Polybrominated Diphenyl Ethers in Marine Ecosystems of the American Continents: Foresight from Current Knowledge

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Abstract: Polybrominated diphenyl ethers (PBDEs) are a class of synthetic halogenated organic compounds used in commercial and household products, such as textiles, furniture, and electronics, to increase their flame ignition resistance and to meet fire safety standards. The demonstrated persistence, bioaccumulation, and toxic potential of these compounds in animals and in humans are of increasing concern. The oceans are considered global sinks for PBDEs, as higher levels are found in marine organisms than in terrestrial biota. For the past three decades, North America has dominated the world market demand for PBDEs, consuming 95% of the penta-BDE formulation. Accordingly, the PBDE concentrations in marine biota and people from North America are the highest in the world and are increasing. Despite recent restrictions on penta- and octa-BDE commercial formulations, penta-BDE containing products will remain a reservoir for PBDE release for years to come, and the deca-BDE formulation is still in high-volume use. In this paper, we review all available data on the occurrence and trends of PBDEs in the marine ecosystems (air, water, sediments, invertebrates, fish, seabirds, and marine mammals) of North and South America. We outline here our concerns about the potential future impacts of large existing stores of banned PBDEs in consumer products, and the vast and growing reservoirs of deca-BDE as well as new and naturally occurring brominated compounds on marine ecosystems.

Keywords: brominated flame retardants, PBDEs, novel BFRs, natural organobromine compounds, biomagnification, marine food webs

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1. INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a class of synthetic halogenated organic compounds used in numerous polymer-based commercial and household products, such as textiles, furniture, and electronics, to increase their flame ignition resistance and to meet fire safety standards /1/. PBDEs and their hydroxylated metabolites OH-PBDEs are structurally similar to thyroxine, and laboratory studies indicate that PBDE exposure can interfere with early neurodevelopment /2/. Since the 1970s, large amounts of PBDEs have been produced and used, resulting in the widespread contamination of the environment. Since the 1990s, PBDEs have been recognized as a global problem as they have been detected throughout the world in all matrices examined (air, water, soil, sediment, sludge, dust, mussels, fish, mammals, and human tissues) /3-6/. The less brominated congeners have been found in remote areas distant from their known use or production, e.g., the Arctic /4/ and the open oceans /7/. PBDEs are lipophilic and readily biomagnify in food webs; marine top predators tend to accumulate higher PBDE concentrations than do terrestrial biota /4/. The oceans are the global sinks for many hydrophobic persistent organic pollutants, such as polychlorinated biphenyls (PCBs) and PBDEs /8-10/. The finding of higher levels of PBDEs in organisms from deep seas compared with those from shallow waters suggests a vertical distribution of the compounds throughout the water column because of their association with organic particles that finally reach the sea bottom. As a result, sediment and biota from deeper waters act as a depot for persistent halogenated contaminants. The detection of PBDEs in diverse deep-sea organisms including sperm whales /7/, cephalapods /11/, and deep-sea fishes /9,10,12,13/ confirms that these contaminants have reached the deep ocean environment.

For the past three decades, North America has dominated the global market for PBDEs, consuming 95% of the penta-BDE formulation and 44% of the deca-BDE /14/. Accordingly, PBDE concentrations in biota and populations from North America are generally the highest in the world and increasing /3/. The distribution of PBDEs in the European, Asian, and Arctic environments has been the subject of numerous studies (reviewed in: /4-6,15, 16/). Despite a much greater usage of PBDEs in North America, until recently, comparatively little research has been done on the occurrence and fate of PBDEs in US or Canadian marine environments (reviewed in /3,17/), and few data exist on PBDEs in Central and South America. Compared with the northern hemisphere, the southern hemisphere is much less densely populated, and PBDE use there is presumed to be limited. Yet, PBDE exposure may be significant for marine biota inhabiting densely populated coastal areas because of the widespread usage of these compounds in several consumer products, including electronics /18/.

PBDEs are similar to PCBs with regard to structure, physicochemical properties, and the volume of global production and use. Based on the global production estimate of PBDEs in 2001 (67,400 tons; /14/) and the production duration of over 30 years, we can roughly estimate that more than 1 million tons of PBDEs have been produced globally, and the production of deca-BDE is still ongoing. For PCBs, the total global production from 1929 to the late 1970s was estimated at 1.2 million tons /19/. Tanabe /19/ predicted that the majority of the PCBs (66%) would remain stockpiled in equipment (transformers and capacitors) long after PCBs were banned from production. A similar scenario could be expected for PBDEs. Despite recent restrictions on certain PBDE mixtures, large amounts of polymer-based products, building materials, and plastics containing PBDEs are still in use and will be disposed of after their lifetimes, creating second-tier outdoor reservoirs (e.g., landfills, wastewater treatment plants, electronic waste recycling facilities or stockpiles of hazardous wastes) for the future dispersal of PBDEs to surface waters and the

Table 1. *Composition of commercial PBDE mixtures*

PBDE Mixtures	Congener composition (% of total)
Penta	24–38% tetra-BDEs, 50–60% penta-BDEs, 4–8% hexa-BDEs
Octa	10–12% hexa-BDEs, 44% heptaBDEs, 31–35% octa-BDEs, 10–11% nona-BDEs, <1% deca-BDEs
Deca	<3% nona-BDEs, 97–98% deca-BDE
BDE congeners[a]	**Substitution pattern**
BDE-47	2,2',4,4'-tetra-BDE
BDE-99	2,2',4,4',5-penta-BDE
BDE-153	2,2',4,4',5,5'-hexa-BDE
BDE-209	2,2',3,3',4,4',5,5',6,6'-deca-BDE

oceans. Moreover, deca-BDE remains the highest volume global PBDE in use today and the discharge of BDE-209 to aquatic and marine ecosystems has been increasing.

In this paper, we review all available data on the occurrence and trends of PBDEs in marine biota in the coastal areas of the American continents, with a particular focus on recent studies from the US and Canada and new data from Central and South America. PBDEs in the Canadian Arctic environment are not covered in this review, as they have received attention elsewhere /4,15/. We outline here our concerns about the potential future impacts of large existing stores of banned PBDEs in consumer products, and the vast and growing reservoirs of deca-BDE as well as new and naturally occurring brominated compounds on marine ecosystems. Whereas obvious difficulties exist in making comparisons among studies because of differences in the congeners measured, units used (for example, wet weight, lipid weight, dry weight), and tissues examined, we made every effort to ascertain that the comparisons and interpretations are logical and meaningful.

1.1 Chemical and Physical Properties and Uses

PBDEs are a class of brominated aromatic compounds consisting of two phenyl rings linked by an ether bond and having variable hydrogen to bromine substitutions (Figure 1).

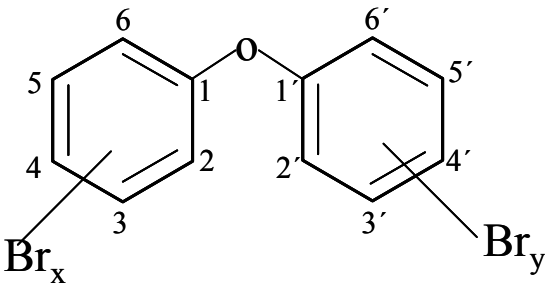

Fig. 1: Structural formula for PBDEs, where x + y = 1–10.

Mixtures of PBDEs are added to a variety of polymers such as polystyrene foams, high-impact polystyrene, and epoxy resins that are applied to consumer products, including electronic equipment (circuit boards, computers, monitors), textiles, commercial and residential construction materials, insulation, and mattresses and foam cushions in furniture, baby products, automobile and aircraft interiors to increase their fire resistance /1,20/. As additive flame retardants, PBDEs are not chemically bound (for example, covalently bound) but rather are physically blended with polymers, thus over

time, PBDEs have leached out of products, accumulated in indoor air and dust, and eventually entered the natural environment /21/.

PBDEs have been produced since the 1960s as three commercial mixtures (penta-, octa-, and deca-BDE) that vary in the degree of bromination (Table 1). Although PBDEs are structurally similar to PCBs and analogous to PCBs, 209 distinct PBDE isomers are possible, each commercial mixture contains only a limited number of congeners from each homologue group. The major constituents of penta-BDE, BDE-47, -99, and -100, with minor contributions from BDE-153, -154, and -85, are highly lipophilic, persistent and bioaccumulative, and subject to long-range transport /1,22/. The major congener found in octa-BDE is hepta-BDE-183; other constituents are nona-BDE-203 and several octa- and nona-BDEs, whereas deca-BDE contains primarily (97%) BDE-209 and low levels of nona-BDEs /1/.

Penta-BDE mixtures were mainly used in polyester and flexible polyurethane foam formulations in amounts that can result in the finished material being composed of up to 30% by weight of flame retardant. The main use of octa-BDE was in a variety of thermoplastic resins, in particular ABS (acrylonitrile-butadiene-styrene) plastic, which can contain up to 12% by weight octa-BDE. Deca-BDE, the most widely used PBDE globally, is added to various plastic polymers, such as polyvinyl chloride, polycarbonates, and high-impact polystyrene, as well as back coating for textiles (commercial furniture, automobile fabrics, and carpets).

Over the past three decades, North America has consumed more than half the global production of PBDEs and 95% of the penta-BDE product, the constituents of which are more persistent and bioaccumulative than those of the octa- or deca-BDEs /23/. Accordingly, PBDE burdens in people and biota from North America are the highest in the world; in contrast to Europe, North American burdens have been increasing over time /3/. In the US and Canada, the concentrations of PBDEs are increasing exponentially in fish, birds, and marine mammals, with concentrations in species from some areas doubling as rapidly as every 3-4 years /3,24-27/. In urbanized regions, the levels of tetra- to hexabrominated PBDEs are surpassing those of the PCBs and organochlorine pesticides as the top contaminant in air, water, and sewage sludge, and are rivaling PCBs in many fish and marine mammal populations /22/.

Largely because of environmental and public health concerns, the penta- and octa-BDE formulations were banned in Europe in 2004 and subsequently withdrawn from commerce in the US /28/. In May 2009, the Stockholm Convention included the penta- and octa-BDEs in the list of persistent organic pollutants (POPs) that by definition, are environmentally persistent, bioaccumulative, toxic to humans and the environment, and subject to long-range transport. Despite such regulations, the presence of large amounts of these compounds in long-lived, in-service, and discarded polymer products and electronics ensures their continuous release into the environment for decades. Moreover, deca-BDE remains a high-production volume flame retardant worldwide. Substantial releases of deca-BDE from industrial sources directly into the environment have been reported /21/. Further, concerns have been raised about the abiotic and biotic debromination of BDE-209, the major constituent of deca-BDE, to more bioaccumulative and potentially toxic congeners /22,29-32/. Comparatively little data have been reported regarding the environmental distribution of highly brominated PBDEs, either present in or derived from octa- and deca-BDE formulations. These hydrophobic congeners bind to particles and are concentrated in soils, sewage sludge, and indoor dusts, especially near urban areas /21/. BDE-209 is the dominant PBDE found in marine sediments /33/, thus, the deep oceans are vast reservoirs for ongoing inputs of deca-BDE and its debromination products to marine ecosystems.

In Europe and in some US states, restrictions have been imposed on the production and use of

deca-BDE /28/. In 2007, the state of Maine enacted legislation prohibiting the use of deca-BDE in mattresses and residential upholstered furniture sold within its borders and will extend the ban to electronics in 2010. Washington State prohibits the use of deca-BDE in mattresses and is considering a ban on its use in furniture and electronics. Similar legislation is pending in other US states and in Asian countries. Nevertheless, large amounts of deca-BDE have been released into the global environment and this flame retardant is still in high-volume use. The global market demand for deca-BDE in 2001 was approximately 56,100 tonnes /14/.

1.2 Sources of PBDEs

The main sources of PBDE input into the environment include emissions to air and surface waters from plants manufacturing the technical mixtures and facilities incorporating these flame retardants into polymers, as well as release during the life cycle of treated consumer products (degradation, recycling, disposal) /22/. For example, estimates are that up to 43 tonnes of penta-BDE per year have been released into the environment by volatilization from polyurethane foam used in various consumer products /34/. Electronic waste recycling facilities in Asia have recently been identified as major sources of PBDEs /35/. Airborne emissions of the less-brominated PBDE congeners (up to hexa-substituted) are expected to exist in both the vapor and the particulate phases and therefore be subject to long-range atmospheric transport processes /36/. Municipal sewage treatment plants (STPs) and landfills are additional point sources of PBDEs entering the environment /37/. High levels of PBDEs have been found in STP effluent and sludge and may be widely dispersed through the use of sludge as fertilizer on agricultural lands /22/. Because of their low vapor pressure and high hydrophobicity, PBDEs strongly adsorb to soils, sediments, and suspended organic material in the water column, facilitating their transfer to aquatic organisms. PBDEs enter coastal waters through municipal and industrial wastewater outfalls, landfill leachate, and atmospheric deposition from multiple sources /4/. Such compounds readily travel through the movement of air, water, particles, and biota and are surpassing PCBs as the top contaminant in many coastal ecosystems of North America /26/. Initially, the highest concentrations of PBDEs are often found near industrial or municipal outfalls /5/. PBDEs have been detected at low concentrations in North American air, water, and sediment, but at much higher levels in aquatic biota in areas receiving discharges /23/. Whereas sediment concentrations of PCBs are determined by environmental processes (for example, sediment accumulation, mixing rates), those of the more recently introduced PBDEs are strongly controlled by proximity to sources and use patterns. In particular, the entry of PBDEs into domestic dust facilitates the transport of these compounds to the coastal oceans by municipal wastewater systems /38/. Because PBDEs, especially the less environmentally mobile, highly brominated congeners (for example, BDE-209), persist in sediments, they are available to foraging by benthic organisms /9/ through which these compounds may re-enter marine food webs.

