five orders smaller than that of prey [80]. Although it is still needed detailed study on PCBs concentration in floating plastics and in prey organisms, in the case of fulmar, PCBs exposure from prey is expected to overcome that from ingested plastics.

The bird with the lowest amount of plastics (FAE2013-003), which plot was colored blue in Fig. 4-6, accumulated the highest concentration of PCBs. FAE2013-003 also accumulated PBDEs at the highest concentration among chick samples (Fig. 4-7). The congener profiles of both PCBs and PBDEs in FAE2013-003 do not seem to be different from the other chicks (Figs 4-3A and 4-8A), and PBDEs in the tissue are dominated by tetra- to hexa brominated congeners. These features suggest that contaminants in FAE2013-003 were accumulated by bio-accumulation. Northern fulmar mainly preys on fish, crustacea, and cephalopods, and also feed on carrion (often mammal)[52]. Among the prey species, mammals are stated at the highest trophic levels, and generally accumulate much higher concentration of PCBs and PBDEs, for example, in a marine food chain of northern Norway, PCBs in seals were approximately 30 times higher than those in sandeels [81]. One explanation is that FAE2013-003 was fed more carrion of mammals and less lower-trophic organisms such as fish than the other chicks. Fewer plastic ingestion of FAE2013-003 may relate to lower frequency of hunting for small marine organisms.

In addition, all of the PCB homologues are decreasing trends, and some of the homologues, such as 4-chrorinated and 5-chrorinated PCBs, decrease significantly with the amount of plastics in both cases of including or excluding FAE2013-003 (Fig. 4-6). However, there were no significant correlation between the number of plastics and any homologues (Fig. 4-9). The number of plastics in stomach can be affected by fragmentation of plastics. The observed decreasing trend suggests that plastic ingestion is possibly related with the feeding habits, e.g., the bird which mainly feeds on lower trophic level species such as planktonic crustaceans may catch more plastics than those which feed on higher trophic organisms such as fish or mammals.

4.3.2. PBDEs in white-chinned petrel and shy albatross of South African waters

The author analyzed twenty-three white-chinned petrels with plastics, at 0.01–2.02 g per bird (Table 4-15, Fig.4-10), and one white-chinned petrel (#3482) and five shy albatrosses without plastic. In the plastics ingested by white-chinned petrels, fragments of end-products accounted for 40% of all pieces, followed by plastic sheets (32%), fibers (19%), and resin pellets (8%). Comparing this profile, white-chinned petrels ingest plastics with higher proportion of sheets and fibers and lower proportion of fragments than short-tailed shearwater or northern fulmars, in which fragments accounted for over 60% and sheets and fibers accounted for less than 20% and 10 %, respectively. Some of sheets in white-chinned petrels could be identified as packaging of snacks or polyethylene bag (Fig.4-10), and fibers are derived from lines or ropes for fishing. White-chinned petrels are known to frequently attend fishing vessels [52]. It is indicated that garbage from fishing vessels are relatively higher proportion of plastics ingested by white-chinned petrels in South African waters. In addition, one bird (#3499) ingested black crinkly sheet of plastics, which seems to be burned cinder (Fig.4-10). A susceptible source of burned plastics is debris from incinerator on ships [82]. The plastic in #3499 may be also derived from vessels.

Total PBDEs in the adipose ranged from 1.84 to 23.4 ng/g-lipid weight with median of 5.9 ng/g-lipid weight (Table 4-16, Fig. 4-11A). The concentration is similar to that of previous report on pelagic seabirds around South African waters [83]. The congener profiles were variable among the birds, but in most of the birds, the profiles were mostly composed of lower-brominated congeners (tetra- to hexa-brominated). On the other hand, in abdominal adipose of two birds, i.e., #3477 and #3484, higher-brominated congeners (octa- to deca-brominated congeners) were dominant in the profiles, and some other birds also accumulated low concentration of BDE209 and nona-brominated congeners, including shy albatross, i.e., # 3486 and #3487 accumulated 0.12 and 0.16 ng/g-lipid weight of BDE209, respectively.

Octa-brominated congeners, i.e., BDE196, BDE197, BDE202, and BDE203, were detected as major components in the profiles of higher-brominated congeners accumulated in two white-chinned petrels (#3477, #3484). The accumulation pattern of higher-brominated congeners in tissue was similar to that observed in short-tailed shearwater, and suggested the transfer of PBDEs from ingested plastic to the tissues. The profile dominated by octa-brominated congeners, may be unique to additive octa-BDE, because the profiles dominated by octa-brominated congeners is uncommon in marine environments, to the best of our knowledge. However, PBDEs were not detected in the plastics in their stomachs (Table 4-17, Fig. 4-11B). In the plastic samples, only low concentration (less than 1ng /individual) of PBDEs were detected in 6 samples, and higher-brominated congeners were detected in one sample, plastics from #3504 (Table 4-17, Fig. 4-11B).

BDE209 were detected in two shy albatross (# 3486 and #3487) (Table 4-16, Fig. 4-11A), however, a previous study reported that the frequency of plastic ingestion of shy albatross were less than 1% [84]. The observed detection frequency of BDE209 in 2 of 5 individuals seemed higher than expected. Low concentration of BDE209 was also detected in most of northern fulmars and white-chinned petrels, although it was not noticeable in congener profile. It is suggested that the low concentration of BDE209 in shy albatross may be derived from the other sources than plastics. In the environment, BDE209 is dominant in hydrophobic solids such as soils and sediments [85]. Some birds in terrestrial food webs accumulated BDE209 in tissue [86], which can be explained by daily exposure to contaminated soils. Pelagic seabirds do not utilize terrestrial food webs, however, they may be exposed to BDE209 through prey which ingest hydrophobic solids. Further studies are needed to distinguish the sources of low concentration of BDE209 accumulated in seabirds' tissue.

Deca-BDE was detected in northern fulmar of North Atlantic Ocean, and deca-BDE, octa-BDE and hexa-BDE were detected in short-tailed shearwaters in North Pacific Ocean. This may relate to the difference of the use of PBDEs between Europe and Asian countries. In Europe, PBDE technical mixtures apart from deca-BDE were phased out, however, some Asian developing countries may still use them. In white-chinned petrels of South African waters, lower concentration of PBDEs, in which was similar profile to octa-BDE, were observed. South African waters is assumed to be more remote from human activities than North Atlantic or North Pacific Ocean. PBDEs are not combined with polymer of plastics, therefore, they can be leached out slowly in the marine environment during floating for long periods. Thus, one possible explanation of the lower accumulation of PBDEs is that the plastics in South African waters retain lower concentration of additive PBDEs. To interpret the difference of the accumulation pattern, and to understand the risks of plastic ingestion to seabirds, it is needed to quantify the PBDEs burden in marine plastics in each of the waters.

4.4. Conclusion

The author detected sporadic accumulation of higher-brominated congeners of PBDEs in tissue of seabirds from North Atlantic Ocean and South African waters. The sporadic detection of PBDEs was corresponded to that observed in short-tailed shearwater. It was indicated to be derived from additives retained in ingested plastics. The obtained data indicate that chemical exposure to seabirds by plastic ingestion occurs all around the ocean.

Some of the northern fulmars and short-tailed shearwaters accumulated much higher concentration of plastic-derived PBDEs than that derived from bio-accumulation. Because plastics are applied many kinds of chemicals during the manufacturing process, marine plastics may retain them in the marine environment. Seabirds which ingested plastics can be exposed to the other additives as well as PBDEs. It is needed to quantify the PBDEs and the other chemicals contained in marine plastics to assess the toxic risks of plastic ingestion.

Figure 4-1. Examples of plastics in the stomach of northern fulmars.

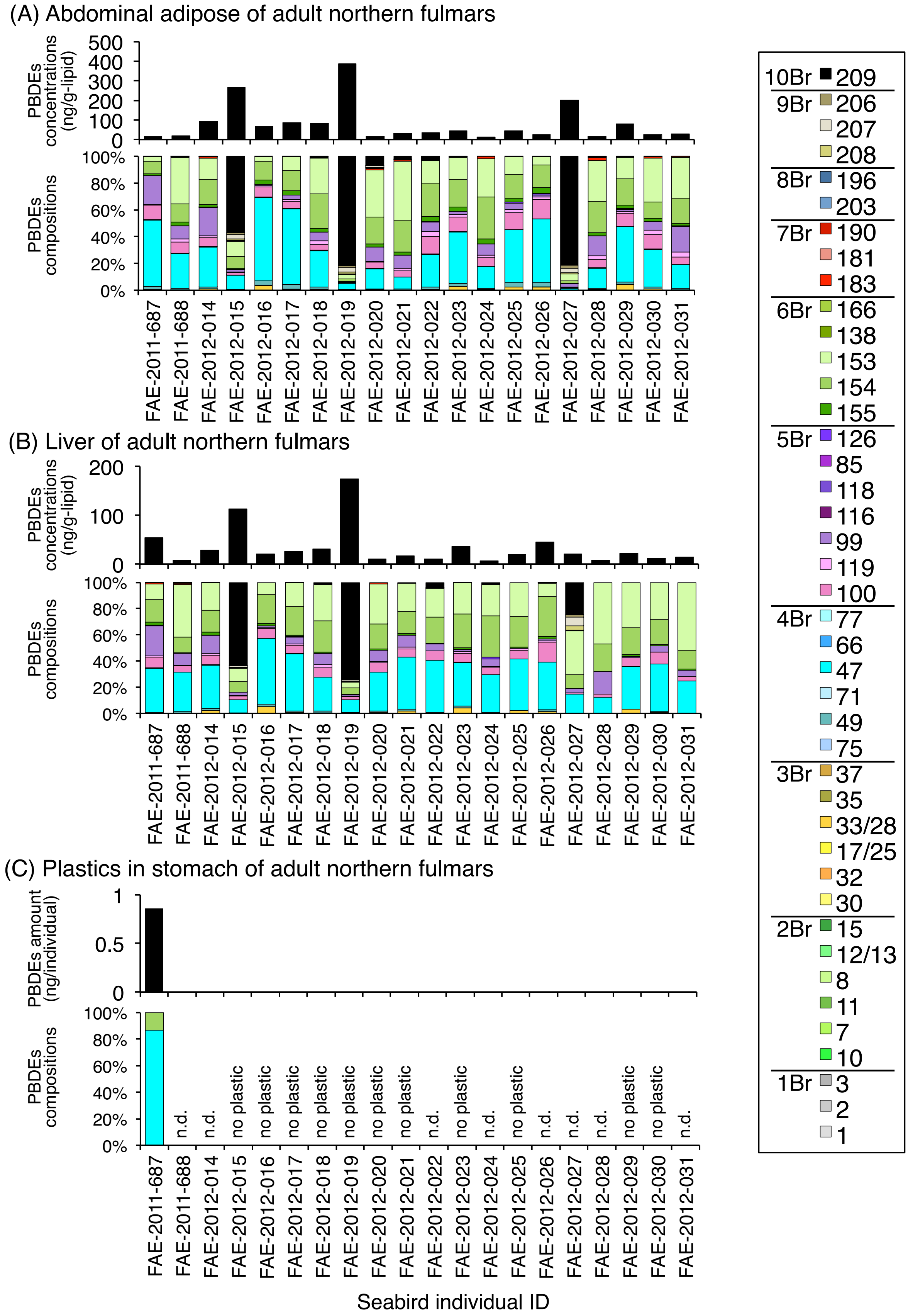


Figure 4-2. PBDE concentrations and compositions in the (A) abdominal adipose, (B) liver, (C) plastics in the stomach of adult northern fulmars. n.d. indicates not detected.

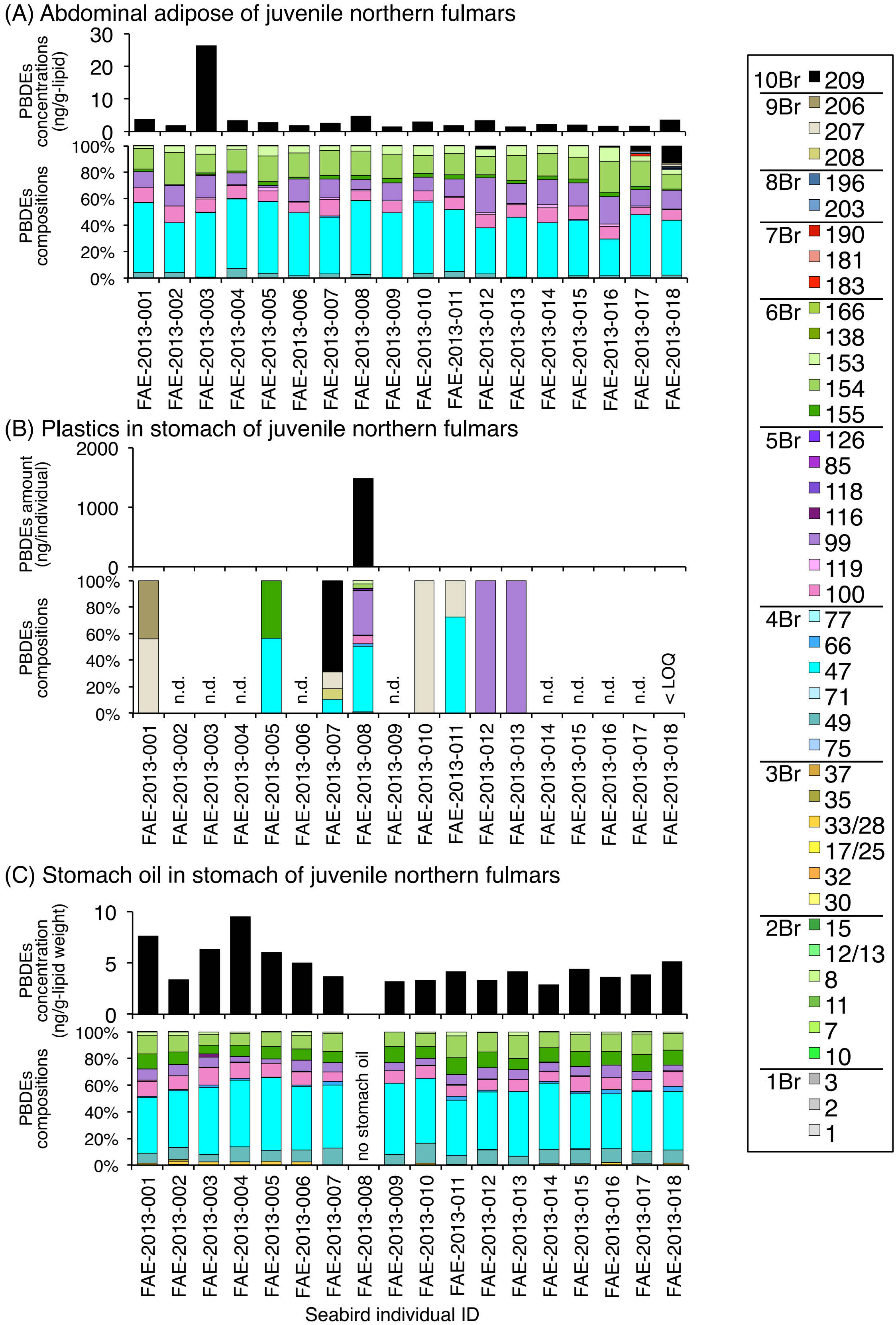


Figure 4-3. PBDE concentrations and compositions in the (A) abdominal adipose, (B) plastics in the stomach, (C) stomach oil in the stomach of juvenile northern fulmars. n.d. indicates not detected.

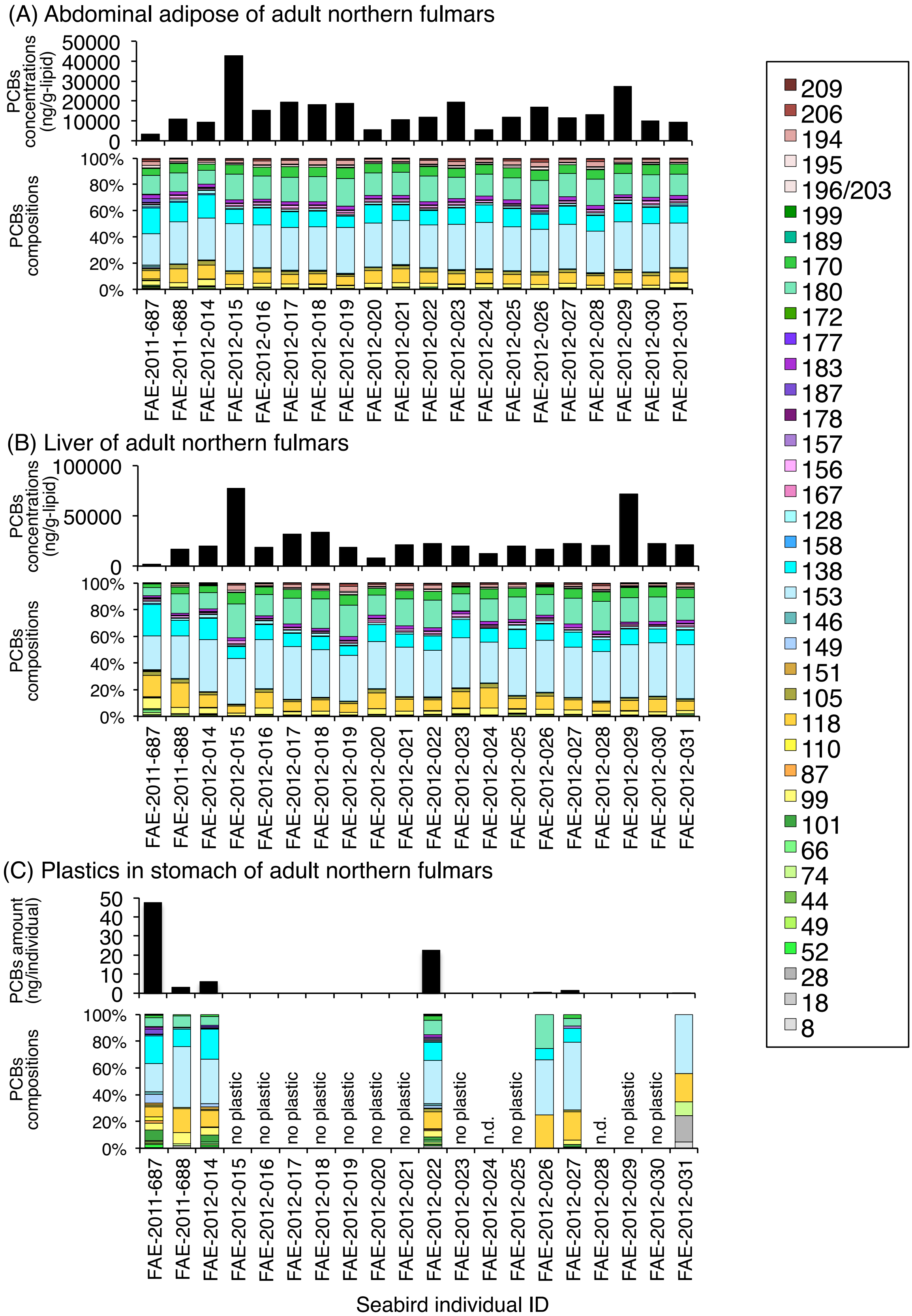
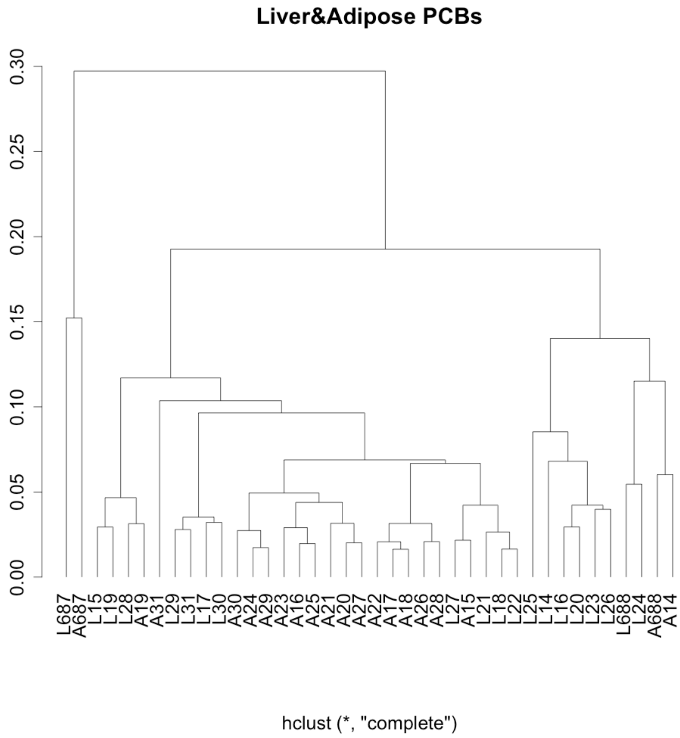


Figure 4-4. PCB concentrations and compositions in the (A) abdominal adipose, (B) liver, (C) plastics in the stomach of adult northern fulmars. n.d. indicates not detected.

(A) cluster dendrogram



(B) NMDS ordination

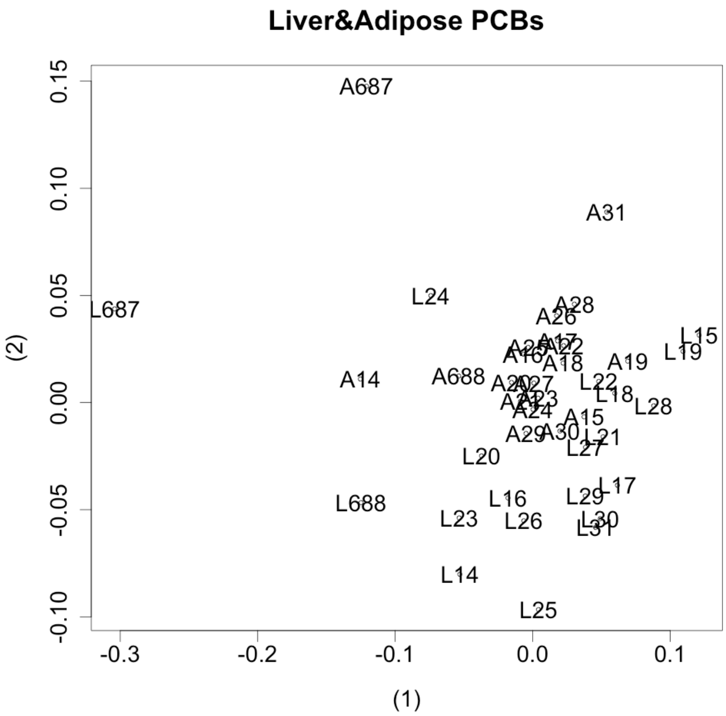


Figure 4-5. **(A)** Cluster dendrogram of PBDE compositions in liver (L-), abdominal adipose (A-), and plastics (P-) in gizzard of short-tailed shearwaters. **(B)** Nonmetric multidimensional scaling (NMDS) of PBDE congener profiles.

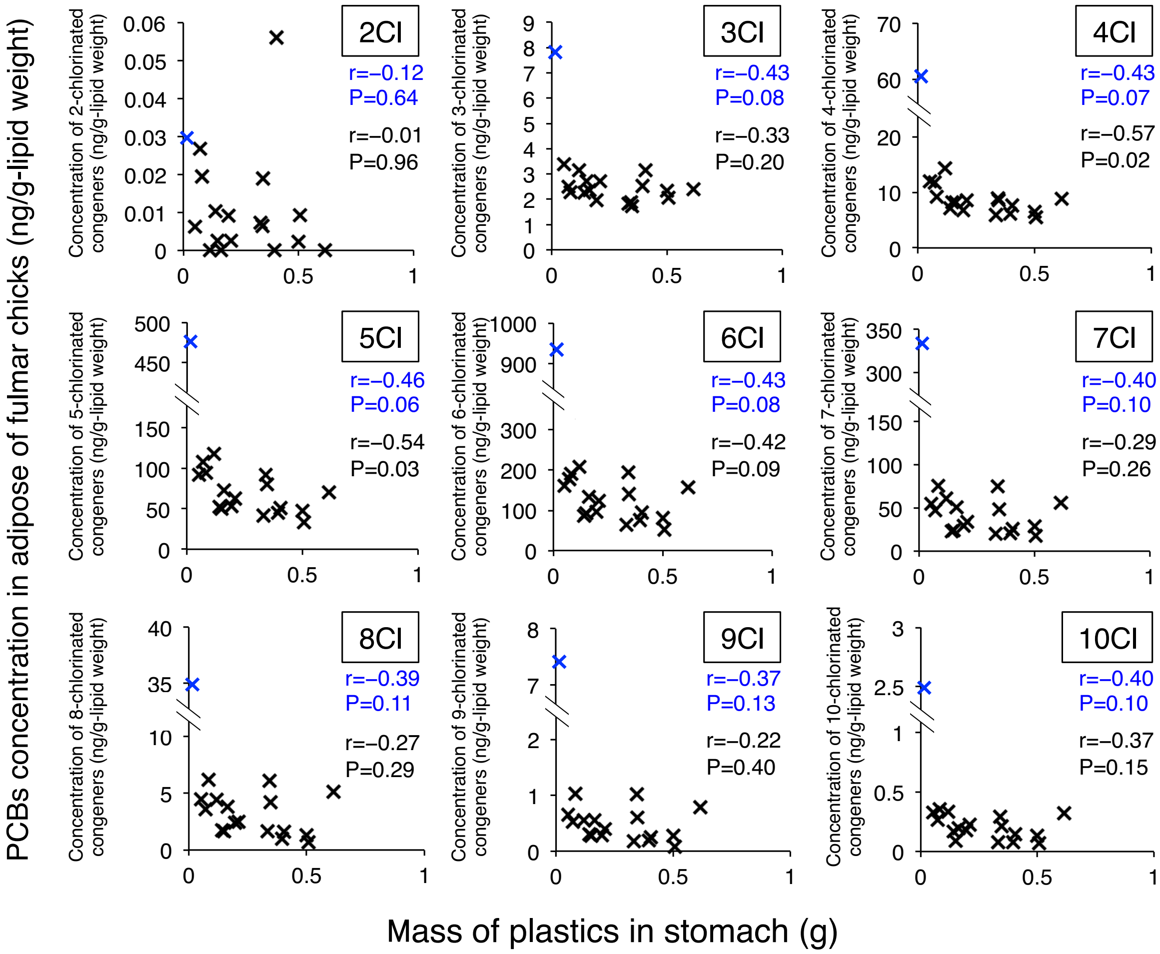


Figure 4-6. PCBs concentration in tissue vs mass of ingested plastics in chicks of northern fulmars. Blue character: r-value and P-value of t-test calculated using all of the samples; Black character: r-value and P-value of t-test calculated excluding an outlier, FAE-2013-003 (blue plot).

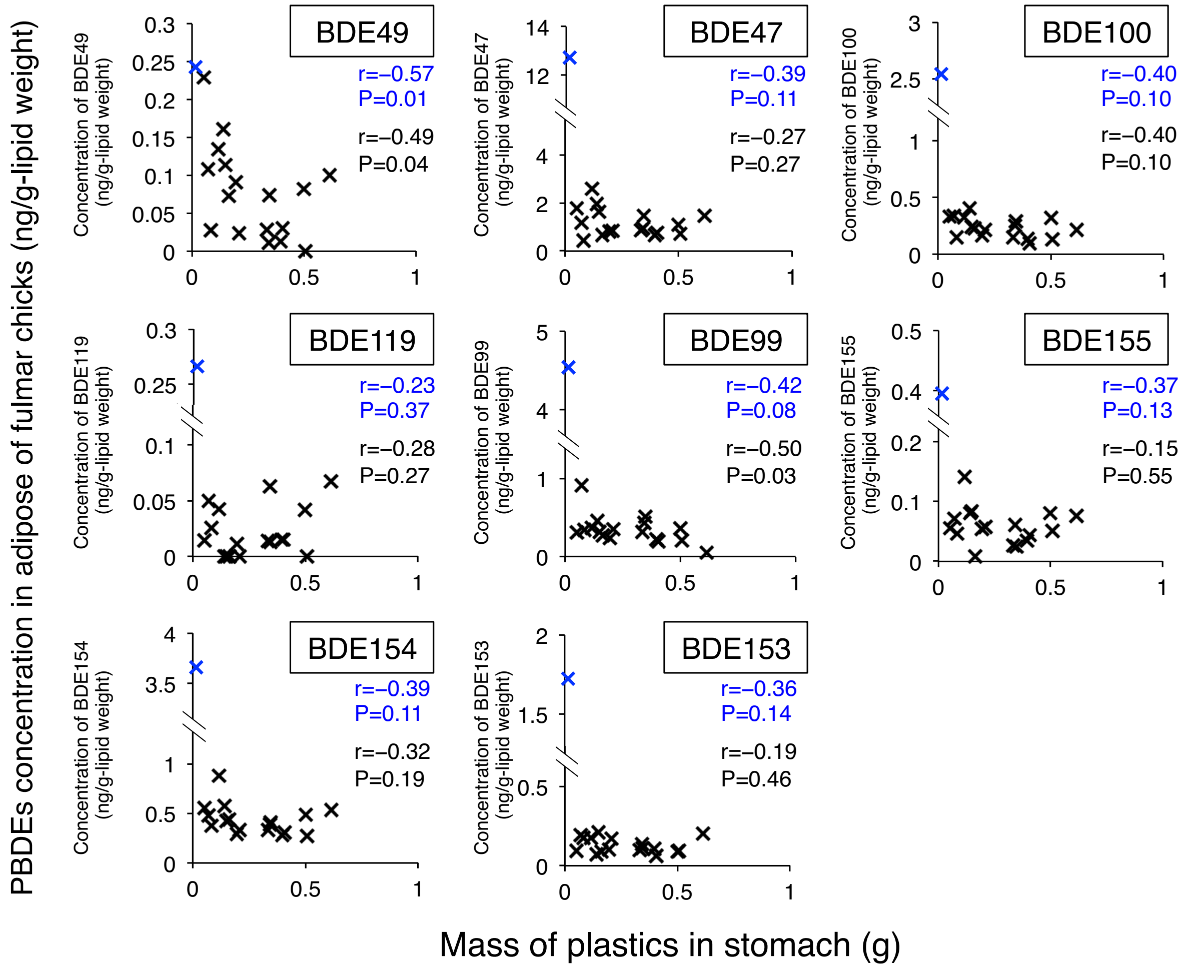


Figure 4-7. PBDEs concentration in tissue vs mass of ingested plastics in chicks of northern fulmars. Blue character: r-value and P-value of t-test calculated using all of the samples; Black character: r-value and P-value of t-test calculated excluding an outlier, FAE-2013-003 (blue plot).

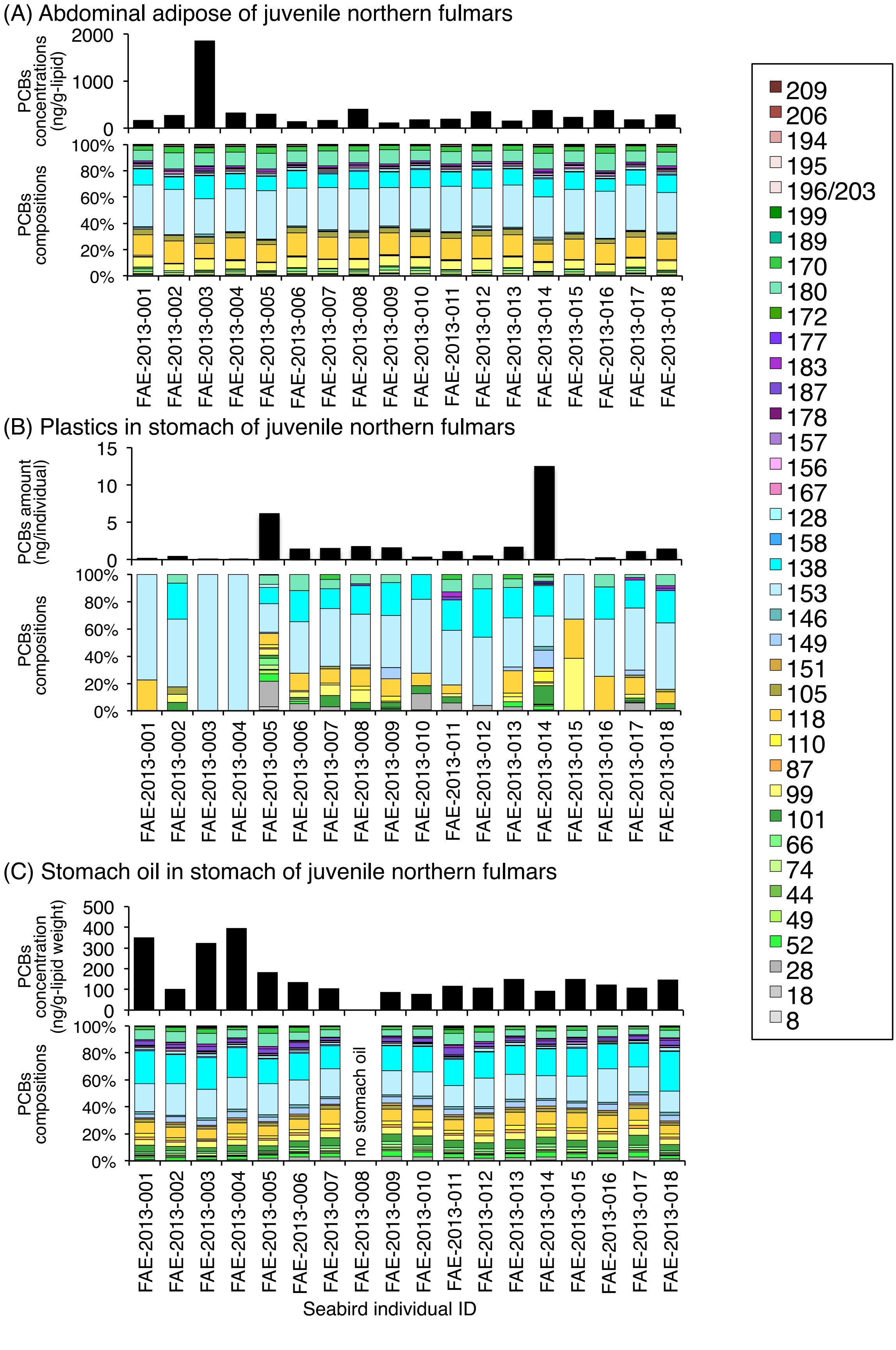


Figure 4-8. PCB concentrations and compositions in the (A) abdominal adipose, (B) plastics in the stomach, (C) stomach oil in the stomach of juvenile northern fulmars. n.d. indicates not detected.