1.3 Toxic Effects

Detailed reviews of the toxicology of PBDEs, including mechanisms of toxicity, can be found in /20,39-42/. Laboratory studies conducted over the past two decades have shown that PBDEs have the potential to disrupt the endocrine system at multiple target sites in amphibians, birds, fish, mice, and rats resulting in effects on thyroid, ovarian, and androgen function. Many reports concern the disruption of thyroid hormone homeostasis by PBDEs (both in vivo and in vitro) and are characterized by a reduction in thyroxine (T_4) concentrations. Possible mechanisms include interference in the transport of T_4 via competitive binding to thyroid transport proteins (transthyretin, TTR) and thyroid hormone receptors, the induction

of thyroid hormone metabolic activity, and interference with the hypo-thalamus-pituitary-thyroid axis /43/. Recent studies have demonstrated effects of PBDEs on estrogen- and androgen-mediated processes as well, notably anti-androgenic effects shown both in vivo and in vitro (reviewed in /40,41/).

The greatest concern for the potential health effects of PBDEs arises from reports of toxicity resulting from developmental exposure (reviewed in /20,44/). In animal models, the toxic effects caused by prenatal or neonatal exposure to PBDEs include effects on liver enzymes /45/, endocrine disruption (altered thyroid hormone levels) /46/, reproductive damage /39,47,48/ neurotoxic effects /49,50/ and immunotoxicity /51/. In the offspring of mice prenatally exposed to BDE-209, immunotoxic effects include a dose-dependent increase in titers of respiratory synctytial virus in the lungs, increased levels of interferon-γ in the broncheoalveolar lavage fluids; at high doses (10,000 μg g^{-1} deca-BDE), an increased incidence of pneumonia was observed /52/. Experiments conducted by Eriksson and coworkers in mice developmentally exposed either to penta- or higher BDEs (49,50,53-56) and in rats exposed to BDE-209 /57,58/ during the period of brain growth spurt showed neurotoxic effects, including the impairment of spontaneous behavior, cholinergic transmitter susceptibility, and habituation capability. Deficits in learning and memory were observed to persist into adulthood and worsen with age. Similarly, a study by Rice et al 2007 /59/ demonstrated that dietary exposure of neonatal mice to deca-BDE resulted in developmental delays, changes in spontaneous locomotor activity, and a dose-related reduction in serum T_4 concentrations. The results of that study underline the critical role of thyroid hormones during brain development and suggest that the neurodevelopmental effect of PBDEs is related to perturbations in thyroid hormone homeostasis in the neonate /20/. The US Environmental Protection Agency (EPA) Integrated Risk Information System (IRIS) recently updated a toxicological review on deca-BDE and published an oral reference dose (RfD) of 0.007 mg/kg day based on neurobehavioral effects /60/.

In comparison with rodent studies, very few studies have examined the effects of PBDEs on wildlife. Mink (*Mustela vison*) are useful sentinels of ecosystem health and have been used as surrogate species for aquatic mammal toxicity studies /61/. Developmental immunotoxicity (increased splenic germinal center development and incidence of B-cell hyperplasia) was reported in ranch mink exposed to a high dose (10 μg g^{-1}) of a penta-BDE mixture DE-71 in the diet /51/. Significant body weight reductions were found in mink fed 5 and 10 μg PBDE/g /51/. The exposure doses of 5 and 10 μg PBDE g^{-1} diet for 8 weeks affected several immunological endpoints, including lymphocyte counts, in mink /51/. The exposure of mink to PBDEs through diet resulted in an accumulation in brain, suggesting that PBDEs can cross the blood-brain barrier /62/. A recent study assessed the effects of environmentally relevant doses (0, 0.1, 0.5, and 2.5 μg g^{-1} ww (wet weight) in feed) of DE-71 on the reproductive performance of mink and on the development of offspring exposed perinatally and post-weaning. A dietary concentration (2.5 μg g^{-1} DE-71) that had no effect on reproduction in rodents resulted in complete reproductive failure in mink, whereas whelping rates were not affected at all lower doses /62/. The developmental effects in offspring were evident in 33-week-old juveniles, which were more sensitive to such effects than were their respective dams. Juvenile thyroid hormone homeostasis was also far more sensitive compared with rodents, and at 0.5 μg g^{-1} DE-71, total triiodothyronine (T_3) was significantly decreased in all males and females, despite a compensatory increase of total thyroxine (T_4) in females /62/. The results of these studies indicate that mink are more sensitive than rodents to the effects of PBDEs.

Fish, predatory birds, and marine mammals may be susceptible to the endocrine-disrupting effects of PBDEs, although the threshold levels for effects in many wildlife species are not established (reviewed in /4/). Plasma thyroxine (T_4) levels

were significantly reduced in juvenile lake trout (*Salvelinus namaycush*) exposed to 13 PBDE congeners at levels somewhat higher than those found in the environment /63/. Repeated oral exposure to BDE-47 reduced sperm production in male fathead minnows (*Pimephales promelas*) /64/. Low-dose embryonic exposure of killifish (*Fundulus heteroclitis*) to DE-71 resulted in neurobehavioral effects and a subtle developmental asymmetry with respect to tail curvature direction, with a J-shaped dose-response curve suggestive of thyroid hormone disruption /65/. Similarly, the exposure of zebrafish (*Danio rerio*) embryos to high doses of BDE-47 resulted in developmental effects, including morphological, cardiac, and neural deficits that impaired later survivorship in the fish larvae /66/. The relevance of these effects observed in fish under laboratory conditions to wild fish inhabiting aquatic environments is unclear.

At high concentrations, PBDEs have been detected in such birds of prey as peregrine falcons *(Falco peregrinus)* and common kestrels (*Falco tinnunculus)*, and recent studies have demonstrated PBDE-related endocrine-disrupting and reproductive effects at environmentally relevant concentrations. In captive American kestrels *(Falco sparverius),* Fernie et al /67/ reported decreased plasma T_4 and vitamin A levels, as well as indications of oxidative stress in kestrels dosed with environmentally relevant levels of the penta-BDE mixture DE-71 in ovo and post-hatch. In a separate study, DE-71 exposure had a negative impact on the timing and frequency of courtship behaviors that are essential for successful reproduction /68/. In a follow-up experiment, exposure to DE-71 and the unintentional exposure to α-HBCD (hexabromocyclododecane) resulted in delayed egg laying, reduced egg size, eggshell thinning, and reduced fertility and reproductive success in kestrels /69/. Kestrels were exposed to DE-71 for 75 d. The low-exposure kestrel pairs were exposed to DE-71 concentrations of 0.3 μg g^{-1} diet per day and laid eggs with total PBDE concentrations of 289 ng g^{-1} ww; the high-exposure pairs were exposed to 1.6 μg g^{-1} diet per day of DE-71 and laid eggs with PBDE concentrations of 1130 ng g^{-1}ww. Control birds received safflower oil only and laid eggs with PBDE concentrations of 3.01 ng g^{-1} ww. Egg laying was delayed with increasing concentrations of BDEs-153, -154, -28, and -17, and multiple effects of DE-71 exposure on egg quality (egg size and mass) were observed in both treatment groups. Delayed egg laying, thinner eggshells, and poorer fledgling success were associated with BDE-153, the dominant congener recently found in kestrels and peregrine falcons /70/, whereas eggshell thinning was consistently inversely associated with the majority of congeners in the DE-71 mixture, especially BDE-153, and - 154, and -28, as well as α-HBCD. The authors of that study /69/ concluded that these changes in reproductive success of captive kestrels exposed to environmentally relevant PBDE concentrations, particularly the eggshell thinning associated with reduced hatching success, may partially explain the ongoing decline of American kestrels across North America. McKernan et al /71/ reported decreased pipping and hatching success in American kestrel embryos following air cell injection of DE-71 at concentrations of 10 and 20 μg g^{-1}, suggesting the lowest observable effect level may be as low as 1800 ng g^{-1} ww (based on the uptake rate).

Similarly, Johansson et al /70/ reported a negative relation between ΣPBDEs and reproductive success in peregrine falcons from Sweden. ΣPBDE concentrations in eggs were negatively related to the average number of young produced from individual breeding females over a 2-7 year period. Van den Steen et al /72/ observed negative effects of ΣPBDEs on reproductive performance in European starlings *(Sturnus vulgaris)* implanted with silastic tubes containing environmentally relevant concentrations of PBDEs. A recent field study /73/ reported a negative relation between reproductive performance and ΣPBDE concentrations in excess of 1000 ng g^{-1} ww in the eggs of wild ospreys *(Pandion haliaetus)* at two river locations in Oregon and Washington, suggesting

that concentrations of ~1000 ng g^{-1} ww may reduce reproductive performance in ospreys.

Marine mammals are long-lived top predators that accumulate high levels of PBDEs and other lipophilic POPs. Whereas threshold levels for the immune- and endocrine-disrupting effects of certain organochlorine contaminants (PCBs, dioxins) have been demonstrated in such species as harbor seals and mink (reviewed in /61/), the toxicological significance of adding PBDEs to an already existing complex mixture of toxicants has been difficult to evaluate. Similar to PCBs, relations between PBDEs and thyroid hormone alterations and immunotoxic effects in marine mammals have been reported. A study of free-ranging young United Kingdom (UK) grey seals reported a significant positive relation between concentrations of ΣPBDE in the blubber—range 61 to 1500 ng g^{-1}, lipid weight (lw) and circulating levels of thyroid hormones (T_3 and T_4) /74/. In a study of free-ranging mature harbor seals, elevated serum T_3 levels were significantly related to higher blubber concentrations of a contaminant mixture that included PBDEs, PCBs, and DDTs /75/. Another study reported an association between blubber concentrations of PBDEs (median 138 ng g^{-1} lw) and PCBs (median 3400 ng g^{-1} lw) and thymic atrophy and splenic depletion in stranded and by caught harbor porpoises from the North and Baltic Seas /76/, although emaciation was a confounding factor in some cases. The results of the two latter studies suggested that PCB-PBDE interactions may be involved in such effects. Similarly, the results of a study of infectious diseases in California sea otters co-exposed to PCBs and PBDEs were suggestive of possible synergistic interactions between these contaminant groups /77/. Although the role of the aryl hydrocarbon receptor (AhR) pathway in mediating the toxicity of PBDEs is unclear, , PBDEs and PCBs appear to bind the same cellular receptors /78/ and therefore, many PBDE-induced toxicities are likely to be similar to those of the non-coplanar (non-dioxin-like) PCBs. PCB-PBDE synergistic interactions have been observed in rodents. In rats exposed orally to PBDE-47 and the commercial PCB mixture Aroclor 1254, synergistic effects were observed on free T_4 in plasma and hepatic ethoxyresofurin (EROD) induction /79/. In a study by Eriksson et al /80/, developmental neurobehavioral defects were enhanced in neonatal mice co-exposed to a single oral dose of PCB-52 + PBDE-99, whereas a low dose of each compound alone had no effect on behavior. Thus, the effects of PCB-PBDE interactions may be more than just additive.

Despite the widespread and escalating PBDE exposure among North Americans, data on the potential adverse health effects of PBDEs in humans are limited. A study by Turyk et al /81/ reported that PBDEs were associated with increased T_4 levels and thyroglobulin antibodies in the blood of adult male consumers of Great Lakes sport fish. Effects were observed at PBDE levels comparable to those found in the general US population, and were independent of PCB exposure and sport fish consumption. Another recent study in China reported elevated serum levels of thyroid stimulating hormone (TSH) in workers exposed to PBDEs, either by working in or by living near an electronic waste-recycling center /82/. Elevated TSH levels could be a compensation for the reduction of circulating thyroid hormones and are indicative of stress on the thyroid system. An earlier study by Julander /83/, however, at an electronic recycling facility in Sweden did not find a significant change in thyroid hormone levels among PBDE-exposed workers. In a study of mother-boy pairs from Denmark and Finland, elevated PBDE levels in breast milk were correlated with cryptorchidism in the children /84/. The PBDE levels associated with cryptorchidism were also positively correlated with serum lutenizing hormone (LH) concentrations in the infants, which suggested a possible compensatory mechanism to achieve normal testosterone levels

and is consistent with the anti-androgenic effects of PBDEs observed in experimental animals. Elevated levels of PBDEs in the breast milk of pregnant Taiwanese women were significantly associated with an adverse birth outcome regarding weight, length, and chest circumference of their infants /85/. In both latter studies, the effects were observed at levels much lower than average PBDE levels in the adult US population. A pilot study conducted by Japanese researchers reported that elevated blood levels of BDE-153 were correlated with decreased sperm count and decreased testes size /86/. The importance of house dust as a major exposure route for PBDEs in humans has been highlighted /87-89/, and a recent study reported a relation between altered hormone levels in American men and PBDE levels in house dust /90/. The findings included significant inverse associations between PBDEs in house dust and serum concentrations of the free androgen index, LH, and follicle-stimulating hormone (FSH) and positive associations between PBDEs and sex hormone binding globulin (SHBG) and free T_4. The positive relation between PBDEs and free T_4 is consistent with the findings of Turyk et al /81/ in adult male fish consumers, but inconsistent with developmental exposure studies in animals that have consistently reported a decrease in T_4 related to PBDE exposure /40,41/. The authors suggest that important differences exist in PBDE effects on thyroid signaling and T_4 levels in developmentally exposed animals compared with adult human exposure. The relations between PBDEs and enhanced thyroid hormone levels reported in studies of marine mammals, however, suggest that in addition to species and maturation differences, possible interactions among contaminant mixtures may be involved in thyroid alterations. In the US, congenital hypothyroidism has been on the rise for the past 20 years. Studies assessing the relation between in utero exposure to compounds that alter thyroid hormone levels (such as PBDEs) and congenital hypothyroidism are needed.