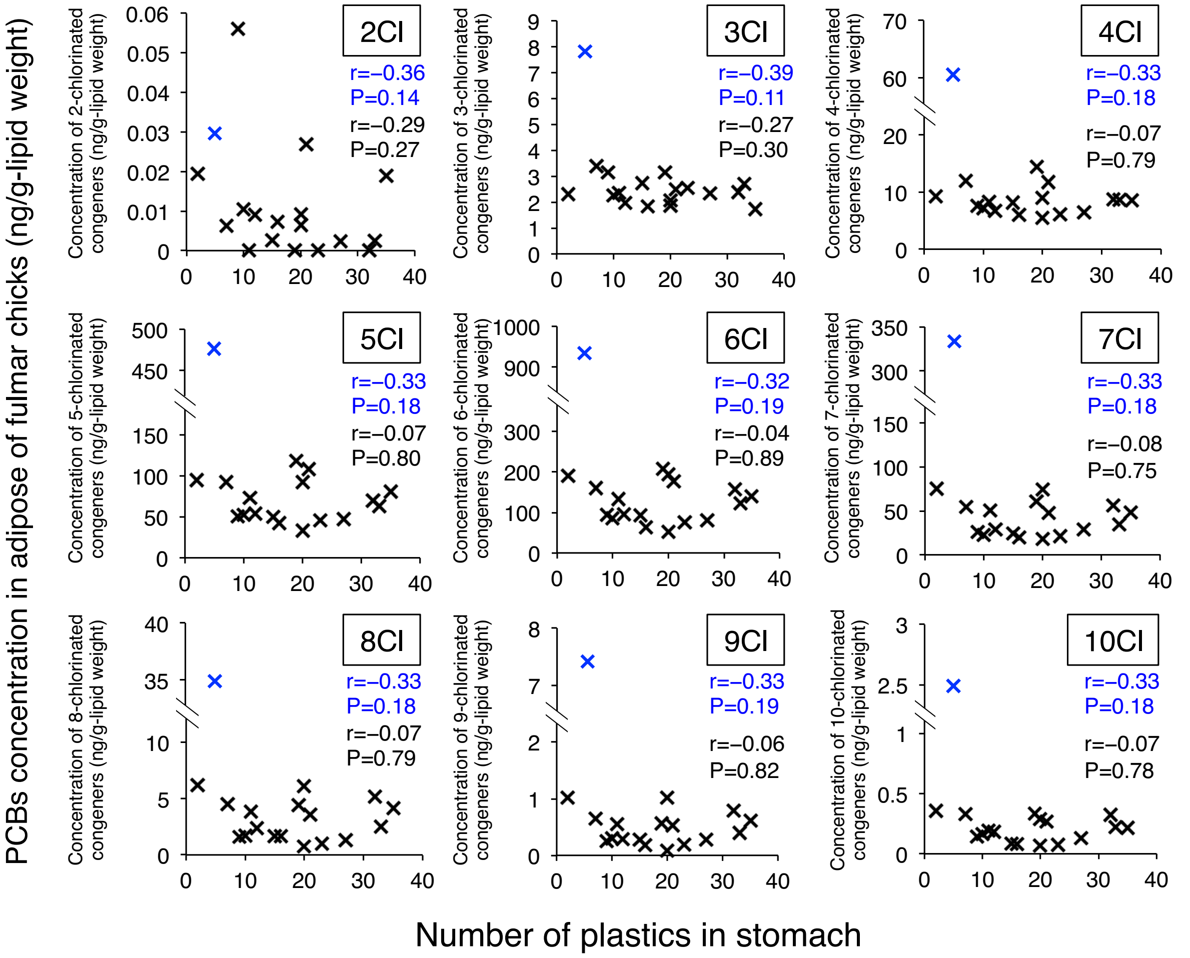


Figure 4-9. PCBs concentration in tissue vs number of ingested plastics in chicks of northern fulmars. Blue character: r-value and P-value of t-test calculated using all of the samples; Black character: r-value and P-value of t-test calculated excluding an outlier, FAE-2013-003 (blue plot).

Figure 4-10. Examples of plastics in the stomach of white-chinned petrels.

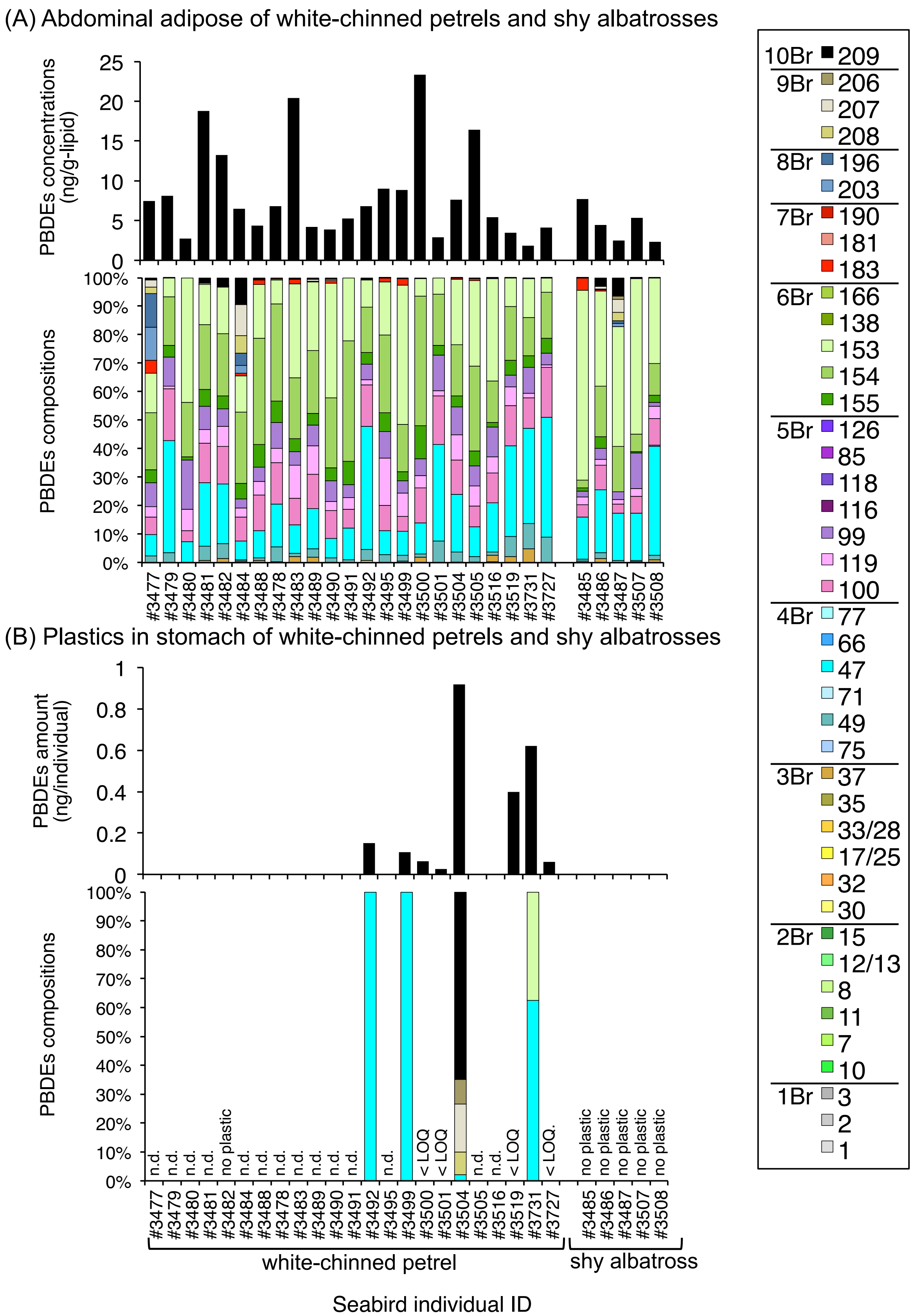


Figure 4-11. PBDE concentrations and congener compositions in (A) abdominal adipose, (B) plastics in stomach of white-chinned petrels and shy albatrosses.

Table 4-1. Amount and Number of ingested plastics and breeding status of adult northern fulmar.

ID	Number	Amount (g)	Breeding status
FAE-2011-687	23	0.18	NB
FAE-2011-688	10	0.10	B
FAE-2012-014	19	0.06	NB
FAE-2012-015	-	-	B
FAE-2012-016	-	-	B
FAE-2012-017	8	0.17	B
FAE-2012-018	-	-	B
FAE-2012-019	-	-	B
FAE-2012-020	-	-	B
FAE-2012-021	-	-	B
FAE-2012-022	6	0.11	B
FAE-2012-023	-	-	B
FAE-2012-024	2	0.02	B
FAE-2012-025	-	-	B
FAE-2012-026	1	0.01	B
FAE-2012-027	5	0.03	B
FAE-2012-028	2	0.02	B
FAE-2012-029	-	-	B
FAE-2012-030	-	-	B
FAE-2012-031	2	0.01	B
Average	3.9	0.03	

NB: non-breeding, B: breeding

Table 4-2. Amount and Number of ingested plastics in chicks of northern fulmar.

ID	Number	Amount (g)
FAE-2013-001	10	0.14
FAE-2013-002	11	0.16
FAE-2013-003	5	0.02
FAE-2013-004	7	0.05
FAE-2013-005	32	0.62
FAE-2013-006	16	0.33
FAE-2013-007	27	0.50
FAE-2013-008	19	0.12
FAE-2013-009	20	0.51
FAE-2013-010	15	0.15
FAE-2013-011	12	0.20
FAE-2013-012	21	0.07
FAE-2013-013	23	0.40
FAE-2013-014	20	0.34
FAE-2013-015	33	0.21
FAE-2013-016	2	0.08
FAE-2013-017	9	0.40
FAE-2013-018	35	0.35
Average	17.6	0.26

Table 4-3. PBDE concentrations in abdominal adipose of adult northern fulmars (ng/g-lipid weight).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-015	FAE-2012-016	FAE-2012-017	FAE-2012-018	FAE-2012-019	FAE-2012-020	FAE-2012-021	FAE-2012-022	FAE-2012-023	FAE-2012-024	FAE-2012-025	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-029	FAE-2012-030	FAE-2012-031
1Br	1																				
	2																				
	3																				
2Br	10																				
	7																				
	11																				
	8																				
	12/13								0.03												
	15							0.08				0.03	0.08								0.01
3Br	30		0.01																		
	32		0.01											0.01							
	17/25			*0.01*											*0.01*	0.01					
	33/28	0.05	0.01	0.89	0.08	2.16	0.52	0.17	0.31	0.05	0.03	0.13	1.24	0.03	1.09	0.63	0.09	0.02	3.38	0.19	
	35					0.40															
	37							*0.07*								0.06		*0.04*			
4Br	75			0.02			0.02									0.01			0.03		
	49	0.40	0.27	1.46	1.02	2.16	3.12	1.54	1.40	0.10	0.23	0.70	0.92	0.16	1.51	0.84	0.13	0.15	1.72	0.42	0.42
	71	0.01		0.02								0.01	0.03	0.01					0.01		
	47	8.16	4.93	27.7	29.0	41.6	48.4	23.2	17.3	2.61	3.01	9.06	17.0	2.07	18.5	13.01	2.42	2.64	33.6	7.25	5.13
	66	0.09	0.03	0.05	0.24	0.06	0.22	0.21	0.18	0.02		0.08			0.03		0.04	0.05	0.07	0.05	
	77	0.04	0.02	0.08		0.13	0.04			0.03			0.04		0.02				*0.02*	0.11	
5Br	100	1.80	1.63	6.20	4.30	4.98	4.53	3.56	2.24	0.81	1.55	4.94	4.87	0.87	5.97	3.93	1.34	1.01	8.21	2.77	1.76
	119	0.07	0.40	1.20	1.71	0.59	1.28	2.38	0.79	0.13	0.56	1.22	0.86	0.24	1.11	0.59	0.29	0.52	1.05	0.77	1.02
	99	3.50	1.90	19.6	4.58	0.78	2.59	5.45	1.68	1.89	3.28	2.73	0.89	1.08	2.02	0.35	5.02	2.57	1.12	1.74	5.60
	116			0.15			*0.04*						*0.05*		*0.07*						
	118			0.16	0.16		0.03	*0.04*	*0.08*			*0.05*			*0.06*	0.31	0.22		0.28		*0.09*
	85			*0.03*			0.02												*0.02*		
	126			0.21	0.17	0.11	0.24	0.16	*0.10*			0.08			0.31	*0.04*	0.14		0.34		*0.08*
6Br	155	0.27	0.56	1.88	2.10	1.93	2.70	2.30	1.35	0.38	0.64	1.39	1.56	0.50	1.36	1.15	0.51	0.36	1.89	0.66	0.60
	154	1.56	2.54	17.1	23.8	9.42	12.8	21.5	7.69	3.51	7.89	9.15	9.10	4.03	8.29	4.67	4.33	4.07	16.2	3.12	5.44
	153	0.52	6.57	14.9	29.8	2.44	8.81	22.5	12.2	6.09	14.5	6.17	7.14	3.71	6.13	1.60	9.78	5.23	12.8	8.44	8.92
	138			0.10											0.08						
	166					*0.01*										*0.01*					
7Br	183	0.04	0.17	0.72	1.01	0.05	0.17	0.74	0.31	0.12	0.22	0.15	0.22	0.20	0.04	0.03	0.17	0.38	0.59	0.26	0.27
	181			0.02																	
	190																				
	188																				
	179																				
8Br	202																				
	197			0.05	0.35				0.28	*0.06*							0.07			0.02	
	203																				
	196		*0.01*	0.02	0.61		0.01	0.06	0.33	*0.09*	*0.09*	*0.01*	*0.02*				0.27				
9Br	208		*0.001*	0.01	2.91			0.02	5.35	0.06							2.08	*0.01*	*0.005*		
	207	*0.001*	*0.01*	0.04	7.86	*0.01*	*0.02*	0.07	12.9	0.19	*0.12*	0.10	*0.04*	*0.005*		0.01	6.15	0.02	0.03	0.02	0.01
	206		*0.003*	0.02	4.41		*0.01*	0.03	4.98	*0.05*	*0.05*	*0.03*	*0.005*			*0.004*	4.37	*0.01*	0.02	*0.01*	*0.004*
10Br	209	*0.01*	*0.02*	0.19	150	0.05	0.09	0.41	316	1.17	0.70	0.96	0.10	*0.02*	0.03	0.09	165	0.10	0.20	0.04	
total		16.5	19.1	92.9	264	66.9	85.7	84.5	386	17.4	32.8	37.0	44.2	12.9	46.6	27.4	202	17.2	81.6	25.9	29.4

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-4. PBDE concentrations in liver of adult northern fulmars (ng/g-lipid weight).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-015	FAE-2012-016	FAE-2012-017	FAE-2012-018	FAE-2012-019	FAE-2012-020	FAE-2012-021	FAE-2012-022	FAE-2012-023	FAE-2012-024	FAE-2012-025	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-029	FAE-2012-030	FAE-2012-031
1Br	1																				
	2																				
	3																				
2Br	10																				
	7																				
	11																				
	8																				
	12/13																				
	15				0.04																
3Br	30																				
	32																				
	17/25																				
	33/28			0.70		1.11	0.25	*0.05*	0.32	0.10	0.34		1.51		0.41	0.63			0.68	*0.08*	
	35																				
	37																				
4Br	75															0.07					
	49	0.52	0.09	0.33	0.31	0.31	0.21	0.49	0.87	0.11	0.19	0.10	0.50	0.05	0.08	0.45				0.08	
	71																				
	47	17.8	2.54	9.60	11.3	10.4	10.9	8.09	16.6	3.05	6.86	4.10	12.0	1.90	7.52	16.7	2.99	0.99	7.17	4.34	3.48
	66	0.09		0.06	0.10		0.08		0.07				0.06								
	77																				
5Br	100	4.48	0.39	2.12	2.95	1.60	1.57	2.22	4.58	0.75	1.14	0.75	2.49	0.33	1.27	6.92	*0.15*	0.18	1.48	1.12	0.47
	119	0.45	*0.05*	0.40	0.83	*0.08*	0.25	0.72	0.59	0.11	0.21		0.57	*0.08*	0.28	0.47					
	99	12.3	0.74	3.99	2.51	0.31	1.30	2.61	1.66	0.85	1.56	0.57	0.38	0.36		0.59	0.68	1.36	*0.24*	0.56	0.66
	116						*0.13*														
	118	*0.14*						*0.09*													
	85																				
	126	*0.12*		*0.07*										*0.10*							
6Br	155	1.13	0.04	0.57	0.22	0.37	0.31	0.31	1.18	0.10	0.29	0.03	0.55		0.19	0.83			0.28	0.14	*0.13*
	154	9.15	1.02	4.84	8.79	4.51	5.42	7.34	7.99	2.02	2.87	2.12	9.32	2.08	4.51	14.0	2.06	1.70	4.50	2.28	2.06
	153	6.58	3.34	6.11	11.7	1.93	4.62	8.64	7.80	3.14	3.71	2.25	8.55	1.56	5.03	4.50	6.75	3.74	7.61	3.42	7.31
	138																				
	166																				
7Br	183	0.48	0.11	0.07	0.51		0.06	0.17	0.51	0.13	0.13		0.15			0.17					
	181																				
	190																				
	188																				
	179																				
8Br	202								0.25												
	197								0.14												
	203																				
	196				0.06				0.10								0.06				
9Br	208				0.35				0.61			*0.01*				0.04	0.70				
	207	*0.01*	*0.02*		0.85			*0.01*	1.38			*0.02*		*0.01*		0.08	1.37				
	206				0.32				0.34			*0.01*				*0.03*	0.38				
10Br	209	*0.03*	*0.03*		71.6			0.32	130			0.45		0.11		0.05	4.91				
total		53.3	8.38	28.9	112	20.6	25.1	31.1	175	10.4	17.3	10.4	36.0	6.59	19.3	45.6	20.1	7.96	22.0	12.0	14.1

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-5. PCB concentrations in abdominal adipose of adult northern fulmars (ng/g-lipid weight).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-015	FAE-2012-016	FAE-2012-017	FAE-2012-018	FAE-2012-019	FAE-2012-020	FAE-2012-021	FAE-2012-022	FAE-2012-023	FAE-2012-024	FAE-2012-025	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-029	FAE-2012-030	FAE-2012-031
2Cl	8					*0.05*															
3Cl	18																				
	28	18.2	27.7	44.5	56.1	57.0	68.0	54.9	29.2	25.9	35.5	44.0	47.2	15.6	57.3	27.5	25.5	36.0	61.3	18.8	49.2
4Cl	52	5.33	0.61	7.73		4.19	3.76	1.37	2.72			0.25	5.29		6.60	1.98	0.56	0.64	2.69		
	49	1.36		2.92		0.88	0.43	58.8	1.16						2.88	0.82			0.88		
	44																				
	74	36.5	207	190	251	168	158	149	105	76.6	150	170	180	98.1	234	164	208	150	412	160	231
	66	30.1	85.2	124	101	70.2	93.6	64.7	43.8	39.8	70.4	102	126	28.5	162	93.0	95.6	101	295	51.2	117
5Cl	101	19.9	3.94	26.8	7.22	33.4	27.6	14.2	12.5	1.84	1.10	12.4	26.3	0.91	35.8	18.3	1.58	1.06	32.3	5.74	4.35
	99	146	791	913	1460	830	797	915	380	318	557	610	828	676	599	572	730	499	2190	529	562
	87	13.1	32.6	44.9	58.8	26.2	43.9	40.2	13.3	13.5	15.5	32.5	46.9	14.2	36.1	21.4	23.5	21.6	57.4	9.8	22.7
	110	8.11	2.00	8.87	3.59	7.98	16.4	5.05	5.39	0.56		4.18	10.7	0.65	12.6	5.99	0.37	0.99	7.75	1.01	3.28
	118	301	3100	1840	4070	2190	2370	2860	1230	939	1910	1860	2500	1930	1510	1700	1690	1270	5580	2150	1490
	105	59.8	441	410	868	452	421	537	222	216	387	375	516	405	382	368	324	264	1280	437	275
6Cl	151	1.68	0.40	5.72	0.86	4.66	3.16					1.60	1.68		3.88	3.06			3.34		
	149	10.2	4.46	27.2	14.4	12.5	19.1	9.9	5.10	0.48	0.90	7.16	10.1	1.30	10.4	8.82	5.76	1.76	22.0	4.26	4.84
	146	12.5	5.17	79.5	34.7	25.6	38.0	28.8	14.0	3.09	4.79	13.6	15.8	5.85	25.1	11.1	12.4	7.40	38.5	7.77	22.0
	153	476	5460	7850	26800	7050	12700	12000	6430	2870	8040	8070	7730	4000	7130	6630	8620	7780	28500	9170	8730
	138	447	1980	3150	6860	2070	3120	3390	1340	1010	2120	2450	2720	1290	2800	2070	2580	1840	8630	2300	2340
	158	16.1	51.7	110	220	90.1	79.9	131	70.8	29.3	45.9	68.9	53.6	38.3	97.1	68.8	75.7	63.8	165	51.7	84.3
	128	33.6	191	479	1020	447	604	612	262	109	220	228	264	72.1	524	350	272	345	1120	357	425
	167	11.9	123	113	659	106	278	159	138	88	205	207	205	74.7	160	126	124	158	432	213	166
	156	20.1	214	187	1171	158	450	337	245	135	309	318	329	134	229	202	263	253	724	78.7	305
	157	5.52	48.1	47.5	222	64.2	87.9	95.2	65.9	35.1	71.3	67.8	58.7	36.2	62.4	60.7	50.6	58.9	127	77.3	76.2
7Cl	178			5.74	0.94	1.58	0.94								1.32	0.83			1.52	0.85	
	187	11.3	4.07	90.8	18.9	22.9	22.3	21.4	9.8	1.13	2.89	11.9	16.1	4.56	22.8	11.2	13.1	4.01	26.5	6.81	15.2
	183	23.6	274	412	1857	375	650	776	457	181	464	494	385	357	445	307	645	491	958	472	483
	177			1.78																	
	172	0.87		12.5	0.72	1.21	2.53	2.17				0.31	0.65		1.01				2.34		
	180	116	2430	2450	19600	2980	6260	7300	4440	1220	4340	4800	2670	2130	3370	2610	4340	4700	13200	4240	3720
	170	51.1	885	987	6610	1130	2000	2140	1400	416	1280	1510	975	980	1280	985	1650	1480	5470	1670	1370
	189	0.90	20.8	18.5	156	27.0	49.0	51.3	39.6	10.7	36.9	31.3	24.0	17.5	29.6	19.3	32.3	33.2	50.0	33.8	28.7
8Cl	199	0.97		17.8	1.69	1.72	2.59	2.61	0.36			1.28	1.19	0.43	3.10	0.64	1.41		0.78		1.09
	196/203	4.60	114	118	1010	80.4	326	316	300	66.3	233	289	132	112	142	118	237	267	554	131	195
	195	1.13	38.7	47.1	258	55.7	91.3	120	101	22.1	58.3	65.4	42.7	36.9	50.5	40.3	76.3	80.7	110	54.5	59.2
	194	7.26	220	131	2640	234	817	901	779	135	534	663	249	286	377	146	377	549	1060	294	383
9Cl	206	1.09	76.6	56.7	946	75.9	213	358	277	49.8	280	270	126	86.5	127	98.6	187	325	337	125	189
10Cl	209	0.62	33.3	39.1	459	70.9	130	195	152	25.8	147	98.2	76.9	32.3	71.1	57.4	47.8	115	222	54.2	80.7
total		1890	16900	20100	77400	18900	32000	33700	18600	8030	21500	22900	20400	12900	20000	16900	22700	20900	71700	22700	21400

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-6. PCB concentrations in liver of adult northern fulmars (ng/g-lipid weight).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-015	FAE-2012-016	FAE-2012-017	FAE-2012-018	FAE-2012-019	FAE-2012-020	FAE-2012-021	FAE-2012-022	FAE-2012-023	FAE-2012-024	FAE-2012-025	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-029	FAE-2012-030	FAE-2012-031
2Cl	8	0.03																			
3Cl	18																				
	28	14.3	15.7	18.5	34.7	29.9	33.6	26.9	16.8	12.0	30.2	27.6	21.5	7.37	22.4	13.3	13.1	16.3	28.9	8.52	22.5
4Cl	52	6.58		1.59			0.47		1.91				0.90			2.43				0.31	0.32
	49	3.52		0.97					0.74				0.92								
	44																				
	74	31.6	115	127	309	143	132	112	101	45.1	106	93.8	126	55.4	82.5	85.8	103	72.8	190	64.2	78.9
	66	26.2	50.8	86.3	163	84.1	94.3	75.8	50.6	24.9	53.8	61.3	88.6	17.6	56.0	48.1	51.3	42.3	116	21.4	43.7
5Cl	101	30.6	0.74	15.6	4.44	8.17	7.55	4.34	15.8	1.31	5.07	2.15	10.0	1.00	6.41	18.4	0.81	0.61	5.99	4.29	1.55
	99	108	381	466	1020	450	511	463	367	166	337	358	501	167	305	429	380	298	772	213	298
	87	16.0	21.5	33.6	41.7	21.6	26.8	25.5	28.5	6.15	14.7	18.6	27.6	9.25	15.2	20.3	17.7	9.51	34.3	7.29	18.2
	110	36.8	2.28	10.8	5.04	8.72	16.4	7.38	13.0	2.31	3.43	4.45	8.88	3.16	8.41	14.2	1.48	1.28	4.61	3.32	4.68
	118	208	1150	992	3430	1300	1410	1390	1280	518	1080	1020	1550	485	850	1220	947	905	2300	715	811
	105	55.5	348	302	824	384	500	428	315	142	288	291	475	140	262	391	233	245	605	226	257
6Cl	151	9.31		2.11	0.58				1.48	1.37			0.91	0.42	0.77	2.00				1.07	
	149	31.7	2.38	11.5	7.89	6.83	6.03	6.33	7.93	0.77	2.59	2.64	7.68	1.69	4.40	8.16	3.58	2.30	4.01	3.62	5.32
	146	53.2	4.59	35.8	18.4	18.6	19.4	15.6	27.5	4.47	4.79	8.64	11.6	5.60	13.6	23.8	6.66	3.54	12.1	7.66	15.1
	153	816	3580	2990	15600	5010	6510	6120	6680	1860	3600	3900	6860	2050	3970	5470	4030	4230	9940	3670	3280
	138	658	1660	1690	4700	2010	2330	2120	1680	762	1240	1290	2370	759	1650	1960	1600	1600	3850	1220	1250
	158	31.4	75.2	91.8	146	60.7	102	102	67.4	22.9	51.1	42.0	95.3	19.9	50.6	105	62.3	63.1	129	39.7	56.2
	128	47.5	188	196	600	227	292	291	210	74.4	159	152	293	59.6	167	220	155	183	441	147	157
	167	22.9	120	62.8	416	142	203	159	205	52.7	101	115	187	42.9	82.1	145	90.1	133	211	115	81.6
	156	32.0	186.4	91.5	662	193	298	247	323	85.3	155	157	277	85.6	127	244	138	190	315	171	144
	157	9.94	52.2	26.7	164	53.0	81.7	68.1	77.1	19.4	43.2	42.8	72.3	23.5	37.4	67.5	41.9	52.1	89.9	47.5	38.5
7Cl	178	5.65																			
	187	103	3.37	43.5	14.4	11.4	21.8	15.7	32.1	6.94	7.49	8.35	16.6	4.79	15.8	38.9	10.1	6.18	5.85	8.29	16.1
	183	97.7	240	234	981	341	512	471	476	129	260	287	504	153	296	407	324	359	560	251	246
	177	5.76																			
	172	9.13		0.93			1.04		1.83												
	180	477	1630	997	8370	2650	3670	3400	3990	942	1840	2280	3190	974	2020	3120	2090	2660	4480	1710	1560
	170	192	790	454	3040	1050	1520	1290	1580	384	722	874	1320	417	872	1310	777	991	1990	754	732
	189	5.05	23.6	7.81	99.4	29.0	41.6	36.6	49.7	8.25	17.0	23.4	45.5	10.6	23.5	40.3	19.5	28.1	37.7	16.1	13.4
8Cl	199	9.79		2.69	0.37	0.87			1.72				0.83			2.39					
	196/203	51.9	90.1	100	456	204	247	277	274	51.2	93.7	150	292	62.9	180	327	155	228	256	107	109
	195	13.0	29.1	22.2	152	59.7	83.0	79.8	81.5	13.1	25.8	40.6	99.6	18.1	62.4	109	39.5	62.1	75.2	33.4	32.6
	194	85.7	179	141	847	475	580	596	633	104	191	344	580	123	386	612	241	498	510	208	191
9Cl	206	46.6	53.7	85.9	408	164	203	196	256	30.5	63.6	126	236	42.2	121	286	101	228	175	72.1	65.5
10Cl	209	48.7	49.4	59.7	235	134	148	95.1	106	20.1	32.8	70.4	239	27.5	108	204	32.2	97.1	127	41.9	33.2
total		3400	11000	9400	42800	15300	19600	18100	19000	5500	10500	11800	19500	5770	11800	17000	11700	13200	27300	9890	9550

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-7. PBDE concentrations in abdominal adipose of juvenile northern fulmars (ng/g-lipid weight).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
1Br	1																		
	2																		
	3																		
2Br	10																		
	7																		
	11																		
	8																		
	12/13																		
	15																		
3Br	30																		
	32																		
	17/25																		
	33/28				0.02											0.01			
	35																		
	37																		
4Br	75																		
	49	0.16	0.07	0.24	0.23	0.10	0.03	0.08	0.14		0.11	0.09	0.11	0.01	0.01	0.02	0.03	0.03	0.07
	71																		
	47	1.96	0.68	12.7	1.79	1.47	0.88	1.11	2.61	0.74	1.64	0.83	1.19	0.66	0.95	0.84	0.45	0.75	1.48
	66	0.02		0.14	0.02			0.03	0.03		0.03					0.01			
	77			0.04															
5Br	100	0.40	0.23	2.54	0.33	0.22	0.15	0.32	0.33	0.13	0.24	0.17	0.34	0.14	0.25	0.21	0.15	0.10	0.29
	119			0.27	*0.01*	0.07	*0.01*	0.04	0.04			*0.01*	0.05	*0.01*	0.06		0.03	*0.02*	*0.01*
	99	0.46	0.27	4.54	0.31	0.05	0.31	0.36	0.37	0.21	0.31	0.24	0.90	0.22	0.43	0.35	0.34	0.19	0.51
	116																		
	118			0.05															
	85																		
	126																		
6Br	155	0.08	*0.01*	0.40	0.06	0.08	0.03	0.08	0.14	0.05	0.08	0.05	0.07	0.04	0.06	0.06	0.05	0.04	0.02
	154	0.57	0.44	3.66	0.55	0.53	0.33	0.48	0.88	0.27	0.42	0.29	0.48	0.27	0.39	0.33	0.37	0.31	0.41
	153	0.07	0.09	1.72	0.09	0.20	0.10	0.09	0.18	0.10	0.21	0.10	0.19	0.11	0.14	0.17	0.18	0.06	0.11
	138																		
	166																		
7Br	183										0.01							0.03	
	181																		
	190																		
	188																		
	179																		
8Br	202																	*0.003*	
	197																		
	203																	*0.02*	*0.04*
	196																		0.03
9Br	208												0.01						0.03
	207		*0.001*	*0.001*	*0.001*	*0.001*	*0.001*						0.01				*0.003*	0.02	0.07
	206												0.001						0.02
10Br	209	*0.01*	*0.002*	*0.005*	*0.002*	*0.004*	*0.004*	*0.004*	*0.01*	*0.004*	*0.001*	*0.002*	0.07	*0.002*	*0.003*	*0.004*	*0.01*	0.05	0.46
total		3.74	1.79	26.3	3.41	2.73	1.85	2.59	4.71	1.50	3.06	1.79	3.42	1.46	2.29	2.01	1.61	1.62	3.56