The cancer potential of PBDEs has not been adequately addressed in animals or humans, as few studies have been conducted. A study by Hardell et al /91/ reported an association between BDE-47 concentrations and an increased risk for non-Hodgkin's lymphoma (NHL). In the highest risk, highest exposure group, BDE-47 was also significantly correlated with elevated titers to Epstein Barr IgG, a human herpes virus that has been associated with certain subgroups of NHL. In a long-term feeding study, a significant increase in the incidence of follicular-cell hyperplasia of the thyroid gland was observed in mice (male and female) exposed to high doses of BDE-209 /92/. During the past several decades, the incidence of thyroid cancer has been increasing in the US, especially among women; one hypothesis is that part of the observed increase in thyroid cancer rates may be related to the increasing population exposure to PBDEs and other thyroid hormone disrupting compounds /43/.

1.4 Human Exposure

The reported concentrations of PBDEs in human tissues from the US, Canada, Mexico, Nicaragua, and Brazil are shown in Table 2. The concentrations of PBDEs in the North American general population are much higher (10-100 fold) than the concentrations reported for populations in other parts of the world /3,87,88,93-95/. Furthermore, the concentrations of PBDEs in body tissues of US children were found to be 2 to 5-fold higher than the concentrations in their parents /96/. The highest concentration of PBDEs (9630 ng g^{-1}, lipid wt) in human adipose fat was reported for a sample collected from New York /93/. Similarly, elevated (relative to the general US population) concentrations of PBDEs in serum samples have been reported from California /97/. In Californian women, elevated PBDE concentrations were observed in breast adipose tissue and milk /98,99/; the median concentrations of BDE-47 in milk were

Table 2. *Concentrations of individual PBDE congeners (ng g^{-1} lw) and mean and range of ΣPBDEs in humans. Reference numbers refer to the corresponding references in the manuscript.*

Location		Tissue	Year	Mean Age	Sex	N	#	BDE Congeners										ΣPBDEs			Ref
Country	State						Cong.	28	47	85	99	100	138	153	154	183	209	Mean	Min	Max	#
United States	California	breast adipose	1996-98	47	F	32	1	nm	29	nm	nm	nm	nm	nm	nm	nm	nm	29	5.2	196	99
	California	breast adipose	1996-99	47	F	23	5	nm	18	nm	6.6	3.2	nm	4.1	5.4	nm	nm	41	17.2	462	98
	New York	adipose	2003-04	32	F/M	52	11	3.3	132	6.9	74	68	nm	92	8.3	nm	nm	398	17	9630	93
	Indiana	cord serum	2001			12	6	nm	25	nm	7.1	4.1	nm	4.4	0.7	0	nm	39	14	460	284
	Maryland	cord serum	2004-05			297	8	0.9	14	1.1	4.3	2.6	nm	2.6	0.9	0.9	nm	27			285
	All US residents	serum	2003-04	12-19	F/M		10	1.3	28	nr	6.9	5.2	nr	8.1	nr	nr	nm				87
	All US residents	serum	2003-04	20-39	F/M		10	1.1	22	nr	5.2	4.4	nr	6.6	nr	nr	nm				87
	All US residents	serum	2003-04	40-59	F/M		10	1.1	18	nr	nd	3.3	nr	4.6	nr	nr	nm				87
	All US residents	serum	2003-04	60+	F/M		10	1.4	20	nr	nd	3.6	nr	5.1	nr	nr	nm				87
	Illinois	serum	1988			12	13	nm	2.7	nm	1	0.53	nm	0.84	nm	0.33	<1	7.1	0.5	134	286
	Tennessee	serum	1985-89			9	6	nm	5.4	<0.5	<2	0.81	nm	0.84	<0.5	nm	nm	9.6	4.6	74	287
	Tennessee	serum	1990-94			14	6	nm	28	0.61	10	4	nm	1.6	<0.5	nm	nm	48	7.5	86	287
	Tennessee/ Washington	serum	1995-99			10	6	nm	46	0.78	13	6.7	nm	4.2	0.88	nm	nm	71	42	120	287
	Tennessee/ Washington	serum	2000-02			7	6	nm	34	0.7	11	5.9	nm	7.3	0.95	nm	nm	61	47	160	287
	Indiana	serum	2001	26	F	12	6	nm	28	nm	5.7	4.2	nm	2.9	0.3	0	nm	37	15	580	284
	California	serum	1959-67	27	F	420	1	nm	nd	nm	nm	nm	nm	nm	nm	nm	nm	nd	nd	nd	99
	California	serum	1997-99	31	F	50	1	nm	51	nm	nm	nm	nm	nm	nm	nm	nm	51	<10	511	99
	California	serum	2003-04	12-60+	F/M	276	6	2.1	36	nm	7.4	6	nm	6.8	0.8	nm	nm	62			97
	California	serum	2004	35	M	1	39	nr	28	nr	4.7	9.7	nr	23	3.5	nr	9.3	69			96
	California	serum	2004	36	F	1	39	nr	58	nr	14	19	nr	35	4.4	nr	7.3	130			96
	California	serum	2004	5	F	1	39	nr	112	nr	32	36	nr	59	5.7	nr	51	245			96
	California	serum	2004	1.5	M	1	39	nr	213	nr	42	77	nr	116	12	nr	93	461			96
	Salinas Valley, CA	plasma	1999-01	26	F	24	7	nm	11	0.3	2.9	1.8	nm	1.5	0.3	<0.1	nm	21	5.3	320	102

Location		Tissue	Year	Mean Age	Sex	N	#	BDE Congeners										ΣPBDEs			Ref
Country	State						Cong.	28	47	85	99	100	138	153	154	183	209	Mean	Min	Max	#
United States	Mississippi	whole blood	2003	49	F/M	29	13	0.71	15	0.19	3.8	3.3	0.06	3.6	0.38	0.19	1.5	31	4.7	362	105
	New York	whole blood	2003	42	F/M	10	13	0.34	11	nm	2.9	2.4	0.07	3.5	0.31	0.22	nd	25	4.6	135	105
	Texas	milk	2001-04	29	F	47	13	1.2	18	0.41	5.7	2.9	0.09	2	0.22	0.07	nm	34	6.2	418	288
	Pacific Northwest	milk	2003	30.5	F	40	12	1.7	28	0.57	5.4	5.3	nm	4.8	0.4	0.2	0.43	50	6.34	321	106
	Massachusetts	milk	2004-05	19 - 41	F	46	12	0.9	14	0.3	2.4	2.4	0.03	3	0.2	0.1	nd	30	4.3	264	124
	Massachusetts	milk	2004	35	F	38	17	0.47	7.7	<0.5	1.5	<0.5	<0.03	1.1	0.05	4.1	nd	20	0.06	1910	289
	Texas	liver	2004	fetus		11	13	0.55	11	0.14	6.1	1.9	0.02	1.1	0.29	0.28	nd	23	4	99	290
	Michigan	gestational membranes	2007-08			5	11	0.14	1.2	0.25	0.98	0.56	nd	0.61	0.36	nd	nd	5.6	3.1	9.5	291
Canada	Ontario /Quebec	milk	1992		F	10	6	0.13	1.8	nr	0.65	0.21	nr	0.29	nr	0.13	nr	3.1	0.79	28.5	292
	Quebec City	plasma	2003-04	48-76	F	110	4	nm	39	nm	12	6.8	nm	5.4	nm	nm	nm	64	0.81	3.1	293
Mexico	Juarez	serum	2006	6-13	F/M	43											nd	4.8			94
	San Luis Potosi	serum	2006	6-13	F/M	16											nd	7.3			94
	Milpillas	serum	2006	6-13	F/M	52											nd	8.6			94
	El Refugio	serum	2006	6-13	F/M	15											nd	15.7			94
	San Juan Tilapa	serum	2006	6-13	F/M	20											nd	3.7			94
	Chihuahua	serum	2006	6-13	F/M	27											nd	2.7			94
Nicaragua	Managua Waste Disposal Site[a]	serum	2002	14	F/M	11	10	6.5	219	11	119	40	nm	20	12	1.7	8.9	438			101
	Managua Waste Disposal Site[b]	serum	2002	14	F/M	23	10	0.69	28	1	10	9	nm	9.7	5.9	1.2	3.6	69			101
	Urban Managua	serum	2002	14	F/M	37	10	0.31	9.6	0.42	4.0	2.5	nm	2.2	1.5	1.4	12	34			101
	Rural Managua	serum	2002	15-20	F	8	10	0.25	8.3	0.28	4.2	2.1	nm	1.2	1.2	0.8	3.9	22			101
	Urban Managua	serum	2002	18-25	F	3	10	1.5	42	0.96	7.9	9.0	nm	7.7	1.3	0.57	3.9	75			101
	Urban Managua	serum	2002	42-44	F	4	10	0.77	33	1.5	15	6.2	nm	3.5	1.8	0.35	2.9	65			101
Brazil	Porto Alegre	breast adipose	2004-05	56	F	25	11	0.08	3.8	0.83	9.1	0.75	0.35	0.54	0.58	0.64	nm	6.6	0.19	132	95

[a] Children working and living at the waste disposal site (WDS)
[b] Children working at the WDS and living in the surrounding city
nd = not detected, nr = not reported, nm = not measured

up to 25 times higher than those reported from Europe /99/. The higher levels in Californians are presumably due to the state's stringent fire regulations and high BFR usage.

Few studies have reported concentrations of PBDEs in populations from Central and South America. In comparison with PBDE concentrations in human samples from the US and Canada, the PBDE concentrations in adipose samples from Brazil were 3 to 100-fold lower /95/. A recent study /94/ reported serum PBDE levels as high as 43.4 ng g^{-1} lw in children living in an urban/ industrialized city (Juarez), Mexico. The Mexican children had two-fold higher PBDE levels than children living in Europe /100/ but almost 20-fold lower levels than children from the US /96/. In contrast, PBDE concentrations in the serum of children working and living at a waste disposal site in Nicaragua were among the highest ever reported /101/. Serum BDE-47 levels (mean 219 ng g^{-1} lw) in these children exceeded those in US children /96/ and were an order of magnitude higher than the median concentration for BDE-47 in the adult US population /87/. Moreover, levels of PBDEs observed in referent Nicaraguan children (who did not work at the site) and young and middle-aged women from a nearby urban area were comparable to current levels reported in the US /3,87,96,102-106/. These observations highlight the need for a worldwide exposure assessment of PBDEs, not just in the developed countries /101/.

The sources of human exposure to PBDEs have been associated with the indoor environment and diet /88,97,107-114/. Nevertheless, in contrast to the pattern reported for PCBs /115,116/, diet contributes only a small proportion to PBDE exposure /109,117,118/. PBDE concentrations in foodstuffs from the US were comparable to those reported for several European countries and Japan /110,119-123/. Sources other than the diet, including house dust, were found to be important contributors to human exposure to PBDEs in the US and Canada /88,96,97,108,111-113/. Elevated concentrations (on the order of several tens to hundreds of μg g^{-1}) of PBDEs in house dust samples from the US and Canada coupled with a daily dust ingestion rate of 20-200 mg by the US general population /107/ suggest the significance of dust as a source of human exposure /113,124/. Recent studies have documented the occurrence of PBDEs at several tens of μg g^{-1} concentrations in dust from US automobiles /125/ and garages /126/, hundreds to thousands of pg/m^3 concentrations of PBDEs in indoor air /89/, and tens to hundreds of pg/m^3 concentrations in automobile air /127/. A significant positive correlation between PBDE concentrations in house dust and breast milk has been shown /124/.

Human exposure to PBDEs, with particular focus on external exposure routes (for example, dust, diet, and air) and the resulting internal exposure (breast milk and blood) has been reviewed recently /128/. Overall, populations in the US and Canada are exposed to higher levels of PBDEs than the general populations in the rest of the world. Lorber /88/ reported that exposure to PBDEs in house dust accounts for 80% to 90% of the overall estimated PBDE intake in Americans. Infants in the US are exposed at higher levels than adults through the ingestion of breast milk /89,96/. In Nicaraguan children working at a waste disposal site, PBDE exposure through the inhalation and ingestion of contaminated dust was of greater magnitude than dietary exposure /101/. Studies suggest that PBDE exposure via dust intake and inhalation of air can contribute to adverse health effects. Despite the high proportion of deca-BDE in house dust and that dust is a major contributor to PBDE exposure in humans, the proportion of BDE-209 in human tissues is minor. This paradox suggests a congener-specific contribution from various sources to the human exposure of PBDEs. The significance of diet as a major contributor to the less brominated congeners like BDE-47 cannot be ruled out. Further studies are needed to evaluate the bioavailability, transformation, and fate of BDE-209 in the indoor environment.

2. PBDE IN MARINE ECOSYTEMS

2.1 Air and Water

PBDEs have been detected in ambient air and indoor environments /129-131/. Despite its low volatility and strong sorption to particulates, BDE-209 was found in samples collected from remote locations, suggesting its potential for long-range transport /24/. Although several studies have reported the occurrence of PBDEs in air samples from North American continental locations (130, 132-135), reports of PBDEs in coastal marine environments are scarce. In 2003, the reported concentrations of PBDEs in air from the North Pacific Ocean ranged from 1.4 to 36.9 pg/m^3 (mean: 12.8; /35/), (Table 3), which are similar to those reported for air samples from the Arctic Ocean, Norwegian coast, and the Baltic Sea (ranging from 1.1 to 17 pg/m^3, /35,129,136/), Eagle Harbor, Lake Superior (5.5 pg/m^3, /132/) and the Indian Ocean (1.5-15.6 pg/m^3; /137/). The reported concentrations of PBDEs in coastal and oceanic air samples are approximately 10-fold lower than those reported for North American urban air /130, 133-135/. Although little information exists for Central and South America, it is noteworthy that the highest levels of PBDEs in air over the North American continent were detected in samples from the small tropical town of Belmopan, Belize /135/, indicating that PBDEs are not just a problem for industrialized, densely populated countries. Higher rates of the open burning of household wastes in developing countries than in the US or Canada have been reported /135/ and might be the reason for the high PBDE levels found in the air samples from Belize.

Oros et al /138/ determined the concentrations of PBDEs in water, sediment, and bivalves from San Francisco Bay in 2002 (Table 3). The levels of PBDEs in water samples ranged from 3 to 513 pg L^{-1} (mean: 103 pg L^{-1}). The highest concentrations were found in the urbanized southern part of the bay. PBDE congeners -47 and -209 were the predominant compounds in water, collectively accounting for 42% and 44%, respectively, of the total PBDE concentrations. More than 78% of the PBDEs in water were associated with the particulate phase. The concentrations of PBDE congeners -47 and -209 in water samples collected during 2002-2006 from San Francisco Bay were in the ranges of 16-337 (mean: 55) and 12-533 (mean: 29) pg L^{-1}, respectively /139/, which were much higher than those reported for the Scheldt estuary and the North Sea along the Dutch coast (0.1-5.6 pg L^{-1} /140/) and similar to those reported for Lake Michigan (31-158 pg L^{-1} /22/). Discharge of STP effluents is a major source of PBDEs in the coastal marine environment /141/. No study has reported the occurrence of PBDEs in air or seawater from South America.

2.2 Sediments and Invertebrates

Surface sediments collected from the San Francisco Bay in 2002 contained PBDE levels ranging from below detection limit to 212 ng g^{-1}, dry wt (mean: 11.9 ng g^{-1} /138/) (Table 3). With the exception of two highly contaminated locations with concentrations of 212 and 69 ng g^{-1} dry wt, PBDE levels in sediments from other locations in San Francisco Bay were below 20 ng g^{-1}, dry wt. BDE-47 was the predominant congener in San Francisco Bay sediment, whereas BDE-209 was not detected. On the other hand, BDE-209 was the dominant congener in sediments from several lakes, rivers, and coastal areas /142-147/. Deca-BDE accounted for 79% to 98% of the total PBDE concentrations in sediments from the Great Lakes /142/, 89% of the total PBDEs in sediments from the Niagara River /144/, and 79% to 90% of the total PBDEs in sediments from the Saginaw River watershed /147/. The concentrations of PBDE (all congeners except BDE-209) and BDE-209 in Lake Michigan surficial sediment were 2.6 and 315 ng g^{-1} dry wt, respectively /148/, which were an order of magnitude higher than the concentrations found in Lake Superior /142/. In general, concentrations of BDE-209 in surficial sediment from the Great

Table 3. *Concentrations of individual PBDE congeners and mean and range of ΣPBDEs in air, seawater, marine sediments, and invertebrates. Reference numbers refer to the corresponding references in the manuscript.*

Matrix/ Species	Units[a]/ Tissue	Location		Year	# Cong.	N	PBDE								Sum			Ref
		Country	State				47	99	100	153	154	155	183	209	Mean	Min	Max	#
Air	pg/m3	US	North Pacific Ocean	2003	11	10	6	2.24	0.56	0.1	0.03	nr	0.04	3.6	12.8	1.4	36.9	35
	pg/m3	US	Lake Michigan	2002-04	26	35	6.2	5.1	1.1	nr	nr	nm	nr	1.5	16	1.2	61	133
	pg/m3	US	Chicago, Illinois	2002-04	26	28	17	7.4	1.8	nr	nr	nm	nr	60	100	13	980	133
	pg/m3	US	Bloomington, Indiana	2002-04	26	38	7.0	5.1	1.0	nr	nr	nm	nr	2.2	19	6.4	44	133
	pg/m3	US	Rohwer, Arkansas	2002-04	26	30	9.2	5.4	1.2	nr	nr	nm	nr	9.0	30	2.7	165	133
	pg/m3	US	Cocodrie, Louisiana	2002-04	26	26	6.9	3.0	0.73	nr	nr	nm	nr	2.8	16	5	42	133
Air	ng/PAS/yr	Canada	Bonavista, Newfoundland	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	20			135
	ng/PAS/yr	Canada	Daniels Harbor, Newfoundland	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	nd			135
	ng/PAS/yr	Canada	East Point, Pr. Edward Island	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	4.1			135
	ng/PAS/yr	Canada	Sable Island	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	0.2			135
	ng/PAS/yr	Canada	Kejimkujik, Nova Scotia	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	13			135
	ng/PAS/yr	Canada	Saturna Island, BC	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	0.1			135
	ng/PAS/yr	Canada	Cape Beale, BC	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	6.5			135
	ng/PAS/yr	US	Solomon, Maryland	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	0.6			135
	ng/PAS/yr	US	Wilmington, North Carolina	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	0.1			135
	ng/PAS/yr	US	Turkey Point, Florida	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	6.1			135
	ng/PAS/yr	Mexico	Chetumal	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	0.4			135
	ng/PAS/yr	Mexico	Tapachula	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	4.9			135
	ng/PAS/yr	Belize	Belmopan	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	25			135
	ng/PAS/yr	Costa Rica	Monteverde	2000-01	5	1	nr	nr	nr	nr	nr	nm	nm	nm	nd			135
Seawater	pg/L	US	California-San Francisco Bay	2002	22	33	43	28	7.2	3.9	2.9	nm	4.4	45	103	0.2	513	138
	pg/L	US	California-San Francisco Bay	2002-06	40		180	nr	nr	nr	nr	nr	nr	270				139
Sediments	ng/g dry wt	Canada	BC-Clover Point at outflow	2006	34	1	1.3	1.1	0.27	0.11	0.09	0.01	0.03	2.6	6.2			155
	ng/g dry wt	Canada	BC-Clover Point at 100m	2006	34	4	0.10	0.07	0.02	0.01	0.01	0.001	0.003	0.6	1.0	0.68	1.3	155
	ng/g dry wt	Canada	BC-Clover Point at 200m	2006	34	3	0.16	0.12	0.03	0.01	0.01	0.001	0.004	0.7	1.2	0.37	2.1	155
	ng/g dry wt	Canada	BC-Clover Point at 400m	2006	34	4	0.07	0.05	0.02	0.01	0.01	0.001	0.003	0.7	0.91	0.51	1.4	155
	ng/g dry wt	Canada	BC-Clover Point at 800m	2006	34	2	0.07	0.05	0.02	0.01	0.01	0.001	0.002	0.6	0.84	0.81	0.87	155
	ng/g dry wt	Canada	BC-Clover Point at reference	2006	34	3	0.03	0.02	0.01	0.003	0.003	0	0.001	0.3	0.39	0.33	0.43	155
	ng/g dry wt	Canada	Strait of Georgia	2003-04	40	7	nr	nr	nr	nr	nr	nr	nr	nr	2.54	0.27	12.6	38
	ng/g dry wt	US	California-San Francisco Bay	2002	22	48	11	4.9	nd	nd	nd	nm	nd	nd	11.9	<0.2	212	138
	ng/g	US	California-San Francisco Bay	2002-06	40		1.9	nr	nr	nr	nr	nr	nr	9.7				139
	ng/g dry wt	US	Alabama	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	0.2	0	0.6	150

Matrix/ Species	Units[a]/ Tissue	Location/ Country	State	Year	# Cong.	N	PBDE 47	99	100	153	154	155	183	209	Sum Mean	Min	Max	Ref #
Sediments	ng/g dry wt	US	California	2004-07	38	15	nr	nr	nr	nr	nr	nr	nr	nm	7.1	0	88	150
	ng/g dry wt	US	Florida	2004-07	38	23	nr	nr	nr	nr	nr	nr	nr	nm	0.7	0	6.7	150
	ng/g dry wt	US	Georgia	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	0.03	0	0.1	150
	ng/g dry wt	US	Louisiana	2004-07	38	19	nr	nr	nr	nr	nr	nr	nr	nm	0.6	0	3.9	150
	ng/g dry wt	US	Maine	2004-07	38	1	nr	nr	nr	nr	nr	nr	nr	nm	0			150
	ng/g dry wt	US	Maryland	2004-07	38	6	nr	nr	nr	nr	nr	nr	nr	nm	0.1	0	0.3	150
	ng/g dry wt	US	Massachusetts	2004-07	38	8	nr	nr	nr	nr	nr	nr	nr	nm	2.5	0	14.4	150
	ng/g dry wt	US	Mississippi	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	2	0	5.6	150
	ng/g dry wt	US	New Jersey	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	2.2	0.1	4.9	150
	ng/g dry wt	US	New York	2004-07	38	12	nr	nr	nr	nr	nr	nr	nr	nm	14.4	2.9	41.3	150
	ng/g dry wt	US	North Carolina	2004-07	38	6	nr	nr	nr	nr	nr	nr	nr	nm	1.3	0	5.4	150
	ng/g dry wt	US	Oregon	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	0.2	0	0.7	150
	ng/g dry wt	US	Puerto Rico	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	1.5	0	4.4	150
	ng/g dry wt	US	Rhode Island	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	4.9	0	12.2	150
	ng/g dry wt	US	South Carolina	2004-07	38	1	nr	nr	nr	nr	nr	nr	nr	nm	0			150
	ng/g dry wt	US	Texas	2004-07	38	24	nr	nr	nr	nr	nr	nr	nr	nm	0.8	0	14.5	150
	ng/g dry wt	US	Virginia	2004-07	38	5	nr	nr	nr	nr	nr	nr	nr	nm	0.1	0	0.4	150
	ng/g dry wt	US	Washington	2004-07	38	15	nr	nr	nr	nr	nr	nr	nr	nm	0.6	0	2.0	150
Maldane worm	Whole body	Canada	St. Lawrence Estuary	1999-00	10	3	31	38	12	nr	nr	nr	nr	nm	93			157
Nereis worm	Whole body	Canada	St. Lawrence Estuary	1999-00	10	2	23	24	13	nr	nr	nr	nr	nm	59			157
Shrimp	Muscle	Canada	St. Lawrence Estuary	1999-00	10	3	19	2.2	3.3	nr	nr	nr	nr	nm	27			157
Zooplankton	Whole body	Canada	St. Lawrence Estuary	1999-00	10	5	4.1	2.9	0.8	nr	nr	nr	nr	nm	9.4			157
Dungeness crab	Hepatopan.	Canada	British Columbia-Crofton	1992-02	37	16	107	13	8.7	1.4	3.8	1.1	nm	nm	175	108	330	154
	Hepatopan.	Canada	BC-Clayoquot Sound	1992-02	37	1	0.02	0.02	0.02	0.01	0.01	0.01	nm	nm	0.26			154
	Hepatopan.	Canada	BC-Elk Falls	1992-02	37	10	132	7.1	8.5	0.8	2.1	0.7	nm	nm	174	73	281	154
	Hepatopan.	Canada	BC-Esquimalt Harbour	1992-02	37	3	74	19	7.8	1.6	2.9	0.7	nm	nm	142	132	160	154
	Hepatopan.	Canada	BC-Howe Sound	1992-02	37	15	372	51	26	3.8	11	3.8	nm	nm	593	251	1226	154
	Hepatopan.	Canada	BC-Quatsino Sound	1992-02	37	5	11	1.7	0.4	0.3	0.2	0.1	nm	nm	16.8	3.53	44.2	154
	Hepatopan.	Canada	BC-Queen Charlotte Island	1992-02	37	1	5.4	0.8	0.3	0.02	0.1	0.1	nm	nm	8.58			154
	Hepatopan.	Canada	BC-Prince Rupert	1992-02	37	13	64	12	6.3	1.1	2.0	0.5	nm	nm	109	33	205	154
	Hepatopan.	Canada	BC-Strait of Georgia	1992-02	37	1	16	2.1	0.9	0.1	0.2	0.1	nm	nm	23.4			154
	Hepatopan.	Canada	BC-Victoria Harbor	1992-02	37	9	183	50	17	7.9	7.2	2.2	nm	nm	355	159	860	154
Horse mussel	Whole body	Canada	BC-Clover Point at outflow	2006	34	1	45	29	7.3	2.2	1.9	0.2	0.0	5.3	96			155
	Whole body	Canada	BC-Clover Point at 100m	2006	34	4	613	424	125	42	39	3.4	0.8	nd	1317	699	1983	155

Table 3. (*continued*)