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-8. PBDE concentrations in stomach oil in stomach of juvenile northern fulmars (ng/g-lipid weight).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008*	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
1Br	1								-										
	2								-										
	3								-										
2Br	10								-										
	7								-										
	11								-										
	8								-										
	12/13								-										
	15								-										
3Br	30								-										
	32								-										
	17/25		0.02			0.03			-							0.02	0.01		
	33/28	0.12	0.08	0.15	0.24	0.16	0.13		-		0.06	0.03	0.02		0.03	0.03	0.06	0.05	0.09
	35		0.05						-										
	37								-										
4Br	75				0.01				-										
	49	0.57	0.29	0.38	1.04	0.48	0.44	0.47	-	0.25	0.49	0.26	0.36	0.29	0.32	0.48	0.37	0.35	0.49
	71				0.01				-				*0.01*			0.01			
	47	3.15	1.43	3.16	4.74	3.29	2.37	1.73	-	1.70	1.60	1.75	1.44	2.00	1.41	1.82	1.47	1.72	2.27
	66	0.08	0.04	0.10	0.14	0.02	0.07	0.09	-			0.10	0.04		0.03	0.05	0.11	0.02	0.20
	77	0.01							-							0.02			
5Br	100	0.82	0.33	0.83	1.09	0.61	0.48	0.26	-	0.30	0.30	0.34	0.27	0.36	0.22	0.49	0.32	0.32	0.56
	119	0.10	*0.01*	0.04	0.06	*0.02*	0.04		-		*0.02*	0.03	0.02			*0.02*			0.04
	99	0.63	0.27	0.46	0.38	0.17	0.41	0.26	-	0.19	0.16	0.31	0.28	0.32	0.18	0.32	0.33	0.23	0.21
	116			0.15					-										
	118	*0.01*							-										
	85								-										
	126								-						*0.02*				
6Br	155	0.85	0.32	0.40	0.83	0.58	0.42	0.31	-	0.38	0.30	0.53	0.40	0.34	0.30	0.48	0.38	0.50	0.59
	154	1.05	0.41	0.51	0.84	0.63	0.52	0.49	-	0.35	0.32	0.69	0.48	0.72	0.34	0.56	0.47	0.58	0.64
	153	0.20	0.08	0.13	0.09	0.02	0.12	0.05	-		0.04	0.12	0.02	0.11	*0.01*	0.09	0.05	0.06	0.06
	138		*0.01*						-										
	166								-										
7Br	183	0.01					0.01		-									0.01	
	181								-										
	190								-										
	188								-										
	179								-										
8Br	202								-										
	197								-										
	203								-										
	196								-										
9Br	208								-										
	207								-										
	206								-										
10Br	209								-										
total		7.61	3.36	6.31	9.49	6.01	4.99	3.66	-	3.17	3.28	4.18	3.33	4.13	2.86	4.39	3.59	3.85	5.15

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

* no oil in stomach.

Table 4-9. PCB concentrations in abdominal adipose of juvenile northern fulmars (ng/g-lipid weight).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
2Cl	8	0.01		0.03	*0.01*		*0.01*	*0.002*		*0.01*	*0.00*	*0.01*	0.03		*0.01*	*0.002*	0.02	0.06	0.02
3Cl	18																		
	28	2.24	2.37	7.81	3.38	2.39	1.83	2.32	3.14	2.06	2.72	1.97	2.48	2.53	1.87	2.70	2.29	3.14	1.72
4Cl	52	0.54	0.08	1.60	0.41	0.35	0.18	0.21	0.82	0.18	0.29	0.36	0.22	0.20	0.31	0.11	0.14	0.28	0.21
	49	0.35	0.07	0.97	0.47	0.12	0.09	0.11	0.40	0.07	0.18	0.08	0.09	0.02	0.22	0.08	0.07	0.12	0.13
	44			0.08	0.03	0.04			0.04					0.04			0.02	0.04	
	74	3.17	4.70	33.2	6.33	4.36	2.87	3.24	7.16	2.81	3.85	3.38	6.32	2.97	5.21	4.39	5.30	3.68	4.86
	66	3.09	3.43	24.6	4.76	3.93	2.83	2.94	5.93	2.47	3.88	2.94	5.13	2.90	3.27	4.03	3.67	3.45	3.35
5Cl	101	2.68	0.74	13.3	2.60	1.47	1.02	1.16	4.24	0.93	1.43	1.10	1.50	0.98	2.64	0.92	0.74	1.45	2.03
	99	12.6	14.2	159	20.1	17.4	10.6	10.7	29.8	8.37	12.9	12.3	29.1	12.3	24.5	15.8	20.7	12.3	20.9
	87	1.33	0.87	10.5	2.07	0.93	0.96	0.97	2.31	0.70	0.98	0.84	2.38	0.97	2.30	0.96	0.94	0.86	1.83
	110	1.24	0.32	2.24	1.13	0.52	0.49	0.40	1.61	0.40	0.52	0.55	0.60	0.54	1.08	0.38	0.35	0.59	0.92
	118	27.3	46.4	207	54.4	39.5	23.4	27.4	64.1	18.4	27.1	31.0	58.9	24.4	50.3	36.2	59.1	28.3	44.0
	105	7.16	10.6	84.1	12.3	10.3	5.62	6.77	16.5	4.51	7.15	8.14	15.4	6.44	11.2	9.00	12.8	7.29	10.6
6Cl	151	0.16	0.06	3.38	0.41	0.13		0.34	0.78	0.09	0.29	0.09	0.13	0.11	0.72	0.19	0.07	0.23	0.22
	149	1.07	0.41	10.3	1.06	0.85	0.48	0.71	1.50	0.37	0.93	0.74	7.46	0.60	2.25	0.92	0.37	0.77	1.13
	146	1.74	1.09	29.4	2.08	1.88	1.31	1.65	3.88	1.00	2.24	1.25	3.17	1.31	5.30	2.25	1.54	1.27	2.93
	153	54.7	93.8	506	106	111	39.0	53.2	130	32.5	56.6	65.7	101	48.7	118	76.7	135	62.9	85.2
	138	21.1	26.1	317	38.0	32.2	17.4	18.0	54.3	13.2	24.6	20.6	48.9	18.9	52.0	30.4	37.1	21.0	37.8
	158	1.05	1.08	12.8	1.68	1.57	0.99	1.04	2.39	0.85	1.41	1.07	3.07	0.82	2.57	1.84	1.50	0.93	2.36
	128	2.98	4.04	29.0	4.68	4.21	2.34	2.24	6.65	1.86	3.19	2.70	6.16	2.43	5.73	4.60	4.68	3.03	4.28
	167	1.05	2.22	8.27	2.27	1.72	0.83	1.18	2.66	0.69	1.03	1.27	2.31	0.96	2.44	1.74	2.90	1.36	1.76
	156	1.83	3.81	13.4	3.68	2.98	1.22	1.68	4.02	1.10	1.90	2.25	3.77	1.46	4.79	3.15	5.19	2.04	3.32
	157	0.48	0.92	4.28	0.85	0.07	0.39	0.52	1.06	0.20	0.54	0.56	0.97	0.40	1.10	0.82	1.30	0.46	0.67
7Cl	178			2.17					0.06			0.06			0.23				0.06
	187	1.11	0.68	26.7	1.44	1.06	0.77	0.96	3.00	0.78	1.23	0.60	1.62	0.64	4.22	1.16	0.92	0.59	1.79
	183	2.48	4.37	40.6	5.13	5.39	2.05	2.67	6.55	1.50	2.75	2.80	5.12	2.01	7.65	3.20	6.63	2.53	5.15
	177			1.48															
	172			3.94		0.05			0.20	0.04						0.05	0.06		
	180	14.0	33.4	187	34.4	35.3	12.0	18.4	35.5	12.3	14.1	18.0	28.3	13.1	44.8	21.1	48.8	15.7	29.9
	170	5.40	12.2	70.0	13.6	13.6	4.84	6.78	15.3	3.35	6.04	7.62	12.1	5.32	17.6	8.73	18.5	6.84	11.2
	189	0.08	0.29	2.40	0.22	0.29	0.05	0.06	0.30	0.04	0.03	0.06	0.26	0.04	0.39	0.23	0.43	0.06	0.15
8Cl	199	0.01		3.29		0.02		0.02	0.10	0.02			0.06		0.38	0.02			
	196/203	0.50	1.03	11.3	1.31	1.53	0.40	0.44	1.58	0.25	0.53	0.65	1.13	0.37	1.98	0.74	1.83	0.59	1.34
	195	0.14	0.35	3.49	0.35	0.49	0.07	0.06	0.35		0.08	0.21	0.28	0.04	0.53	0.17	0.58	0.08	0.19
	194	1.08	2.44	16.9	2.85	3.09	1.17	0.81	2.38	0.47	1.04	1.51	2.14	0.59	3.22	1.55	3.76	0.94	2.63
9Cl	206	0.29	0.55	7.41	0.65	0.79	0.18	0.28	0.56	0.08	0.27	0.28	0.53	0.19	1.02	0.40	1.02	0.25	0.61
10Cl	209	0.17	0.20	2.49	0.33	0.32	0.08	0.13	0.34	0.07	0.09	0.18	0.27	0.08	0.29	0.22	0.36	0.14	0.22
total		173	273	1860	329	300	136	167	409	112	180	191	351	152	380	235	379	183	284

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-10. PCB concentrations in stomach oil in stomach of juvenile northern fulmars (ng/g-lipid weight).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008*	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
2Cl	8	0.01	0.03	0.01	0.02	0.03	0.06	0.05	-	0.04	0.03	0.02	0.05	0.04	0.04	0.04	0.05	0.05	0.03
3Cl	18		0.04	0.54	0.20	0.16	0.33	0.22	-	0.27	0.17	0.28	0.30	0.45	0.37	0.26		0.44	0.34
	28	3.32	0.01	3.37	3.91	3.14	3.29	2.73	-	2.80	2.13	2.32	1.70	3.01	2.45	3.15	3.01	2.57	2.07
4Cl	52	7.43	2.54	5.97	8.36	4.15	3.78	3.09	-	3.33	2.71	3.19	2.42	4.04	3.13	4.07	3.19	3.70	3.04
	49	2.10	0.65	1.39	2.76	1.29	0.99	0.84	-	1.04	0.84	0.88	0.70	1.19	0.86	1.31	0.74	1.08	0.90
	44	1.44	0.87	0.99	2.52	1.17	1.28	0.82	-	1.40	0.95	1.02	0.81	0.58	1.18	1.02	1.02	1.12	0.96
	74	4.16	1.65	5.48	4.83	1.74	1.96	1.89	-	1.73	1.25	1.65	1.59	2.05	1.51	2.87	1.89	1.62	1.68
	66	4.76	1.76	4.57	4.56	2.13	2.22	2.17	-	2.03	1.50	2.03	1.90	2.50	1.92	2.71	1.99	1.97	1.92
5Cl	101	17.3	3.29	9.27	19.1	6.72	6.32	5.90	-	4.90	5.04	6.18	4.81	9.71	4.84	7.48	6.14	7.81	6.91
	99	15.0	3.18	14.4	16.3	6.87	5.81	5.52	-	4.06	3.73	4.26	5.51	7.58	4.60	7.09	6.38	5.22	5.39
	87	5.89	1.41	3.08	7.73	2.82	2.03	1.99	-	1.81	1.39	1.42	1.75	3.06	1.83	3.14	1.93	2.39	2.19
	110	9.91	1.69	3.39	8.11	3.33	3.51	3.06	-	2.43	2.66	2.59	2.33	4.63	2.64	3.44	3.17	3.85	3.46
	118	29.2	7.88	23.1	33.1	13.7	10.4	11.3	-	7.67	7.08	9.46	10.4	14.3	8.45	16.0	11.7	9.30	9.39
	105	7.81	2.16	6.70	8.89	3.61	2.86	3.01	-	2.26	2.00	2.27	2.97	3.91	2.25	4.31	2.92	2.78	2.39
6Cl	151	3.66	1.33	2.77	5.20	2.16	2.12	1.14	-	1.45	0.76	1.69	1.00	1.62	1.39	1.59	1.57	1.77	2.15
	149	10.3	4.04	11.2	18.4	5.55	6.37	3.96	-	4.06	3.79	4.62	3.18	6.69	3.98	5.12	5.57	6.08	6.00
	146	5.77	1.18	6.09	7.46	3.36	2.83	1.56	-	1.33	1.41	2.13	1.57	2.58	1.50	2.04	1.94	2.42	3.34
	153	73.1	23.4	68.9	91.4	41.5	24.8	21.5	-	15.5	14.1	18.2	22.4	27.4	15.6	27.6	29.8	19.8	22.9
	138	85.4	21.6	74.6	88.1	33.3	26.5	17.9	-	16.0	14.6	22.3	20.9	31.4	18.4	30.9	22.4	18.3	42.2
	158	2.55	0.49	3.78	3.20	1.08	1.22	0.70	-	0.46	0.55	0.70	0.60	0.77	0.58	0.94	0.67	0.77	1.13
	128	5.80	1.78	5.27	5.41	3.02	2.13	1.30	-	1.15	0.99	1.39	1.98	2.26	1.21	2.93	1.20	1.36	3.03
	167	2.02	0.64	1.54	2.30	0.90	0.75	0.63	-	0.30	0.34	0.43	0.46	0.72	0.46	0.89	0.72	0.39	0.76
	156	2.66	1.32	3.75	4.04	1.75	1.17	0.99	-	0.63	0.54	0.23	0.85	1.26	0.74	1.62	0.81	0.65	1.19
	157	0.91	0.19	1.04	0.84	0.56	0.31	0.26	-	0.13	0.15	1.20	0.25	0.25	0.21	0.57	0.09	0.13	0.23
7Cl	178	0.67	0.26	0.81	0.48	0.65	0.25	0.06	-	0.20	0.06	0.34	0.12	0.09	0.16	0.16			0.49
	187	10.4	2.68	11.1	8.63	5.29	4.90	1.89	-	2.60	2.14	5.79	2.40	3.54	2.33	2.88	2.26	2.58	5.67
	183	3.66	1.37	5.23	3.44	3.05	1.67	0.81	-	0.77	0.77	1.72	1.33	1.43	1.00	1.47	1.08	0.90	1.61
	177		0.47	0.69		0.30	0.62		-	0.21	0.37	0.54	0.19	0.29	0.34	0.50	0.17	0.26	0.97
	172			0.31	0.46		0.16		-					0.09					
	180	25.5	7.56	24.9	22.7	17.4	8.46	5.62	-	4.25	4.18	9.65	7.52	6.82	5.35	7.79	7.02	4.43	7.84
	170	8.08	3.28	10.6	8.92	8.04	4.52	2.24	-	1.75	1.46	3.15	3.84	3.39	2.57	3.68	2.14	1.91	3.51
	189		0.02	0.10	0.05	0.06	0.05		-			0.03	0.02	0.04		0.02			
8Cl	199	0.46	0.16	2.33	0.68	0.53	0.50	0.10	-	0.09	0.16	1.56	0.12	0.10	0.19	0.14	0.11	0.14	0.23
	196/203	0.52	0.19	1.46	0.43	0.67	0.28	0.10	-	0.12	0.11	0.41	0.15	0.15	0.15	0.17	0.10	0.09	0.15
	195			0.15		0.24	0.04		-				0.03			0.03		0.03	0.08
	194	0.60	0.39	1.52	1.22	0.21	0.43	0.22	-	0.15	0.13	0.65	0.35	0.27	0.30	0.28	0.07	0.09	0.43
9Cl	206	0.37	0.07	0.87	0.24	0.27	0.10	0.08	-	0.08	0.04	0.38	0.14	0.13	0.07	0.12	0.05	0.04	0.11
10Cl	209	0.34	0.10	0.58	0.25	0.23	0.12	0.15	-	0.07	0.06	0.20	0.18	0.12	0.09	0.15	0.09	0.07	0.13
total		351	100	322	394	181	135	104	-	87	78	115	107	148	93	148	122	106	145

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-11. PBDE concentrations in plastics in stomach of adult northern fulmars (ng/individual).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-017	FAE-2012-022	FAE-2012-024	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-031
1Br	1										
	2										
	3										
2Br	10										
	7										
	11										
	8										
	12/13										
	15										
3Br	30										
	32										
	17/25										
	33/28										
	35										
	37										
4Br	75										
	49										
	71										
	47	0.74									
	66										
	77										
5Br	100										
	119										
	99										
	116										
	118										
	85										
	126										
6Br	155										
	154	0.11									
	153										
	138										
	166										
7Br	183										
	181										
	190										
	188										
	179										
8Br	202										
	197										
	203										
	196										
9Br	208										
	207			*0.01*		*0.01*					
	206										
10Br	209			*0.01*		*0.01*					
total		0.85		*0.02*		*0.02*					

Blank cells mean "not detected" (no peak was detected on chromatogram).
Italics in gray-highlighted cells show the values < LOQ.