Matrix/	Units[a]/	Location			#		PBDE								Sum			Ref
Species	Tissue	Country	State	Year	Cong.	N	47	99	100	153	154	155	183	209	Mean	Min	Max	#
Horse mussel	Whole body	Canada	BC-Clover Point at 200m	2006	34	3	745	544	156	49	46	3.7	0.8	0.2	1621	18	2939	155
	Whole body	Canada	BC-Clover Point at 400m	2006	34	4	255	186	57	18	18	1.6	0.2	0.5	564	21	1255	155
	Whole body	Canada	BC-Clover Point at 800m	2006	34	2	881	580	172	57	56	5.1	0.6	nd	1841	1532	2150	155
	Whole body	Canada	BC-Clover Point at reference	2006	34	3	3.0	1.3	0.5	0.1	0.1	0.02	nd	1.1	6.7	5.6	8.5	155
Oyster	Whole body	US	Alabama	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	88	27	149	150
Mussel	Whole body	US	Alaska	2004-07	38	9	nr	nr	nr	nr	nr	nr	nr	nm	46	2	150	150
	Whole body	US	California	2004-07	38	36	nr	nr	nr	nr	nr	nr	nr	nm	504	21	8202	150
Oyster	Whole body	US	CA-San Francisco Estuary	2002	22	8	3390	1230	730	nr	nr	nm	nr	nr	5360	nd	11100	138
Mussel	Whole body	US	CA-San Francisco Estuary	2002	22	10	1410	659	310	nr	nr	nm	nr	nr	2380	nd	4270	138
Clam	Whole body	US	CA-San Francisco Estuary	2002	22	4	7590	2100	3810	nr	nr	nm	nr	nr	13500	13100	14000	138
Mussel	Whole body	US	Connecticut	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	282	74	495	150
Oyster	Whole body	US	Delaware	2004-07	38	1	nr	nr	nr	nr	nr	nr	nr	nm	18			150
Mussel	Whole body	US	Delaware	2004-07	38	1	nr	nr	nr	nr	nr	nr	nr	nm	14			150
Oyster	Whole body	US	Florida	2004-07	38	32	nr	nr	nr	nr	nr	nr	nr	nm	101	2	436	150
	Whole body	US	Georgia	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	44	30	64	150
	Whole body	US	Hawaii	2004-07	38	2	nr	nr	nr	nr	nr	nr	nr	nm	192	65	319	150
	Whole body	US	Louisiana	2004-07	38	16	nr	nr	nr	nr	nr	nr	nr	nm	75	11	133	150
Mussel	Whole body	US	Maine	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	73	21	143	150
Oyster	Whole body	US	Maryland	2004-07	38	6	nr	nr	nr	nr	nr	nr	nr	nm	88	37	185	150
Mussel	Whole body	US	Massachusetts	2004-07	38	15	nr	nr	nr	nr	nr	nr	nr	nm	126	11	720	150
Oyster	Whole body	US	Mississippi	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	302	79	438	150
Mussel	Whole body	US	New Hampshire	2004-07	38	1	nr	nr	nr	nr	nr	nr	nr	nm	167			150
Oyster	Whole body	US	New Jersey	2004-07	38	2	nr	nr	nr	nr	nr	nr	nr	nm	135	102	168	150
Mussel	Whole body	US	New Jersey	2004-07	38	6	nr	nr	nr	nr	nr	nr	nr	nm	334	75	784	150
Mussel	Whole body	US	New York	2004-07	38	17	nr	nr	nr	nr	nr	nr	nr	nm	714	23	2189	150
Oyster	Whole body	US	North Carolina	2004-07	38	7	nr	nr	nr	nr	nr	nr	nr	nm	19	4	35	150
Mussel	Whole body	US	Oregon	2004-07	38	5	nr	nr	nr	nr	nr	nr	nr	nm	146	41	299	150
Oyster	Whole body	US	Puerto Rico	2004-07	38	3	nr	nr	nr	nr	nr	nr	nr	nm	42	13	89	150
Mussel	Whole body	US	Rhode Island	2004-07	38	4	nr	nr	nr	nr	nr	nr	nr	nm	111	36	216	150
Oyster	Whole body	US	South Carolina	2004-07	38	5	nr	nr	nr	nr	nr	nr	nr	nm	47	4	104	150
	Whole body	US	Texas	2004-07	38	24	nr	nr	nr	nr	nr	nr	nr	nm	176	2	728	150
	Whole body	US	Virginia	2004-07	38	7	nr	nr	nr	nr	nr	nr	nr	nm	81	0	170	150
Mussel	Whole body	US	Washington	2004-07	38	18	nr	nr	nr	nr	nr	nr	nr	nm	211	7	567	150

[a] Concentrations expressed in ng g^{-1} lw for invertebrates and in units indicated for abiotic matrices; nd = not detected, nr = not reported, nm = not measured [b] PAS – passive air sampler

Lakes were in the range of 10-300 ng g^{-1} dry wt /148/, whereas those in sediment from the Saginaw River and the Niagara River were in the ranges of 0.05-50 and 2.4-34 ng g^{-1} dry wt, respectively /144, 147/. The highest concentration of BDE-209 was found in sediments from Chesapeake Bay (Maryland and Virginia), where the concentration ranged from 9,000 ng g^{-1}, near a wastewater treatment plant, to 2,400 ng g^{-1} 6 km downstream from that plant /149/.

The first comprehensive assessment of PBDE concentrations in US coastal waters was performed by the National Oceanic and Atmospheric Administration (NOAA) through its Mussel Watch Program /150/. PBDEs were found in sediments collected throughout US coasts during 2004-2007. The concentrations of total PBDEs (sum of 38 congeners) ranged from below detection limit to 88 ng g^{-1}, dry wt. The highest concentration in this study was found in sediments from an urbanized location, Marina del Rey in California (88 ng g^{-1}). High PBDE concentrations were also found in sediments from the Hudson-Raritan Estuary in New York (19-41 ng g^{-1}). The concentrations of total PBDEs in sediments from Galveston Bay (Texas) and Narragansett Bay (Rhode Island) were 15 and 12 ng g^{-1}, dry wt, respectively. The highest concentrations were found near industrial and urban locations, and sediment PBDE concentrations were directly correlated with local human populations. The concentrations of PBDEs found in the urbanized coastal areas were similar to those found in sediments from inland waterbodies (142-144, 147) and lower than those reported for coastal sediments from industrialized areas in Korea and China /145,146,151/. The sediment concentrations reported globally from highly polluted industrial locations had congener concentrations for BDE-47, -99, and -153 on the order of 1000 ng g^{-1} dry wt /152/.

The NOAA's Mussel Watch Program also reported the concentrations of PBDEs in mussels (*Mytilus* spp.) and oysters (*Crassostrea virginica*) collected throughout the US coasts in 1996 and in 2004-2007 /150/. Most tissue measurements of PBDEs (sum of 38 congeners) were above detection limits, with concentrations between 1 and 270 ng g^{-1} lw (lipid wt). Anaheim Bay, California, located in an industrialized area that includes a military base, had the highest concentration (8200 ng g^{-1} lw). Similar to those for coastal sediments, elevated PBDE concentrations in bivalves were found in developed and industrialized areas including the Hudson-Raritan Estuary in New York. The PBDE concentrations in bivalves from coastal locations were significantly positively correlated with human population within 20 km of a site. In an earlier study in California, much higher concentrations of PBDEs were reported in mussels, oysters, and clams from the San Francisco Bay (Table 3) /138/. The respective mean concentrations of PBDEs in blue mussels from Massachusetts Bay and Boston Harbor were 46 and 212 ng g^{-1}, dry weight /17/. Such concentrations are similar to those reported for bivalves from San Francisco Bay, but 2-3 orders of magnitude lower than those reported for mussels from Asian coastal waters /145,146,153/.

The Dungeness crab (*Cancer magister*) has been used as a sentinel species in the assessment of spatial trends in PBDE concentrations in coastal waters of British Columbia, Canada /154/. The highest PBDE concentrations were found in the hepatopancreas of crabs collected during 1992-2002 near heavily urbanized areas. An enrichment of less brominated PBDE congeners, particularly BDE-28/33, -47 and -49 was found for crabs, which was different from what was found for English sole and dogfish enriched with BDE-99, -100, -153 and -154, from the same location.

Marine mussels (*Modiolus modiolus*) and sediment collected in 2006 near a municipal outfall in Vancouver Island, British Columbia, were analyzed for PBDEs /155/. Total PBDE concentrations ranged from 5.59 to 96.1 ng g^{-1} dry wt, in mussels and from 0.32 to 6.2 ng g^{-1} dry wt, in sediments. The predominant congeners were BDE-47, -99, -100 and -209 in both matrices. PBDE

concentrations exhibited a parabolic relation of the biota-sediment accumulation factor (BSAF) versus the log octanol-water partition coefficient (log K_{ow}). The BSAFs for PBDEs were approximately 2-3 fold higher than those for PCBs with similar K_{ow} values. The calculated BSAFs for PBDE congeners indicated that they have a pattern of bioaccumulation potential in mussels similar to that of PCBs, and that in mussels certain PBDE congeners may be more bioaccumulative than PCBs.

2.3 Fish

The occurrence of PBDEs in fish from US waters was reported as early as 1995 /156/. Nevertheless, limited data exist on PBDE concentrations in coastal marine fishes from North and South America. Some of the highest ΣPBDE concentrations (> 1000 ng g^{-1} lw) are found in teleost fishes near urbanized areas on the west coast of British Columbia, in the St. Lawrence Estuary, in San Francisco Bay, and along the coasts of California, Georgia, and Florida. PBDE concentrations in many North American fishes are two orders of magnitude higher than the levels reported in European fish (reviewed in /157/), and their congener profiles generally show a penta-BDE signature, reflecting the heavy usage of penta-BDE in North America over the past decades. A few recent studies, however, have reported the occurrence of highly brominated PBDEs in fish from polluted coastal waters, indicating that these food webs are also contaminated by octa- and deca-BDE formulations. Table 4 presents a summary of the reported concentrations of PBDEs in wild and farm-raised fishes in coastal waters of the Americas. Wherever possible, the concentrations are shown on a ng g^{-1} lw basis to facilitate comparisons among tissue matrices (whole fish composites, fillet, muscle, liver).

PBDE concentrations, particularly BDE-47, have been increasing exponentially over the last decade on Canada's west coast and are particularly elevated in coastal areas surrounding urban centers and industrial/pulp mill towns /24,158/. Ikonomou et al /154/ detected PBDEs (37 mono- to hepta-BDEs) in muscle and liver of English sole *(Pleuronectes vetulus)* and spiny dogfish (*Squalas acanthias)*, a true shark, from various coastal locations in British Columbia. The average concentrations of PBDEs (sum of six major BDEs) were 1612, 150, and 372 ng g^{-1} lw in sole muscle, sole liver, and dogfish liver, respectively. The highest concentration (4332 ng g^{-1} lw) was found in a sole from a site near Vancouver, one of the four top urbanized zones in Canada. PBDE congener profiles showed large species-specific variation, reflecting differences in metabolic capacity or selective uptake and dietary preferences. Marked species differences were observed in the BDE-47/-99 ratios. In spiny dogfish, a higher ratio suggested a possible biotransformation of BDE-99. In laboratory exposure studies, significant debromination of BDE-99 was observed in the intestinal tract of the common carp *(Cyprinus carpio)*, resulting in the conversion of BDE-99 to BDE-47 /30/. In addition to debromination, the preferential excretion of BDE-99 has been suggested as a reason for the lower accumulation of BDE-99 relative to BDE-47 in carnivorous marine fish and Irrawaddy dolphin *(Orcaella brevirostris)* /159, 160/. Spatial/geographic variations in congener patterns were minimal, with the exception of a few sole samples showing a more pronounced penta-BDE signature, presumably indicating proximity to a point source.

The St. Lawrence Estuary, located downstream from the Great Lakes-St. Lawrence River, receives riverine inputs of contaminants from one of the most industrialized areas of the world. A study of contaminant loading to the St. Lawrence Estuary food web measured PBDE congeners in a variety of marine organisms collected during 1999-2000, including several species of pelagic and demersal fishes /157/. The highest average PBDE levels (sum of 10 major congeners) were found in the liver of Atlantic tomcod (*Microgadus tomcod*) (1305 ng g^{-1} lw), smooth flounder (*Pleuronectes putmani*)

(745 ng g^{-1} lw) and rainbow smelt (*Osmerus mordax*) (630 ng g^{-1} lw). The BDE-47/-99 ratios in certain fish species like rainbow smelt, smooth flounder, American plaice (*Hippoglossoides platessoides*), and American eel (*Anguilla rostrara*) were much higher than in other species, whereas the BDE-47/-100 ratios were similar among all species.

Among US coastal environments, the highest PBDE concentrations in marine fishes have been reported in teleost fishes from the California coast. Compared with other US locations, the PBDE levels were 2- to 4-fold higher in the serum of California residents and in house dust, presumably owing to the state's strict furniture flammability standards and high usage /97/. Brown et al /161/ measured PBDEs in 15 species of teleost fishes collected in 2000 at various locations along the California coast. For all fish species from all sampling areas, the mean PBDE concentration (sum of BDEs 47, -99, -100, 153, and -154) in fillets was 302 ng g^{-1} lw. The highest concentrations of PBDEs were found in fish near areas of high population density and in semi-enclosed bays, e.g., San Diego Bay, Los Angeles, and San Francisco Bay. Higher PBDE concentrations were reported in marine fishes collected in 2002 from contaminated "hot spots" in San Francisco Bay /162/. The PBDE concentrations in whole individual fish (minus head, tail, and guts) ranged from 306 ng g^{-1} lw in jack smelt (*Atherinopsis californiensis*) to 2235 and 1925 ng g^{-1} lw in halibut (*Hippoglossus stenolepis*) and striped bass (*Morone saxatilis*), respectively. One leopard shark (*Triakis semifasciata*) contained a ΣPBDE concentration of 489 ng g^{-1} lw. The PBDE concentrations in California coastal fishes are higher than those reported in marine fishes from Europe /163,164/, Japan /165,166/, eastern and southern China /167, 168/, and Asia-Pacific waters /168/, but are an order of magnitude lower than the maximum levels reported in freshwater fish from urbanized watersheds of North America /37/ and Europe /169/.