Table 4-12. PBDE concentrations in plastics in stomach of juvenile northern fulmars (ng/individual).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
1Br	1																		
	2																		
	3																		
2Br	10																		
	7																		
	11																		
	8																		
	12/13																		
	15								0.16										
3Br	30																		
	32																		
	17/25								1.07										
	33/28								3.60										
	35																		
	37																		
4Br	75								0.48										
	49								12.5										
	71								1.24										
	47					0.11		0.17	730			0.31							
	66								27.1										
	77								1.07										
5Br	100								91.1										
	119								1.92										
	99								498				*0.12*	*0.11*					
	116																		
	118								3.61										
	85								16.9										
	126								0.84										
6Br	155					0.08			2.47										
	154								49.0										
	153								33.6										
	138								3.55										
	166																		
7Br	183								1.12										
	181																		
	190																		
	188																		
	179																		
8Br	202																		
	197																		
	203																		
	196																		
9Br	208	*0.02*		*0.02*				0.18	*0.02*	*0.04*	*0.06*								
	207	0.12	*0.03*	*0.12*	*0.02*			0.24	*0.02*	*0.03*	0.10	0.15	*0.01*	*0.01*	*0.01*	*0.01*	*0.01*		*0.04*
	206	0.09		*0.06*				*0.11*	*0.005*		*0.06*								
10Br	209	*0.23*	*0.11*	*0.33*	*0.08*	*0.06*	*0.08*	1.22	*0.03*	*0.08*	*0.26*	*0.03*	*0.02*		*0.06*	*0.05*	*0.05*	*0.06*	*0.21*
total		0.46	0.14	0.53	0.10	0.26	0.08	1.92	1480	0.15	0.49	0.49	0.16	0.12	0.07	0.05	0.06	0.06	0.26

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-13. PCB concentrations in plastics in stomach of adult northern fulmars (ng/individual).

		FAE-2011-687	FAE-2011-688	FAE-2012-014	FAE-2012-017	FAE-2012-022	FAE-2012-024	FAE-2012-026	FAE-2012-027	FAE-2012-028	FAE-2012-031
2Cl	8		*0.004*			*0.01*			*0.005*		*0.002*
3Cl	18					0.04					
	28	0.23	0.05	0.05		0.33					0.01
4Cl	52	1.16		0.11		0.23					
	49	0.42				0.04					
	44	0.08				*0.02*					
	74	0.55	0.05	0.06		0.39			*0.01*		*0.005*
	66	0.33	*0.01*	0.07		0.32					
5Cl	101	3.73		0.34		0.57			0.03		
	99	2.31	0.26	0.33		1.04			0.05		
	87	0.98				0.18					
	110	1.32		0.04		0.14					
	118	3.55	0.57	0.75		2.81		0.12	0.32		0.01
	105	0.68	0.04	0.05		0.49			0.02		
6Cl	151	0.63		0.11		0.16					
	149	3.04		0.16		0.39					
	146	0.93				0.31					
	153	10.1	1.46	2.03		7.36		0.21	0.78		0.02
	138	9.76	0.41	1.38		3.00		0.04	0.16		
	158	0.35				0.07					
	128	0.12	0.03	0.04		0.24					
	167	0.07		0.02		0.10					
	156	0.14				0.20					
	157	0.05							0.02		
7Cl	178										
	187	1.80		0.06		0.29					
	183	0.52	0.02			0.33					
	177	0.07		0.07							
	172	0.14									
	180	3.19	0.26	0.40		2.50		0.13	0.09		
	170	0.85	0.04	0.10		0.76			0.05		
	189										
8Cl	199	0.28									
	196/203	0.06				0.05					
	195										
	194					0.11					
9Cl	206	*0.02*				0.04					
10Cl	209	*0.01*				0.02					
total		47.4	3.21	6.16		22.5		0.50	1.54		0.05

Blank cells mean "not detected" (no peak was detected on chromatogram).
Italics in gray-highlighted cells show the values < LOQ.

Table 4-14. PCB concentrations in plastics in stomach of juvenile northern fulmars (ng/individual).

		FAE-2013-001	FAE-2013-002	FAE-2013-003	FAE-2013-004	FAE-2013-005	FAE-2013-006	FAE-2013-007	FAE-2013-008	FAE-2013-009	FAE-2013-010	FAE-2013-011	FAE-2013-012	FAE-2013-013	FAE-2013-014	FAE-2013-015	FAE-2013-016	FAE-2013-017	FAE-2013-018
2Cl	8					0.03	*0.004*				*0.003*	*0.004*							
3Cl	18					0.15													
	28					1.15	0.07	0.05	0.02		0.05	0.06	0.02	0.05	0.09			0.06	0.02
4Cl	52					0.33	0.03		*0.01*	*0.02*				0.06	0.35				
	49					0.17									0.09				
	44					*0.02*									0.06				
	74					0.20	0.02			*0.01*								*0.01*	
	66					0.31				*0.01*					*0.01*			*0.01*	
5Cl	101		0.03			0.14	0.02	0.12	0.08	0.06	0.02	0.05			1.73			0.03	0.06
	99		0.03			0.27	0.06	0.12	0.17	*0.01*		0.03		0.06	0.17	0.03		0.03	
	87					0.06									0.17				
	110					0.15	0.01	0.02	0.05	0.06				0.04	0.94				
	118	0.04				0.49	0.18	0.16	0.23	0.20	0.04	0.08		0.28	0.27	0.02	0.08	0.14	0.13
	105		0.02					0.03	0.02									0.02	0.02
6Cl	151														0.09				
	149					0.05			0.03	0.13				0.05	1.59			0.04	
	146														0.33				
	153	0.14	0.23	0.03	0.06	1.28	0.53	0.64	0.67	0.61	0.21	0.46	0.24	0.62	2.78	0.03	0.13	0.52	0.71
	138		0.12			0.73	0.32	0.23	0.38	0.38	0.07	0.25	0.17	0.38	2.74		0.07	0.23	0.34
	158														0.13				
	128					0.14									0.06				
	167																		
	156														0.02				
	157																		
7Cl	178																		
	187								0.02			0.03			0.11				0.03
	183					0.01						0.04						0.02	0.02
	177														0.09				
	172																		
	180		0.03			0.42	0.17	0.10	0.13	0.10		0.11	0.05	0.11	0.39		0.03	0.03	0.12
	170					0.03		0.06				0.04		0.06	0.26				
	189														0.02				
8Cl	199																		
	196/203																		
	195																		
	194																		
9Cl	206																		
10Cl	209																		
total		0.18	0.46	0.03	0.06	6.17	1.41	1.53	1.81	1.59	0.39	1.14	0.49	1.71	12.5	0.09	0.31	1.13	1.47

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

Table 4-15. Number (pieces) and mass (g) of plastic found in white-chinned petrels.

ID	Total number	Total mass (g)
#3477	1	2.02
#3478	4	0.08
#3479	1	0.06
#3480	2	0.04
#3481	3	0.10
#3482	–	–
#3483	10	0.27
#3484	1	0.01
#3488	1	0.01
#3489	5	0.12
#3490	2	0.07
#3491	1	0.12
#3492	5	0.10
#3495	11	0.06
#3499	7	0.16
#3500	1	0.02
#3501	2	0.03
#3504	6	0.09
#3505	9	0.11
#3516	2	0.04
#3519	4	0.10
#3727	12	0.11
#3731	14	1.51
Average	4.5	0.2

Table 4-16. PBDE concentrations in abdominal adipose of white-chinned petrels and shy albatrosses (ng/g-lipid weight).

		white-chinned petrel															
		#3477	#3479	#3480	#3481	#3482	#3484	#3488	#3478	#3483	#3489	#3490	#3491	#3492	#3495	#3499	#3500
1Br	1																
	2																
	3																
2Br	10																
	7																
	11																
	8																
	12/13																
	15																
3Br	30																
	32																
	17/25																
	33/28													0.05			
	35																
	37				0.14	0.18	*0.04*	*0.04*	*0.02*	0.45	0.08					0.04	0.47
4Br	75																
	49	0.17	0.28		0.96	0.72	0.03	0.03	0.36	0.20	0.13	0.07	0.06	0.27	0.25	0.18	0.23
	71																
	47	0.57	3.20	0.20	4.18	2.77	0.42	0.42	1.03	2.07	0.59	0.26	0.58	2.95	0.75	0.74	2.56
	66																
	77																
5Br	100	0.46	1.48	0.11	2.61	1.74	0.54	0.54	0.99	1.91	0.51	0.37	0.35	0.99	0.81	0.48	2.86
	119	0.26	*0.07*	0.21	0.88	0.93	0.21	0.21	0.34	2.33	0.43	0.13	0.21	0.13	1.49	0.71	1.02
	99	0.63	0.83	0.47	1.56	0.83	0.21	0.21	0.62	0.99	0.30	0.28	0.24	0.38	0.83	0.38	1.38
	116																
	118																
	85																
	126																
6Br	155	0.34	0.33	0.03	1.09	0.58	0.35	0.35	0.52	0.91	0.17	0.17	0.43	0.28	0.59	0.30	2.73
	154	1.50	1.38	0.51	4.31	2.92	1.61	1.61	2.32	4.36	0.94	0.94	2.24	1.09	2.46	1.47	10.6
	153	1.04	0.54	1.19	2.63	2.14	0.82	0.82	0.58	6.72	1.02	1.55	1.17	0.65	1.68	4.33	1.47
	138																
	166																
7Br	183	0.34				0.04	0.07	0.07	0.04	0.34	0.01	0.04			0.11	0.19	0.06
	181								0.02								
	190																
	188																
	179																
8Br	202																
	197																
	203	0.86	*0.01*				0.18										
	196	0.87					0.27										
9Br	208	0.17			*0.02*	*0.01*	0.40										
	207	0.19			0.05	*0.02*	0.71	*0.01*		0.03	0.03	0.02		0.01	*0.003*	0.02	
	206	0.03			*0.01*	0.02	0.02										
10Br	209	0.03			0.37	0.35	0.59	0.03		0.08	0.03	*0.02*		0.04	*0.01*	*0.02*	
total		7.44	8.13	2.71	18.8	13.3	6.47	4.34	6.83	20.4	4.23	3.84	5.30	6.84	9.00	8.86	23.4

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

(continued)

		white-chinned petrel							shy albatross				
		#3501	#3504	#3505	#3516	#3519	#3731	#3727	#3485	#3486	#3487	#3507	#3508
1Br	1												
	2												
	3												
2Br	10												
	7												
	11												
	8												
	12/13												
	15												
3Br	30												
	32				0.01								
	17/25												
	33/28								*0.01*				
	35												
	37		*0.004*	*0.03*	0.13	0.08	0.09		*0.03*	*0.06*		*0.02*	*0.02*
4Br	75												
	49	0.21	0.29	0.33	0.06	0.25	0.16	0.36	0.06	0.09	0.02	0.02	0.04
	71												
	47	0.97	1.54	1.72	0.93	1.12	0.61	1.73	1.13	0.99	0.41	0.89	0.87
	66												0.01
	77												
5Br	100	0.48	0.93	1.17	0.57	0.49	0.20	0.72	0.34	0.39	0.08	0.31	0.21
	119	0.05	0.68	1.18	0.31	0.23	0.03	0.04	0.21	0.09	*0.04*	0.15	0.10
	99	0.36	0.73	1.15	0.56	0.14	0.16	0.17	0.16	0.17	0.07	0.67	0.03
	116												
	118												
	85												
	126												
6Br	155	0.09	0.29	0.84	0.10	0.18	0.08	0.22	0.09	0.18		0.02	0.06
	154	0.51	1.38	4.91	0.78	0.66	0.24	0.66	0.21	0.80	0.39	0.32	0.25
	153	0.17	1.75	4.93	1.94	0.35	0.25	0.21	5.15	1.49	1.05	2.93	0.69
	138												
	166												
7Br	183		0.04	0.10	0.02				0.33	0.03		0.01	
	181												
	190												
	188												
	179												
8Br	202												
	197												
	203										0.02		
	196										0.02		
9Br	208									0.01	0.08		
	207		*0.002*	0.04	*0.002*	*0.001*	*0.004*	*0.001*	*0.004*	0.04	0.11	*0.005*	*0.003*
	206									0.01	0.03		
10Br	209			*0.04*			*0.004*	*0.001*	*0.01*	0.12	0.16	*0.002*	
total		2.86	7.65	16.4	5.40	3.50	1.84	4.10	7.72	4.48	2.49	5.36	2.29

Table 4-17. PBDE concentrations in plastics in the stomach of white-chinned petrels and shy albatrosses (ng/individual).

		white-chinned petrel															
		#3477	#3479	#3480	#3481	#3482	#3484	#3488	#3478	#3483	#3489	#3490	#3491	#3492	#3495	#3499	#3500
1Br	1																
	2																
	3																
2Br	10																
	7																
	11																
	8																
	12/13																
	15																
3Br	30																
	32																
	17/25																
	33/28																
	35																
	37																
4Br	75																
	49																
	71																
	47													0.15		0.06	
	66																
	77																
5Br	100																
	119																
	99																*0.03*
	116																
	118																
	85																
	126																
6Br	155																
	154																
	153																
	138																
	166																
7Br	183																
	181																
	190																
	188																
	179																
8Br	202																
	197																
	203																
	196																
9Br	208																
	207																
	206																
10Br	209															*0.05*	*0.04*
total														0.15		0.11	0.06

Blank cells mean "not detected" (no peak was detected on chromatogram).

Italics in gray-highlighted cells show the values < LOQ.

(continued)

		white-chinned petrel							shy albatross				
		#3501	#3504	#3505	#3516	#3519	#3731	#3727	#3485	#3486	#3487	#3507	#3508
1Br	1												
	2												
	3												
2Br	10												
	7												
	11												
	8												
	12/13												
	15												
3Br	30												
	32												
	17/25												
	33/28												
	35												
	37												
4Br	75												
	49												
	71												
	47		0.02				0.14						
	66												
	77												
5Br	100												
	119												
	99												
	116					*0.08*							
	118												
	85												
	126												
6Br	155												
	154						*0.02*						
	153						0.08						
	138												
	166												
7Br	183												
	181												
	190												
	188												
	179												
8Br	202												
	197												
	203												
	196												
9Br	208		0.07			*0.04*	*0.03*						
	207		0.15			*0.10*	*0.21*						
	206		0.08										
10Br	209	*0.03*	0.59			*0.18*	*0.13*	*0.06*					
total		0.03	1.00			0.40	0.62	0.06					

Chapter 5.

General discussion

5.1. Behavior of plastic derived PBDEs in the tissues and toxicity on seabirds

5.1.1. Distribution of PBDEs in liver and adipose of seabirds

The ratios of lipid-normalized concentration of PBDEs in liver to those in adipose tissue (L/A ratio) of major congeners were calculated for each individual of short-tailed shearwaters and adult northern fulmars (Figs 5-1 and 5-2). L/A ratio values of BDE47, 99, 100, 153, and 154 in most of the birds are between 0.1 and 1. On the other hand, one individual of short-tailed shearwater (OS10-008) and one northern fulmar (FAE2012-027) show remarkably deviated values of L/A ratio of BDE209 from the others (L/A ratio of 1000 and 0.02, respectively) (Figs 5-1 and 5-2).

Lower-brominated congeners, i.e., BDE47, 99, 100, 153, and 154 in seabirds are derived from their prey and accumulated by daily exposure. L/A ratio of PCBs, which were also accumulated from daily food intake, was in the same value range as the lower-brominated congeners in northern fulmar (Fig. 5-3). Moreover, in chickens from an electronic waste recycling area, where the chickens are always exposed to high concentration of PBDEs including BDE209, L/A ratio within 0.1 to 1 of BDE47, 99, 100, 153, 154 and 209 were observed (Fig. 5-4) [87]. These data indicates that L/A ratio of around 0.1 to 1 may be related to the equilibrium state of the chemicals in birds' body under daily exposure. On the other hand, two individuals with greatly deviated L/A values were indicated to be in non-equilibrium state. Deviated L/A values can be caused by the difference of accumulation and depletion rate between liver and adipose tissue. Both of accumulation and depletion rate of BDE209 are far more rapid in liver than in adipose tissue [88-90]. BDE209 absorbed in intestine is readily transferred to liver and then blood-rich tissues, but slowly distributed to the other tissues such as adipose tissue [88,89]. Half-life of BDE209 in liver is much shorter than that in the other tissues [90], and adipose seemed to be the last tissue to redistribute stored chemicals [91]. Therefore, high L/A ratio value observed in OS10-008 may indicate recent initiation of exposure to high concentration of BDE209, and low L/A ratio value observed in FAE2012-027 may indicate recent termination of exposure. In fact, OS10-008 ingested plastic with additive BDE209 (deca-BDE), and FAE2012-027 did not ingested plastics with BDE209. The greatly deviated L/A ratio of BDE209 from equilibrium state in the birds can be formed by exposure to BDE209, which are usually not exposed to them, for a limited certain period of time with high concentration. This way of exposure is not likely to occur in nature, and greatly deviated L/A ratio of BDE209 may be specific to exposure from ingested plastic with additive-derived PBDEs.

The author made a calculation on the elapsed time after excretion of plastic with BDE209 in the bird with the lowest L/A ratio (FAE2012-027). In one compartment model [92], elimination of chemicals in tissue are represented as follows:

$$lnC = lnC_0 - Kt \qquad (1)$$

$$K = \frac{ln2}{T_{1/2}} \qquad (2)$$

where C is concentration at time t, C_0 is initial concentration at t =0, K is the elimination rate constant, and $T_{1/2}$ is half-life of chemical. According to equation (1) and equation (2),

$$t = ln\frac{C_0}{C} \times \frac{T_{1/2}}{ln2} \qquad (3)$$

Therefore,

$$t_{adipose} = ln\frac{C_{0,adipose}}{C_{adipose}} \times \frac{T_{1/2,adipose}}{ln2} \qquad (4)$$

$$t_{liver} = ln\frac{C_{0,liver}}{C_{liver}} \times \frac{T_{1/2,liver}}{ln2} \qquad (5)$$

There were two reports on half-lives of BDE209 in birds; 13 days and 14 days of half-lives in plasma were observed in American kestrels (*Falco sparverius*) and European starlings (*Sturnus vulgaris*), respectively [93,94]. Half-life of BDE209 in liver is few days longer than that in plasma [90], therefore, 20 days of half-life in liver of birds was roughly estimated. Mean of L/A value (0.598), observed in northern fulmar except BDE209 of FAE2012-027, was used for calculation as L/A value in equilibrium state.

Half-life of BDE209 in adipose seemed to be few months or longer in mammals, in which the half-life of BDE209 in blood was between 8.5 and 13 days [91]. In FAE2012-027, assuming that BDE209 in adipose hasn't decreased after the end of BDE209 exposure, as a maximum estimation, BDE209 concentration in liver at the end of exposure period was calculated as 98.7 ng/g-lipid weight (165 ng/g-lipid weight in adipose (Table 4-3) × 0.598 (L/A ratio in equilibrium state)). When BDE209 concentration in liver of FAE2012-027 have been decreased from 98.7 ng/g-lipid weight to 4.91 ng/g-lipid weight (present concentration, Table 4-4), 87 days of elapsed time after the end of BDE209 exposure was estimated (C_{liver} = 4.9, $C_{0,\ liver}$ = 98.7, $T_{1/2,\ liver}$ = 20 days, were assigned to equation (5)).

Assuming that the half-life of BDE209 in adipose is twice that in liver (20days), 40 days of half-life was obtained as a minimum estimation. In FAE2012-027, when $t_{adipose} = t_{liver}$, $T_{1/2,adipose}$ = 40 days, $T_{1/2,liver}$ = 20 days, $C_{0,\ liver} = C_{0,\ adipose} \times 0.598$ (L/A ratio in equilibrium state), $C_{adipose}$ = 165 ng/g-lipid weight (Table 4-3), C_{liver} = 4.91 ng/g-lipid weight (Table 4-4), combining Equations (4) and

(5) gives rise to 173 days of the elapsed time after the end of BDE209 exposure. Therefore, the periods after quitting BDE209 exposure was estimated to be 87 –173 days in FAE2012-027.

BDE209 was also accumulated in the two other fulmars, FAE2012-015 and FAE2012-019. The elapsed time after the end of BDE209 exposure of these birds was estimated by the same way of the calculation above. When BDE209 in adipose hasn't decreased after the end of BDE209 exposure as a maximum estimation, the elapsed time after the end of BDE209 exposure in FAE2012-015 and FAE2012-019 were calculated as 7 days (C_{liver} = 71 ng/g-lipid weight (Table 4-4), $C_{0, liver}$ = 150 ng/g-lipid weight in adipose (Table 4-3) × 0.598 (L/A ratio in equilibrium state), $T_{1/2,liver}$ = 20 days), and 11 days (C_{liver} = 130 ng/g-lipid weight (Table 4-4), $C_{0, liver}$ = 316 ng/g-lipid weight in adipose (Table 4-3) × 0.598 (L/A ratio in equilibrium state), $T_{1/2,liver}$ = 20 days), respectively.

Assuming that the half-life of BDE209 in adipose to be 40 days as a minimum estimation, when $t_{adipose} = t_{liver}$, the elapsed time after the end of BDE209 exposure in FAE2012-015 and FAE2012-019 were calculated by combining Equations (4) and (5) as 13 days ($T_{1/2,adipose}$ = 40 days, $T_{1/2,liver}$ = 20 days, $C_{0, liver} = C_{0, adipose}$ × 0.598 (L/A ratio in equilibrium state), $C_{adipose}$ = 150 ng/g-lipid weight in adipose (Table 4-3), C_{liver} = 71 ng/g-lipid weight (Table 4-4)) and 22 days ($T_{1/2,adipose}$ = 40 days, $T_{1/2,liver}$ = 20 days, $C_{0, liver} = C_{0, adipose}$ × 0.598 (L/A ratio in equilibrium state), $C_{adipose}$ = 316 ng/g-lipid weight in adipose (Table 4-3), C_{liver} = 130 ng/g-lipid weight (Table 4-4)), respectively. Therefore, the periods after quitting BDE209 exposure was estimated to be 7 –13 days in FAE2012-015, and 11 –22 days in FAE2012-019.

As a result, it is indicated that FAE2012-027 excreted plastic with BDE209 approximately 3 –6 months before the death, and FAE2012-015 and FAE2012-019 excreted plastic with BDE209 within several weeks. The proportion of BDE209 concentration in liver to adipose can be useful for estimation of the time when they excrete plastic with additives. The information on the period in which BDE209 remains in the tissue may lead to reveal the frequency of exposure to plastic-derived BDE209 by comparing it with detection frequency of BDE209 in seabirds. Although the author made an estimation using data from previous studies [93,94], half-lives of PBDEs may differ among species. Further study on the difference of metabolic activity or systems, which is also suggested in the next section (5.1.2.), is needed for more precise estimation.

Estimation of the period elapsed after initiation of BDE209 exposure in OS10-008 (with the highest L/A ratio of BDE209) or the period after reaching equilibrium between tissues in the other birds (with L/A value in equilibrium state) were not able to calculated, because there have been no reports on the accumulation rate of BDE209 in tissues of avian species and difficult to estimate.

5.1.2. Debromination of BDE209 in seabird

In the tissues of five short-tailed shearwaters (OS10-008, WK10-019, WK10-023, WK05-018, and WK05-022) which contained BDE209, all three nona-brominated congeners together with BDE209 were detected. BDE207 was dominant over BDE206 and BDE208 in the tissues of each bird (Fig. 5-5f–m), but no such pattern was evident in the ingested plastics (Figs 5-5a–e and 5-6). The difference in isomeric compositions of nona-BDEs between the tissue and the ingested plastics can be explained by biological debromination of BDE209 at the *meta*-position to selectively generate BDE207.

In the tissues of three northern fulmars (FAE-2012-015, FAE-2012-019 and FAE-2012-027) and one white-chinned petrel (#3477), all of nona- to deca- brominated congeners were detected. Because all of them did not retained plastics with additive PBDEs, congener profiles of exposed PBDEs were unidentified. However, BDE207 was dominant over BDE206 and BDE208 in the tissues of each bird (Fig. 5-7), of which profile corresponded to that in short-tailed shearwater. A similar predominance of BDE207 in debromination of BDE209 among nona-isomers was found in other birds [93-97]. Debromination of BDE209 to BDE207 is generally observed in biological debromination and suggested to be catalyzed by deiodinases [98-100]. Deiodinases are membrane-bound enzymes in vertebrates, and they remove iodine atom from the meta-positions of thyroid hormones [101]. As other debromination processes such as photodegradation do not cause a predominance of BDE207 [102,103], this indicates biological debromination of BDE209 in the birds after absorption.

To compare the results of debromination, the proportion of BDE207 to BDE209 was calculated and shown in Figs 5-5 and 5-7. Among short-tailed shearwaters, WK10-019 showed a unique predominance of BDE207 over BDE209 in the liver and adipose (Fig. 5-5g, j). This composition may be related to the fact that the plastics in this bird did not contain higher-brominated congeners. Birds ingesting plastic with deca-BDE are exposed to PBDEs dominated by BDE209, and the accumulation of PBDEs and the debromination of BDE209 occur simultaneously in the tissues. This results in a higher concentration of BDE209 in the tissues. In contrast, WK10-019 excreted the plastic after the PBDEs accumulated in its tissues, and only debromination of BDE209 to BDE207 took place, resulting in the higher proportion of BDE207 than BDE209. In addition, among northern fulmars, the proportion of BDE207 / BDE209 in FAE-2012-027 (0.28) were higher than that of FAE-2012-015 and FAE-2012-019 (0.01 and 0.01, respectively). FAE-2012-027 has the lowest L/A ratio and it is expected that the congeners profile in liver of FAE-2012-027 was strongly affected by debromination and re-distribution from the other tissue.

In most of the birds, BDE207 / BDE209 ratios were higher in adipose tissue than in liver. Similar distribution was observed in rainbow trauts (*Oncorhynchus mykiss*) after exposure to BDE209, and the author expected that debromination products generated in liver were not accumulated in liver but transported to the other tissues through blood [89]. In addition, preferential accumulation of less brominated compounds is possible, which is due to large molecular sizes and/or high plasma protein binding affinities of BDE209 [89]. Slow distribution of BDE209 from blood to lipid-rich tissues was also reported in mouse [88]. In the birds in this study, likely explanation of differences of BDE207 / BDE209 ratios between liver and adipose is that debromination products are released into blood from liver, and/or preferential accumulation of lower-brominated congeners into adipose.

In comparison between the species, northern fulmar showed lower proportions of octa- and nona-brominated congeners to BDE209 in tissues. For example, BDE207 / BDE209 ratios in fulmars were ranged from 0.04 to 0.05 in adipose and from 0.01 to 0.28 in liver, and those in the other birds were ranged from 0.27 to 1.5 in adipose and from 0.37 to 1.0 in liver (Figs 5-5 and 5-7). The difference of proportion between avian species may indicate the difference of metabolisms and/or excretion systems of PBDE congeners among them. For example, in northern fulmar, slower debromination of BDE209 than the other species and/or faster excretion of octa- to nona- brominated congeners than debromination rate of BDE209 are expected.

Although some differences are indicated, there have been no reports on the difference of metabolism or excretion systems of PBDEs among avian species. More studies are necessary on them for each bird species, for more precise discussion on behavior of PBDEs in birds' tissue. Moreover, studies on metabolisms are important for assessment of toxic risks of plastic-derived PBDEs, because the metabolites of BDE209, such as debrominated congeners or further hydroxylated PBDEs generally have higher toxic activity than the native compound [104].

5.1.3. Toxicity of PBDEs in seabirds

PBDEs are generally known to cause disruption of thyroid hormone homeostasis and contribute to neurotoxicity in various organisms [105]. There are a limited number of studies examining PBDE toxicity in birds. Exposure to lower-brominated PBDEs can induce changes in thyroid, vitamin A, glutathione homeostasis, oxidative stress, growth rate, and pipping and hatching success in American kestrel [106-109]. European starlings exposed to BDE-209 via silastic implants accumulated it at the concentration of 250 ng /g-lipid weight in liver, and body mass was significantly lower in exposure group compared to control group [94]. In American kestrels, higher hepatic EROD activity in the exposed birds compared to control birds was observed after oral dose of 116,000 ng BDE209 per day, at the accumulated concentration of 34071 ng /g-lipid weight in liver and 8943 ng /g-lipid weight in adipose tissue.

In the present study, the highest concentrations of BDE209 were detected in FAE2012-019 at 130 ng /g-lipid weight in liver and 316 ng /g-lipid weight in adipose (Tables 4-3 and 4-4). The highest burden in plastics per a bird were detected in OS10-008 at 5080 ng /individual (Table 2-5). The rate of leaching of BDE209 by stomach oil in the leaching experiment was 15% during 15days. Therefore, the exposure amount per a day would be 50.8 (ng / kg-body weight / day) of BDE209 (15% of 5080 ng per 15 days per 0.5 kg of body weight [52]). The observed concentration in tissues or estimated exposure in the present study is lower than the levels in which toxic effect on avian species were observed [93,94]. However, higher exposure of PBDEs to seabirds would be occurred when they ingest more plastics with additive PBDEs, or they ingest plastics with higher concentration of PBDEs. For example, 10% of PBDEs, which is general concentration for application [18], in 0.01g of plastic (mean weight of a piece of ingested plastic) result in 1,000,000 ng of PBDEs burden in a bird.

After absorption, PBDEs are transferred and distributed in the tissues of seabirds. Even after excretion of plastics with PBDEs, gradual redistribution from tissues possibly cause for continuous toxic effect on seabirds. Exposure concentration from plastics, transfer and accumulation rate, and metabolic activity decide the behavior of PBDEs in seabirds. Understanding on them makes it possible to assess the toxic effect of plastic derived PBDEs. Studies on the concentration and frequency of additive PBDEs in marine plastics, and studies on their behavior in seabirds' tissue are needed.

5.2. Conclusion

In chapter 2 and 4, the author detected sporadic accumulation of higher-brominated congeners in tissue of seabird from three oceans, i.e., North Pacific Ocean, North Atlantic Ocean and South African waters, which is suggested to be derived from additives retained in ingested plastics. The obtained data suggest that chemical exposure to seabirds by plastic ingestion occurs all around the ocean. Moreover, some of the short-tailed shearwaters and northern fulmars accumulated much higher concentration of plastic-derived PBDEs than that derived from bio-accumulation. Plastic ingestion should be recognized as another important and major chemical source to seabirds.

In chapter 3, leaching experiments were conducted to examine the leaching of PBDEs from plastics to simulated digestive fluid. Trace amounts were leached into distilled water, seawater, and acidic pepsin solution. In contrast, over 20 times as much materials were leached into stomach oil, and over 50 times as much into fish oil (a major component of stomach oil). It is concluded that stomach oil, which is present in the digestive tract of birds in the order Procellariiformes, acts as an organic solvent, facilitating the leaching of hydrophobic chemicals from ingested plastics. Up to now, it has been thought that hydrophobic chemicals in plastics do not leach out to digestive fluid of marine organisms. The obtained data in this study indicate that in case of seabirds, especially in the order Procellariiformes, the leaching of chemicals from plastics could be greatly facilitated in their stomach by oils derived from prey.