A study by Lunder and Sharp /103/ measured PBDE concentrations (sum of BDEs-28 -33, -47, -99, -100, -153, and -154) in five species of teleost fishes and in leopard sharks caught by anglers in 2002 in San Francisco Bay. The respective highest mean concentrations were found in fillets of halibut and striped bass (2009 and 1756 ng g^{-1} lw). Halibut and striped bass, the two most commonly eaten species, are highly carnivorous, migratory species. PBDE concentrations were also relatively high in other fish species from the Bay (672 and 652 ng g^{-1} lw, in walleye surf perch [*Hyperprosopona argenteum*] and white croaker [*Genyonemus lineatus*], respectively).

Sajwan et al /170/ investigated PBDEs and organochlorines in six species of teleost fishes collected in 2005 from coastal waters off Savannah, Georgia. The Savannah River estuary receives contaminant inputs from industries and chemical manufacturing plants in the port of Savannah, as well as wastewaters and toxic discharges from the Savannah River, which ranks 7th among 50 highly contaminated US rivers. The mean PBDE concentrations in whole fish (sum of BDE-28/33, -47, -66, -99, -100, and -154) ranged from 15.5 ng g^{-1} lw in rock seabass (*Centropristis philadelphica*) to 337 ng g^{-1} lw in silver perch (*Bidyanus bidyanus*). The PBDE congener profiles varied between species but generally followed the order: BDE-47> -99> -100 ~ -28/33. The PBDE concentrations in fish from three coastal locations rivaled PCB concentrations (12-493 ng g^{-1} lw) and surpassed the levels of organochlorine pesticides.

Only a few studies have reported on the concentrations of BDE-209 and other highly brominated PBDEs in coastal marine fishes. Johnson-Restrepo et al /25/ measured PBDEs (tetra- to deca-BDEs) in several species of commercially important teleost fishes and cartilaginous fishes (sharks, stingrays) collected in 2004 from coastal Florida waters. The mean concentrations of PBDEs (sum of 12 congeners) in muscle tissues of teleost fishes ranged from 8 ng g^{-1} lw, in silver perch to 88 ng g^{-1} lw,

Table 4. *Concentrations of individual PBDE congeners (ng g⁻ lw) and mean and range of ΣPBDEs in marine fish. Reference numbers refer to the corresponding references in the manuscript.*

Species	Tissue	Location		Year	# Cong.	N	PBDE (ng g^{-1} lipid wt)								Sum PBDE			Ref #
		Country	State				47	99	100	153	154	155	183	209	Mean	Min	Max	
Wild Fish																		
American eel	muscle	Canada	St. Lawrence Estuary	1999-00	10	5	271	7.1	63	nr	nr	nr	nr	nr	421			157
Greenland halibut	muscle	Canada	St. Lawrence Estuary	1999-00	10	3	88	11	40	nr	nr	nr	nr	nr	178			157
Greenland halibut	liver	Canada	St. Lawrence Estuary	1999-00	10	2	43	4.7	5.3	nr	nr	nr	nr	nr	63			157
Rainbow smelt	liver	Canada	St. Lawrence Estuary	1999-00	10	4	407	19	107	nr	nr	nr	nr	nr	630			157
Atlantic herring	liver	Canada	St. Lawrence Estuary	1999-00	10	3	212	47	35	nr	nr	nr	nr	nr	382			157
Atlantic tomcod	liver	Canada	St. Lawrence Estuary	1999-00	10	6	576	367	171	nr	nr	nr	nr	nr	1305			157
American plaice	liver	Canada	St. Lawrence Estuary	1999-00	10	3	130	3.9	25	nr	nr	nr	nr	nr	221			157
Smooth flounder	liver	Canada	St. Lawrence Estuary	1999-00	10	3	545	7.3	105	nr	nr	nr	nr	nr	745			157
Sockeye salmon	cleaned fish	Canada	British Columbia	2000	38	2	0.92	0.36	0.13	0.03	0.09	0.02	nm	nd	1.7	1.4	1.9	172
English sole	muscle	Canada	British Columbia	2000	37	7	706	442	181	89	62	6.2	nr	nr	1612	266	4332	154
English sole	liver	Canada	British Columbia	2000	37	13	77	25	21	9.2	8.0	1.7	nr	nr	150	52	323	154
Spiny dogfish	liver	Canada	British Columbia	2000	37	2	198	42	83	15	11	2.4	nr	nr	372	362	381	154
Winter flounder	whole fish	US	Gulf of Maine	2006	35	1	35	2.5	6.4	0.78	2	1.3	nd	nd	52			32
Atlantic herring	whole fish	US	Gulf of Maine	2006	35	6	40	6.9	6.8	0.45	1.4	0.61	nd	nd	82	71	91	32
American plaice	whole fish	US	Gulf of Maine	2006	35	1	42	4	7	1.2	2.8	3.6	0.13	1.8	69			32
White hake	whole fish	US	Gulf of Maine	2006	35	2	25	0.63	7.2	0.35	1.9	0.31	nd	0.91	42	38	47	32
Alewife	whole fish	US	Gulf of Maine	2006	35	2	8.3	3.6	1.7	0.3	0.72	0.19	nd	nd	18	18	19	32
Atlantic mackerel	whole fish	US	Gulf of Maine	2006	35	4	20	7.5	4.1	1.4	1.4	0.52	7.9	1.6	69	26.3	152	32
Silver hake	whole fish	US	Gulf of Maine	2006	35	1	18	6.3	4	0.31	2.2	0.94	nd	nd	38			32
Flounder	whole fish	US	GA, Savannah River	2005	13	3	64	6.3	1.2	nr	<1.0	nr	nr	nr	89.5	71	108	170
Anchovy	whole fish	US	GA, Savannah River	2005	13	30	31	20	13	nr	4.5	nr	nr	nr	69			170
Silver perch	whole fish	US	GA, Savannah River	2005	13	2	241	73	<1.0	nr	<1.0	nr	nr	nr	337			170
Anchovy	whole fish	US	GA, Lazaretto Creek	2005	13	26	31	27	3.3	<1.0	<1.0	nr	nr	nr	67			170
Flounder	whole fish	US	GA, Lazaretto Creek	2005	13	2	46	17	3.1	0.5	1.4	nr	nr	nr	71.5	69	74	170
Rock seabass	whole fish	US	GA, Lazaretto Creek	2005	13	2	12	<1.0	0.8	<1.0	0.9	nr	nr	nr	15.5	10	21	170
Silver perch	whole fish	US	GA, Lazaretto Creek	2005	13	1	26	<1.0	<1.0	<1.0	<1.0	nr	nr	nr	26			170
Silver perch	muscle	US	Florida coast	2004	13	6	3.6	1.7	0.4	0.2	0.2	nm	0.1	<0.02	8	3.7	14.3	25
Striped mullet	muscle	US	Florida coast	2004	13	6	21	2.5	1.5	1.1	2	nm	<0.001	<0.02	31	2	63	25
Spotted seatrout	muscle	US	Florida coast	2004	13	7	24	3.7	3.9	0.6	1	nm	0.8	<0.02	36	12	78	25
Red drum	muscle	US	Florida coast	2004	13	11	32	5.7	5.6	1	1.6	nm	<0.001	<0.02	52	2.7	154	25
Hardhead catfish	muscle	US	Florida coast	2004	13	8	26	18	7.4	11	9.3	nm	0.04	4.5	88	1.8	306	25

Species	Tissue	Location		Year	# Cong.	N	PBDE (ng g^{-1} lipid wt)								Sum PBDE			Ref #
		Country	State				47	99	100	153	154	155	183	209	Mean	Min	Max	
Atlantic stingray	muscle	US	Florida coast	2004	13	7	20	14	2.4	2.6	1.2	nm	0.4	0.1	42	2.3	83	25
Spiny dogfish	muscle	US	Florida coast	2004	13	5	11	2.7	3.3	0.7	1.6	nm	<0.001	17	38	16	60	25
Atl. sharpnose shark	muscle	US	Florida coast	2004	13	5	29	12	16	4.3	8.8	nm	<0.001	514	589	45	1930	25
Bull shark	muscle	US	Florida coast	1993-94	13	6	25	6.1	4.2	0.8	2.8	nm	<0.001	39	78	16	287	25
Bull shark	muscle	US	Florida coast	2002-04	13	7	483	15	155	44	139	nm	<0.001	778	1630	12	4190	25
Chinook salmon	skin-on filet	US	Alaska	2003-04	9	3	5.4	1.2	1.1	0.69	0.69	nm	nm	nm	11	5.1	21	175
Chinook salmon	cleaned fish	US	Alaska	2000	38	1	2.8	0.97	0.43	nd	nd	0.03	nm	nd	4.9			172
Chum salmon	cleaned fish	US	Alaska	2000	38	1	0.67	nd	0.1	nd	nd	nd	nm	nd	0.9			172
Chinook salmon	skin-on filet	US	Alaska	2002	44	3	2	0.55	0.34	0.06	0.09	0.03	0.01	0.35	4.1	3.1	5.3	174
Shiner surf perch	filet	US	California coast	2001	5	18	200	23	102	3.3	26	nm	nm	nm	355	23	1024	161
Striped bass	filet	US	California coast	2001	5	10	228	18	88	7.9	21	nm	nm	nm	363	265	419	161
Spotted sand bass	filet	US	California coast	2001	5	5	186	80	75	21	27	nm	nm	nm	388	84	957	161
White croaker	filet	US	California coast	2001	5	18	136	22	112	20	64	nm	nm	nm	354	136	512	161
Walleye surf perch	filet	US	CA, San Francisco Bay	2002	7	4	508	133[a]		16[b]		nr	nr	nr	672			103
Shiner surf perch	filet	US	CA, San Francisco Bay	1997	7	40	669	185[a]		23[b]		nr	nr	nr	903			103
Halibut	filet	US	CA, San Francisco Bay	2002	7	4	1286	548[a]		151[b]		nr	nr	nr	2009			103
Halibut	filet	US	CA, San Francisco Bay	1997	7	3	641	154[a]		2[b]		nr	nr	nr	821			103
Jack smelt	filet	US	CA, San Francisco Bay	2002	7	5	122	121[a]		29[b]		nr	nr	nr	282			103
Jack smelt	filet	US	CA, San Francisco Bay	1997	7	15	158	124[a]		15[b]		nr	nr	nr	312			103
White croaker	filet	US	CA, San Francisco Bay	2002	7	4	444	166[a]		28[b]		nr	nr	nr	652			103
White croaker	filet	US	CA, San Francisco Bay	1997	7	10	395	131[a]		25[b]		nr	nr	nr	564			103
Striped bass	filet	US	CA, San Francisco Bay	2002	7	4	1021	526[a]		179[b]		nr	nr	nr	1756			103
Striped bass	filet	US	CA, San Francisco Bay	1997	7	6	349	118[a]		26[b]		nr	nr	nr	516			103
Leopard shark	filet	US	CA, San Francisco Bay	2002	7	1	310	110[a]		28[b]		nr	nr	nr	474			103
Leopard shark	filet	US	CA, San Francisco Bay	1997	7	12	300	60[a]		8[b]		nr	nr	nr	438			103
Marlin	liver	US	Puerto Rico coast	1991	?	9	nr	nr	nr	nr	nr	nr	nr	nr	100			22
Skipjack tuna	muscle	Brazil	Coast	2000	11	4	7.1	1.9	1.8	0.45	1.2	nm	<0.05	<5.0	13			168
Farmed Fish																		
Atlantic salmon	skin-on filet	Canada	East	2003-04	9	6	3.3	0.91	0.62	0.37	0.37	nm	nm	nm	6.4	3.1	11	175
Atlantic salmon	skin-on filet	Canada	East	2002	44	24	12	2.9	2.2	0.33	0.56	0.32	0.08	0.21	22	9.0	38	174
Chinook salmon	cleaned fish	Canada	British Columbia	2000	38	2	13	2.0	2.3	0.31	0.65	0.31	nm	nd	20	8.5	32	172
Atlantic salmon	skin-on filet	US	Maine	2003-04	9	3	4.6	1.2	0.86	0.48	0.48	nm	nm	nm	8.7	4.7	14	175
Atlantic salmon	skin-on filet	US	Maine	2002	44	6	7.3	1.9	1.4	0.22	0.35	0.2	0.05	0.1	14	13	15	174
Atlantic salmon	skin-on filet	Chile	Coast	2004	35	14	10	1.9	2.3	0.6	0.7	nr	nr	nr	16	12	24	176

[a] Sum of BDE-99 and BDE-100; [b] Sum of BDE-153 and BDE-154; nd = not detected, nr = not reported, nm = not measured

in hardhead catfish (*Arius felis*), with an overall mean (±SD) concentration of 43 ± 30 ng g^{-1} lw. The mean concentrations in muscle of sharks ranged from 37.8 ng g^{-1} lw in spiny dogfish to 1620 ng g^{-1} lw in bull sharks (*Carcharhinus leucas*). The maximum PBDE concentration (4190 ng g^{-1} lw) was detected in a bull shark. For teleost fishes and Atlantic stingrays (*Dasyatis sabina*), the congener profiles followed the order: BDE-47 (55%)> -99> -100> -153> -154. The occurrence of BDE-183, -203, and -209 in certain teleost fish species indicated exposure to the octa-BDE and deca-BDE mixtures. In contrast to the general perception that BDE 209 does not accumulate in higher trophic level aquatic organisms, levels up to 514 and 778 ng g^{-1} lw were detected in Florida sharpnose (*Rhizoprionodon terraenovae*) and in bull sharks, respectively. BDE-209 was the dominant congener in sharks, accounting for 60% of the total PBDE content.

A study by Shaw et al 2009 /32/ analyzed PBDEs (26 tri-deca BDEs) in seven species of commercially important teleost fishes collected from the Gulf of Maine in 2006. The PBDE concentrations (sum of 10 BDE congeners) in whole fish samples ranged, on average, from 18.3 in alewife (*Alosa pseudoharengus*) to 81.5 ng g^{-1} lw in Atlantic herring (*Clupea harengus*), with an overall mean of 62 ± 34 ng g^{-1} lw. The PBDE levels in Gulf of Maine fishes were similar to those reported in muscle of Florida coastal fishes /25/, but 3-15 times lower than levels in marine fishes from the Georgia coast /170/ or the California coast /161/. BDE 47 accounted for 45% to 68% of the total PBDE content in Gulf of Maine fish /32/. The congener profiles were highly variable, and large differences in BDE-47/-99 ratios were apparent among the species (including those belonging to the same family). Highly brominated congeners (BDEs-181, -183, 197, -203, -207 and -209) were detected at low levels in three fish species—American plaice, Atlantic mackerel (*Scomber scombrus*), and white hake (*Urophycis tenuis*). This congener pattern was suggestive of BDE-209 debromination in the fish, as previously shown in freshwater fish /30, 31/, resulting in the enrichment of less brominated congeners.

The consumption of farmed salmon has dramatically increased in North America. In the US alone, salmon consumption has increased annually by more than 26% since 1987, and at present, more than half of the salmon consumed globally is farmed /171/. Early reports of higher levels of POPs, including PBDEs, in farm-raised versus wild fish raised concerns about the rising salmon consumption by Americans /172,173/. Several studies have examined PBDE levels in wild and farmed salmon in North and South America. A pilot study by Easton et al /172/ measured PBDEs in farmed and wild salmon from Alaska and British Columbia. The mean concentrations (sum of 31 detected BDEs) in wild salmon (skin-on fillets) from Alaska in 2000 ranged from 0.9 to 4.0 ng g^{-1} lw in chum and Chinook salmon, respectively. PBDE concentrations in wild sockeye salmon from British Columbia were 1.7 ng g^{-1} lw in contrast to 20 ng g^{-1} lw in farmed Chinook salmon.

Hites et al /174/ conducted a global assessment of mono- through nona-PBDEs in farmed and wild salmon collected in 2002 from suppliers in Europe, eastern and western Canada, Chile, and the US states of Oregon, Maine, Washington, and Alaska. Among the five wild salmon species from British Columbia, Oregon, and Alaska, the mean PBDE concentrations (sum of 43 BDE congeners in skin-on fillets) were approximately 17 times higher in Chinook salmon (*Oncorhynchus tshawytscha*) than in pink (*Oncorhynchus gorbuscha*), coho (*Oncorhynchus kisutch*), chum (*Oncorhynchus keta*), and sockeye salmon (*Oncorhynchus nerka*). The highest concentrations were found in wild Chinook salmon from highly urbanized areas of British Columbia (4.2 ng g^{-1} ww) and the Oregon coast (2.1 ng g^{-1} ww), in contrast to much lower concentrations (0.49 ng g^{-1} ww) in Chinook from the Alaskan coast. The higher concentrations in Chinook samples in both studies reflect species

differences with respect to feeding behavior and the higher trophic level of Chinook salmon. With the exception of the wild Chinook from British Columbia, farm-raised salmon had significantly higher PBDE levels than the wild salmon. Farmed salmon from Scotland and western Canada had the highest concentrations (3.8 and 3.3 ng g^{-1} ww, respectively), whereas salmon from Chile and Washington state (1.2 ng g^{-1} ww) had the lowest concentrations (1.3 and 1.2 ng g^{-1} ww, respectively). Intermediate concentrations (3.1 and 2.6 ng g^{-1} ww) were found in farmed salmon from eastern Canada and Maine. An analysis of PBDEs in feed samples indicated that the higher concentrations in farmed salmon were a function of their diet /174/. Farmed salmon are fed a combination of fish oils and fish meal derived from small forage fishes, whereas the diet of wild salmon is varied and includes lower trophic organisms (invertebrates and zooplankton) as well as fish.

A study by Shaw et al /175/ measured PBDEs (di- through hexa-BDEs) in farm-raised Atlantic salmon (*Salmo salar*) collected in 2003-2004 from Maine, eastern Canada, and Norway, and wild Alaskan Chinook salmon. In contrast to the findings of Hites et al /174/, the PBDE concentrations (sum of BDEs-47, -99, -100, and -28) in the farmed salmon (0.4-1.4 ng g^{-1} ww) were not significantly different from those in the wild Alaskan Chinook (0.4-1.2 ng g^{-1} ww), nor were significant differences found among regions, although intra-regional (intra-producer) differences in concentrations were observed in the salmon from various Canadian farms. Concentrations in the Canadian samples (0.85 ng g^{-1} ww or 6.3 ng g^{-1} lw) were three times lower than those reported in Canadian farmed salmon two years earlier by Hites et al /174/, presumably reflecting industry efforts to reduce the contaminant levels in salmon feed. The PBDE concentrations in farmed salmon from Maine (0.95 ng g^{-1} ww or 7.3 ng g^{-1} lw) did not show a similar reduction over time /175/. The PBDE concentrations in farmed salmon from Maine and eastern Canada were similar to those reported in farmed salmon from Chile /174,176/ and Japan /119/, and 3- to 6-fold lower than levels reported in European farmed salmon /173,177/. PBDE levels were compared in skin-on and skin-off fillets to determine whether removal of skin and subdermal fat lowers contaminant levels in farmed salmon /175/. Skin removal resulted in no overall reduction in PBDE concentrations and in certain cases PBDE levels were higher in the skin-off samples. Moreover, PBDE levels were correlated with lipids only in the skin-off samples, implying a greater accumulation and retention of PBDEs in muscle lipids of fish on high energy diets than in skin-associated fat. Interestingly, the Norwegian salmon analyzed in the study were labeled “organically farmed”, but were later found to be conventionally farmed salmon that had been exported to the US and falsely labeled /178/. These samples had the highest levels of PBDEs, indicating that purchasing highly priced salmon labeled as organic does not necessarily protect the consumer from exposure.

The data on PBDE concentrations in marine fishes from Central and South America are very sparse. Montory and Barra /176/ measured PBDEs (mono- through deca-BDEs) in farmed salmon from five different regions in southern Chile. PBDE concentrations (sum of BDEs- 28, -47, -49, -99, -100, -153, and -154) in all salmon (skin-on fillets) averaged 1.46 ng g^{-1} ww, and no difference was observed in concentration or congener profiles across the five regions. These concentrations are relatively low, on a global scale, and are similar to the PBDE levels previously reported in Chilean farmed salmon /174/ and those reported in farmed salmon from eastern Canada and Maine /175/ and Japan /119/. Congener profiles in salmon followed the order BDE-47 (40%) >-99 > -100 ~ -49. Hepta- through deca-BDE congeners were also present at trace levels. The PBDE levels measured in five salmon feed samples (0.21-5.4 ng g^{-1} ww) were higher than the levels in fish muscle and contained 6.9% by weight of BDE-209, suggesting possible debromination of this congener in the fish. A strong

Table 5. *Concentrations of individual PBDE congeners (ng g^{-1} lw) and mean and range of ΣPBDEs in seabirds. Reference numbers refer to the corresponding references in the manuscript.*

Species	Tissue	Location		Year	#		PBDE								Sum PBDE			Ref
		Country	State		Cong.	N	47	99	100	153	154	155	183	209	Mean	Min	Max	#
	eggs	Canada	BC.-Mandarte Isl.	1979	18	1	2.3	1.2	0.62	0.62	0.21	nr	nd	nm	5.0			27
	eggs	Canada	BC-Mandarte Island	1985	18	5	33	46	23	10	8.6	nr	0.53	nm	130			27
Double-crested	eggs	Canada	BC -Mandarte Island	1990	18	11	129	139	131	40	38	nr	0.75	nm	486			27
cormorant[1]	eggs	Canada	BC-Mandarte Island	1994-5	18	13	2248	831	1285	493	332	nr	1.3	nm	5259			27
	eggs	Canada	BC -Mandarte Island	1998	18	6	888	727	911	426	248	nr	nd	nm	3223			27
	eggs	Canada	BC-Mandarte Island	2002	18	3	248	140	339	147	63	nr	0.61	nm	950			27
Leach's storm petrel	eggs	Canada	BC-Hippa Island	2000	6	1	0.93[a]	0.76[a]	0.49[a]	0.93[a]	0.2[a]	nr	nr	nr	3.4[a]			27
	plasma	Canada	BC-Fort St. James	2001	8	4	nr	31	nr	nd	nd	nm	nd	nd	57	nd	179	199
	plasma	Canada	BC-Barkley Sound	2003	8	3	164	120	71	22	nd	nm	nd	nd	377	62	774	199
Bald eagle	plasma	Canada	BC-Nanaimo-Crofton	2003	8	7	425	162	183	29	nd	nm	nd	nd	801	403	1780	199
	plasma	Canada	BC-Delta-Richmond	2003	8	6	145	78	70	58	nd	nm	nd	nd	342	112	644	199
	plasma	Canada	BC-Abbotsford-Chilliwack	2003	8	6	99	44	51	15	nd	nm	nd	nd	205	131	391	199
Common eider	eggs	US	Maine Coast	2007	13	6	nr	nr	nr	nr	nr	nr	nr	nr	30			190
Leach's storm petrel	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	110			190
Arctic tern	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	143			190
Common tern	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	134			190
Least tern	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	110			190
DC cormorant	eggs	US	Maine Coast	2007	13	5	nr	nr	nr	nr	nr	nr	nr	nr	596			190
Black guillemot	eggs	US	Maine Coast	2007	13	3	nr	nr	nr	nr	nr	nr	nr	nr	439			190
Atlantic puffin	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	577			190
Herring gull	eggs	US	Maine Coast	2007	13	6	nr	nr	nr	nr	nr	nr	nr	nr	2340			190
Peregrine falcon	eggs	US	Maine Coast	2007	13	1	nr	nr	nr	nr	nr	nr	nr	nr	2551			190
Osprey	eggs	US	Maine Coast	2007	13	6	nr	nr	nr	nr	nr	nr	nr	nr	4204			190
Bald eagle	eggs	US	Maine Coast	2007	13	4	nr	nr	nr	nr	nr	nr	nr	nr	8627			190
	eggs	US	Delaware Bay-North	2000-01	7	6	276[a]	111[a]	99[a]	40[a]	37[a]	nm	nm	nm	572[a]	442	820	189
Osprey	eggs	US	Delaware Bay-Central	2000-01	7	6	124[a]	18[a]	35[a]	11[a]	13[a]	nm	nm	nm	206[a]	141	429	189
	eggs	US	Delaware Bay-South	2000-01	7	2	47[a]	7.9[a]	12[a]	5.3[a]	7.9[a]	nm	nm	nm	82[a]	71	94	189