Finally, it was revealed that the comparison of PBDEs concentration in liver to that in adipose tissue could give a new insight into the timing of exposure to plastic-derived PBDEs. Debromination of BDE209 occurs in seabirds' organs, and generates lower-brominated congeners. The information about exposure periods and metabolisms of chemicals is essential to assess the effects of plastic-derived chemicals on seabirds. The difference of metabolism or excretion rate among seabird species were suggested and need for studies on them were underlined.

As a conclusion, the present study revealed the way of exposure to plastic-derived additive chemicals as follows: plastics are transported in the marine environment with retention of additives, and after their ingestion by seabirds, additive chemicals in plastics are efficiently leached out to digestive fluid, and result in their accumulation in the tissue of seabirds.

5.3. Future direction

In this study, sporadic accumulation of higher-brominated PBDE congeners was observed in tissues of three seabird species in the order Procellariiformes, ascribing to transfer from ingested plastics. Leaching experiments suggested that stomach oil facilitates this transfer. The sporadic occurrence of BDE209 was reported in other studies, namely northern fulmar (*Fulmarus glacialis*)[57,79,110,111], but also in other species, i.e., glaucous gull (*Larus hyperboreus*) [112], and African penguin (*Spheniscus demersus*)[113]. This occurrence might also be due to plastic ingestion [12,75,114]. Plastic-mediated bioaccumulation of PBDEs in these species should be investigated. A wide range of seabird species, some of which have stomach oil, ingest plastics. Other organic digestive fluids such as bile may also facilitate leaching and bio-accumulation of additive-derived PBDEs from ingested plastics.

Marine plastics contain many chemicals. Comprehensive analysis of chemicals in plastics ingested by fulmars of Faroe islands revealed that they retain various additives and impurities other than PBDEs, such as Hexabromocyclododecane (HBCD) (30ppm), benzotriazole UV absorbers (1200ppm), benzophenone UV absorbers (570ppm), and styrene oligomers (260ppm) (Tanaka et al., unpublished data). Some of these chemicals are known to be endocrine disruptors [115-119]. These additives and impurities in plastics can be exposed to seabirds in the same way as PBDEs. This multiple contamination by plastic ingestion can result in toxic effects which have not been expected with the exposure through bio-concentration. Further investigations of plastic-mediated multiple exposures of additive chemicals are needed and their toxicological effects on seabirds should be studied in the future.

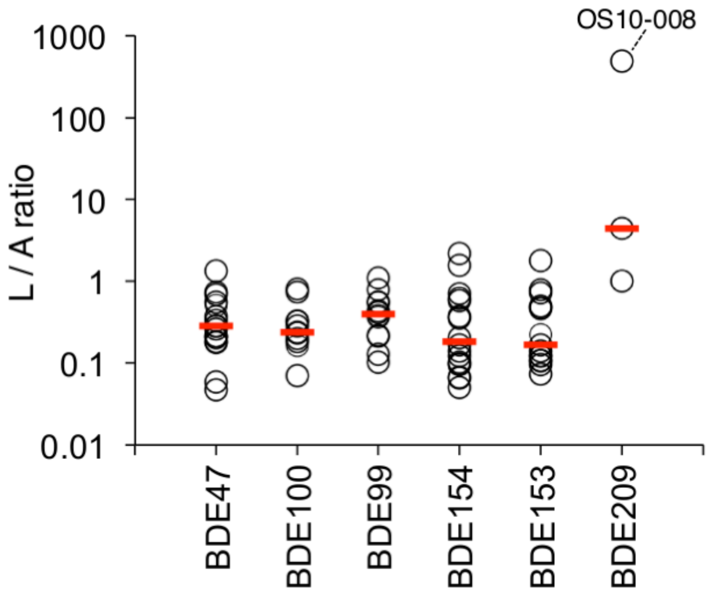


Figure 5-1. The ratio of PBDE concentration in liver to that in abdominal adipose (L/A ratio) of short-tailed shearwaters (*n*=18).

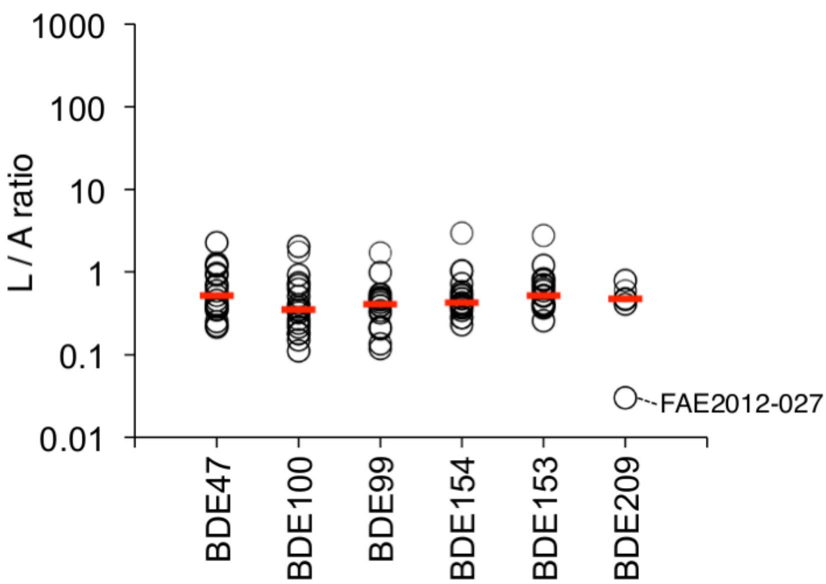


Figure 5-2. The ratio of PBDE concentration in liver to that in abdominal adipose (L/A ratio) of adult northern fulmars excluding FAE2011-687 (*n*=19).

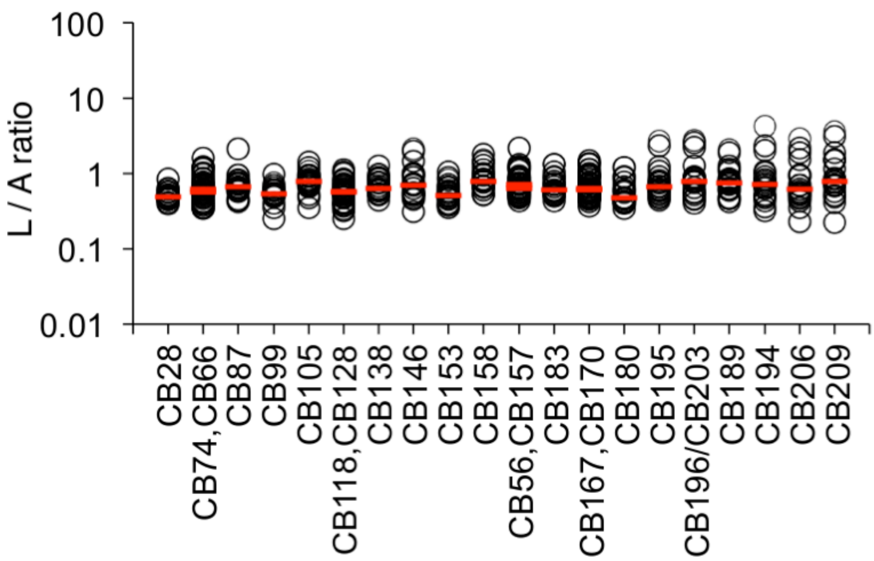


Figure 5-3. The ratio of PCB concentration in liver to that in abdominal adipose of adult northern fulmars excluding FAE2011-687 (n=19).

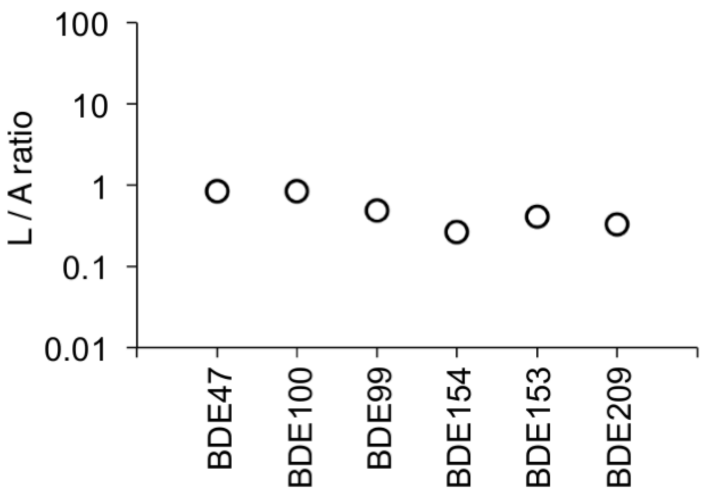


Figure 5-4. The ratio of PBDE concentration in liver to that in abdominal adipose of chicken from an electronic waste (e-waste) recycling area in southeast China (Qin et al., 2011[87]).

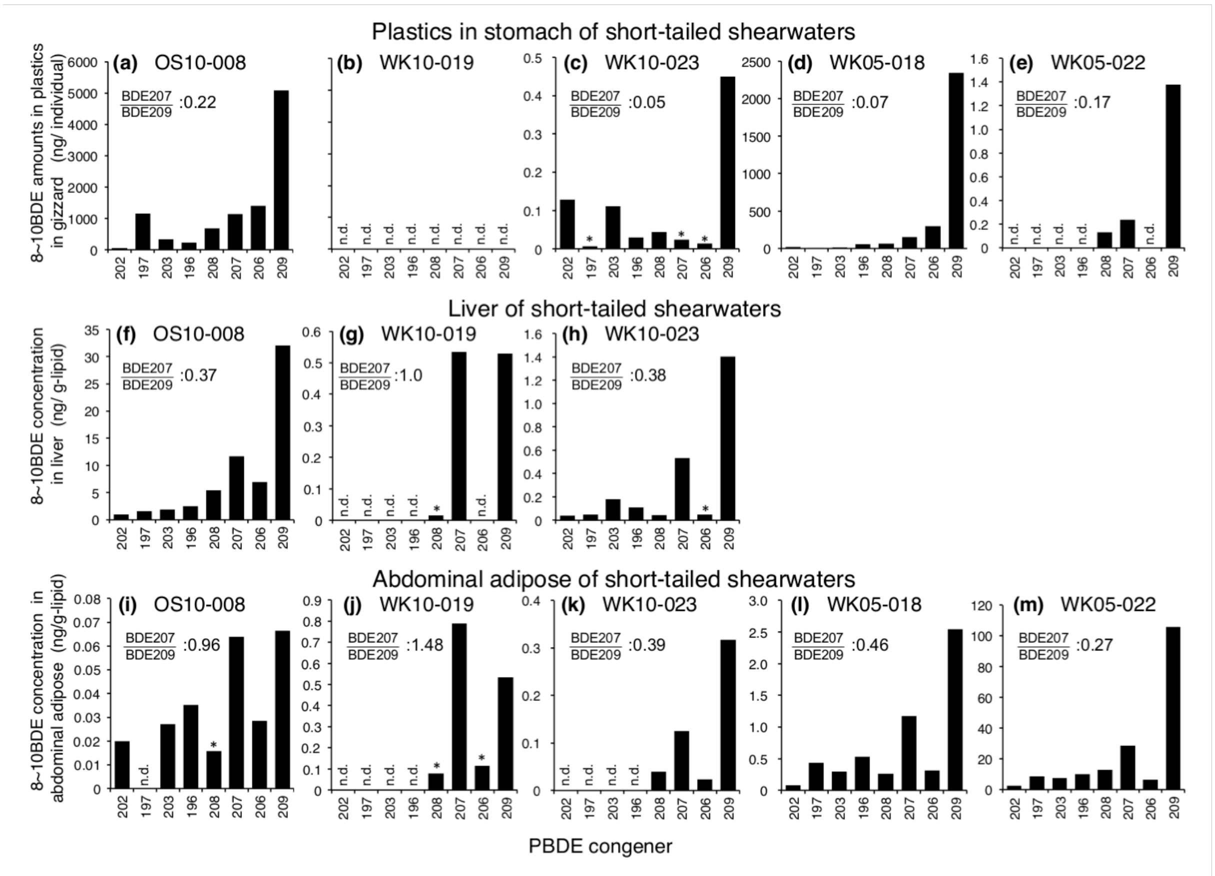


Figure 5-5. Congener profiles of octa- to deca-brominated PBDEs in (a−e) plastic in the stomach, (f−h) the liver, and (i−m) abdominal adipose of short-tailed shearwaters; n.d. indicates not detected. Asterisks indicate <LOQ.

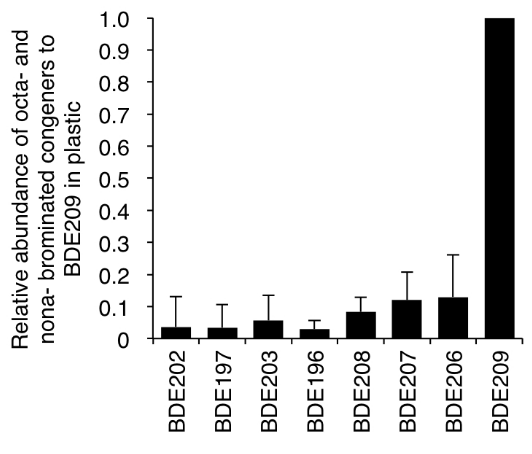


Figure 5-6. Abundance of octa- and nona-brominated congeners relative to BDE209 in ingested plastics which contained BDE209.

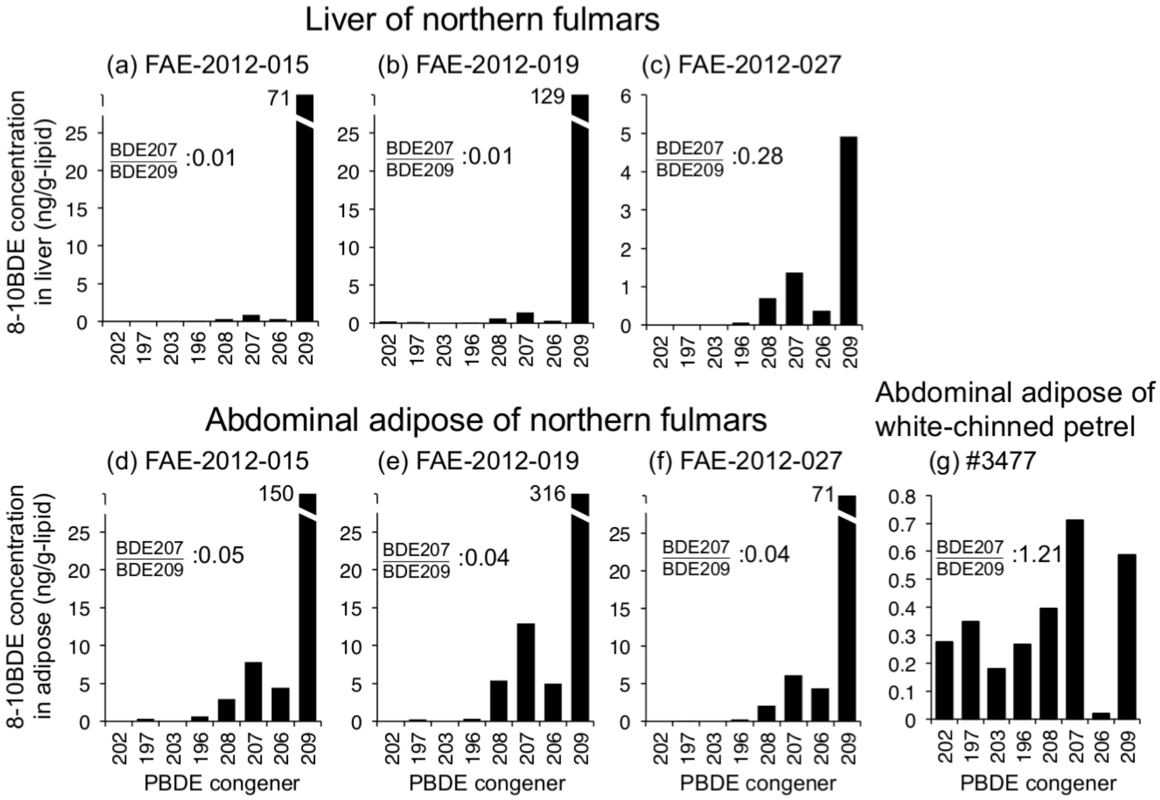


Figure 5-7. Congener profiles of octa- to deca-brominated PBDEs in (a−c) liver, (d−f) abdominal adipose of northern fulmars, and (g) abdominal adipose of white-chinned petrel.

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