Species	Tissue	Location		Year	#		PBDE								Sum PBDE			Ref
		Country	State		Cong.	N	47	99	100	153	154	155	183	209	Mean	Min	Max	#
Osprey	eggs	US	Ches. Bay-South Riv.	2000	7	15	114[a]	13[a]	24[a]	8.6[a]	9.8[a]	nm	nm	nm	176[a]	87	275	188
	eggs	US	Ches. Bay-Baltimore Harbor	2000	7	14	198[a]	45[a]	32[a]	16[a]	15[a]	nm	nm	nm	320[a]	272	357	188
	eggs	US	Ches. Bay-Potomac River	2000	7	16	480[a]	106[a]	79[a]	30[a]	21[a]	nm	nm	nm	725[a]	560	928	188
	eggs	US	Ches. Bay-South River	2001	7	15	156[a]	21[a]	31[a]	16[a]	12[a]	nm	nm	nm	253[a]	157	397	188
	eggs	US	Ches. Bay-Elizabeth River	2001	7	15	120[a]	17[a]	26[a]	8.6[a]	6.6[a]	nm	nm	nm	195[a]	154	248	188
	eggs	US	Delaware Bay-South	2000-01	7	2	47[a]	7.9[a]	12[a]									
	eggs	US	Ches. Bay-South Riv.	2000	7	15	114[a]	13[a]	24[a]									
	eggs	US	Delaware Bay-South	2000-01	7	2	47[a]	7.9[a]	12[a]	5.3[a]	7.9[a]	nm	nm	nm	82[a]	71	94	189
	eggs	US	Ches. Bay-South Riv.	2000	7	15	114[a]	13[a]	24[a]	8.6[a]	9.8[a]	nm	nm	nm	176[a]	87	275	188
	eggs	US	Ches. Bay-Balt/ Harbor	2000	7	14	198[a]	45[a]	32[a]	16[a]	15[a]	nm	nm	nm	320[a]	272	357	188
	eggs	US	Ches. Bay-Potomac Riv.	2000	7	16	480[a]	106[a]	79[a]	30[a]	21[a]	nm	nm	nm	725[a]	560	928	188
	eggs	US	Ches. Bay-South Riv.	2001	7	15	156[a]	21[a]	31[a]	16[a]	12[a]	nm	nm	nm	253[a]	157	397	188
	eggs	US	Ches. Bay-Elizabeth Riv.	2001	7	15	120[a]	17[a]	26[a]	8.6[a]	6.6[a]	nm	nm	nm	195[a]	154	248	188
Peregrine falcon	eggs	US	Maine	1996-06	30	1	43.8	175	62.5	200	47	nm	141	29	885			182
	eggs	US	New Hampshire	1996-06	30	12	30	242	107	704	87	nm	382	168	2188	523	9062	182
	eggs	US	Massachusetts	1996-06	30	8	28.6	214	76.1	673	63	nm	169	117	1763	901	3128	182
	eggs	US	Connecticut	1996-06	30	3	24.5	196	63.4	567	96	nm	193	175	1800	1148	2442	182
	eggs	US	Rhode Island	1996-06	30	2	99.5	147	51.8	277	49	nm	55	94	1001	493	1510	182
	eggs	US	Vermont	1996-06	30	9	12.3	101	33.2	526	46	nm	201	114	1430	278	5896	182
	eggs	US	Ches. Bay-S. Marsh Isld.	1993	12	1	nr	nr	nr	nr	nm	nm	nr	1.2[a]	222[a]			181
	eggs	US	Ches. Bay-Spring Isld.	1999	12	1	nr	nr	nr	nr	nm	nm	nr	2.4[a]	221[a]			181
	eggs	US	Ches. Bay-Chincoteague	1993	12	1	nr	nr	nr	nr	nm	nm	nr	0.2[a]	77[a]			181
	eggs	US	Ches. Bay-Fort Eustis	1993	12	1	nr	nr	nr	nr	nm	nm	nr	1.1[a]	33[a]			181
	eggs	US	Ches. Bay-Ben. Harrison Brg.	1999	12	1	nr	nr	nr	nr	nm	nm	nr	5.7[a]	148[a]			181
	eggs	US	Ches. Bay-Fisherman's Isld.	2002	12	2	nr	nr	nr	nr	nm	nm	nr	3.2[a]	127[a]	122	132	181

[a] Concentrations expressed in ng g^{-1} wet weight; nd = not detected, nr = not reported, nm = not measured;

[b] BC – British Columbia Ches. – Chesapeake; SF – San Francisco; Isld - Island

Table 5. (*continued*)

Species	Tissue	Country	Location / State	Year	#		PBDE								Sum PBDE			Ref
					Cong.	N	47	99	100	153	154	155	183	209	Mean	Min	Max	#
Peregrine falcon	eggs	US	Ches. Bay-Elkins Marsh	2002	12	2	nr	nr	nr	nr	nm	nm	nr	13[a]	240[a]	186	294	181
	eggs	US	Ches. Bay-James Riv. Brg.	2001-02	12	4	nr	nr	nr	nr	nm	nm	nr	7.7[a]	108[a]	126	169	181
	eggs	US	Ches. Bay-Berkeley Brg.	2001-02	12	8	nr	nr	nr	nr	nm	nm	nr	30[a]	262[a]	168	354	181
	eggs	US	Ches. Bay-Norfolk South. Brg	1993	12	1	nr	nr	nr	nr	nm	nm	nr	38[a]	201[a]			181
	eggs	US	Ches. Bay-Legg Mason Build.	1999	12	1	nr	nr	nr	nr	nm	nm	nr	21[a]	262[a]			181
Glaucous gull	eggs	US	Alaska-Chuckchi Sea	2005	10	3	nr	nr	nr	nr	nr	nm	nr	nm	76.8	59.5	470	197
	eggs	US	Alaska-Bearing Sea	2005	10	3	nr	nr	nr	nr	nr	nm	nr	nm	92.8	51.1	142	197
Glaucous-winged gull	eggs	US	Alaska-Bearing Sea	2005	10	3	nr	nr	nr	nr	nr	nm	nr	nm	61.4	47.2	99.2	197
	eggs	US	Alaska-Gulf of Alaska	2005	10	3	nr	nr	nr	nr	nr	nm	nr	nm	348	243	4130	197
Caspian tern	eggs	US	Washington-Gray's Harbor	2001	5	20	2660	1210	564	145	97	nr	nr	nr	4670	1970	15000	193
	eggs	US	California-SF Bay	2000	5	5	1350	521	399	108	60	nr	nr	nr	2590	2010	3540	193
	eggs	US	California- SF Bay	2001	5	13	3630	977	950	204	171	nr	nr	nr	5930	1340	17300	193
	eggs	US	California- SF Bay	2002	5	20	2760	1590	1150	717	342	nr	nr	nr	6760	1270	36100	193
	eggs	US	California- SF Bay	2003	5	20	2900	925	744	184	127	nr	nr	nr	5160	1200	26300	193
Forster's tern	eggs	US	California- SF Bay	2000	5	5	1036	672	157	167	57	nr	nr	nr	2160	1080	3560	193
	eggs	US	California- SF Bay	2001	5	29	5400	1320	573	167	156	nr	nr	nr	7610	1460	62400	193
	eggs	US	California- SF Bay	2002	5	20	3920	2550	1220	1000	578	nr	nr	nr	9420	2590	63300	193
	eggs	US	California- SF Bay	2003	5	22	3280	1260	429	281	205	nr	nr	nr	5610	666	26000	193
Least tern	eggs	US	California- SF Bay	2001	5	5	2550	1440	1030	186	220	nr	nr	nr	5420	4070	7540	193
	eggs	US	California- SF Bay	2002	5	5	2520	1900	967	224	165	nr	nr	nr	5870	4210	7820	193
Peregrine falcon	eggs	US	California coast	1986-07	16	95	nr	nr	nr	nr	nr	nr	nr	490	7850	80	53000	185
Bald eagle	plasma	US	CA-Santa Catalina Island	2003	8	3	2685	638	568	114	46	nm	266	nd	4755			199

[a] Concentrations expressed in ng g^{-1} wet weight; nd = not detected, nr = not reported, nm = not measured;

[1] BC – British Columbia Ches. – Chesapeake; SF – San Francisco

positive correlation was found between the PBDE levels in fish tissue and fish feed, indicating that feed is the main source of PBDEs for Chilean farmed salmon.

Greaves and Harvey /179/ (cited in /22/) reported on PBDEs in livers of blue marlins (*Makaira nigricans*) collected off the coast of Puerto Rico in 1991. The total PBDE concentrations (BDEs- 17, -28/33, -47, -66, -85, -99, -100, -138, -153, and -154) in the marlins were less than 100 ng g^{-1} lw. PCBs and DDT/DDE were present in tissue at 10-fold higher concentrations. Interestingly, two methoxy derivatives of tetra-BDEs were detected at concentrations exceeding those of the PBDEs in the marlins. Not clear was whether these compounds were PBDE metabolites or derivatives of natural products originating from marine sponges or algae /180/.

Ueno et al /168/ measured PBDEs in muscle tissue of skipjack tuna *(Katsuwonus pelamis)* that were collected in 2000 off the Brazilian coast, as part of a global study of PBDE contamination in offshore ocean waters and the southern hemisphere. Generally, PBDE concentrations were higher in tuna samples from the northern hemisphere than those from the southern hemisphere, reflecting the greater usage of these compounds in industrialized northern areas. In all samples, the PBDE levels (sum of 11 BDE congeners) ranged from <0.1 to 53 ng g^{-1} lw. The highest mean concentrations were found in tuna from Taiwan offshore waters (ΣPBDEs 53 ng g^{-1} lw) and at sites around the East China Sea and the South China Sea. The levels of tuna from Brazilian waters (13 ng g^{-1} lw) were comparable to those in Asian regions. The sources in Asian regions are presumed to be electronics manufacturers located in coastal areas of developing Asian countries and massive amounts of electronic wastes exported to these areas from the US, Canada, Japan, and other developed nations. Interestingly, the congener profiles in tuna shifted with latitude and suggested that less brominated congeners (di-, tri-, and tetra-BDEs) are preferentially transported from pollution sources to the northern colder regions, whereas the proportion of BDEs-153, -154, and -183 decreased with increasing latitude. The results suggested that less brominated BDEs have high transportability.

2.4 Seabirds

The reported concentrations of PBDEs in seabirds from coastal areas of the US and Canada are shown in Table 5. The available data suggest that PBDE concentrations in North American birds of prey are among the highest reported for wildlife, and for species that feed in terrestrial food chains, the PBDE congener profiles differ markedly with respect to highly brominated congeners, including BDE-209. Tetra- and penta-BDE congeners typically dominate in piscivorous birds, akin to the pattern in their diet. However, the constituents of the octa- and deca-BDE formulations are prominent congeners in peregrine falcon eggs, especially those from highly urbanized areas, reflecting their terrestrial-dependent food web. PBDE burdens in North American aquatic bird eggs are generally higher than those in eggs from Europe or Asia, reflecting the greater usage and release of PBDE flame retardants in North America.

The Chesapeake Bay is the largest estuary in the US, with a human population in the watershed that recently surpassed 16 million. The breeding success of peregrine falcons (*Falco peregrinus*) in the region has been poor. A study by Potter et al /181/ measured PBDE congeners (BDEs -47, -99, -100, -153, -183, -196, -197, -206, -207, -208, and -209) in the eggs of peregrines collected from shore locations in the Chesapeake Bay region between 1993 and 2002. The overall median total PBDE concentration for all egg clutches was 201 ng g^{-1}, ww, which was lower than that recently reported for peregrine eggs from the northeastern US (median 440 ng g^{-1} ww) /182/. Peregrine falcon eggs were enriched in the more highly brominated congeners, whereas BDE-47 constituted only 4.4% of the total PBDE content /181/. The sum of octa- to nona-brominated congeners (BDEs -196, -197, -206, -207, -and -208) contributed, on average,

14% of the total, whereas BDE-209 contributed 5.9% of the total PBDE concentration. Interestingly, the median BDE-209 concentrations were significantly correlated with the human population density of the area surrounding the nest, whereas total PBDEs, PCBs, and DDE were not. The concentrations of the tetra- to hexa-BDEs were similar between urban and rural sites, but the eggs from urban nests contained higher levels of highly brominated compounds.

Chen et al /182/ analyzed PBDEs in peregrine falcon eggs collected from various locations in the northeastern US (Connecticut, Massachusetts, Maine, New Hampshire, and Vermont) between 1996 and 2006. The mean PBDE concentrations (sum of 17 tetra- to deca-BDEs) in the eggs were 7660 ng g^{-1} lw. Two eggs exhibited extremely high PBDE levels (47,400 and 72,000 ng g^{-1} lw, respectively). These concentrations rival the highest PBDE burdens reported for wildlife to date. The congener profiles, especially in eggs from urban nests, were dominated by BDE-153, and were enriched in BDE-183 and -209, whereas BDE-47 contributed < 2% of the total PBDE content. The occurrence of higher BDEs in the peregrine eggs may reflect recent exposure to the octa- and deca-BDE products and/or BDE-209 debromination processes. A short half life (11 to 18 days) for BDE-209 has been reported in European starlings /183/. BDE-209 was detectable in all peregrine eggs, with concentrations ranging from 11.4 to 175 ng g^{-1} lw /182/. Five octa- and three nona-BDEs were also frequently detected, with some possibly derived from BDE-209 biodegradation. BDE-183, the marker for technical octa-BDE mixtures, was the third most prominent congener in peregrine eggs. A recent study indicated that BDE-183 is highly biomagnified in the passerine-sparrowhawk food chain /184/; thus, the elevated BDE-183 levels in peregrines could derive from its high biomagnification potential in terrestrial food chains. Peregrines living in urban environments can be exposed to deca-BDE used in thermoplastics and in textile backcoatings.

A recent study by Holden et al /185/ examined the homologue patterns of hepta-, octa-, and nona-BDEs in California peregrine falcon eggs collected between 1986 and 2007 as evidence for the biological debromination of BDE-209. The congener patterns in the peregrine eggs were characterized by high levels of BDE-153, -183, and -209. The mean levels were ΣPBDEs = 7850 ng g^{-1} lw (range 80-53000 ng g^{-1} lw); BDE-209 = 490 ng g^{-1} lw; nona-BDEs = 180 ng g^{-1} lw; octa-BDEs = 800 ng g^{-1} lw; hepta-BDEs = 950 ng g^{-1} lw; hexa-BDEs = 1950 ng g^{-1} lw; penta-BDEs = 1900 ng g^{-1} lw; and tetra-BDEs = 540 ng g^{-1} lw. Two congeners were detected in eggs (an unidentified hepta-BDE and BDE-202) that are not present in commercial mixtures. BDE-202 has been reported as a possible indicator of biotransformation in fish /31,186,187/. In addition, BDE-208 was present at 10-fold higher proportions in the eggs than in commercial mixtures /185/. The nona-hepta homologue profiles of peregrine eggs differed significantly from those of the weathered BDEs present in various abiotic matrices (sludge, sediments, and dust): BDE-207 was the major nona-BDE in eggs, whereas BDE-206 was the major nona-BDE in abiotic matrices. Thus, the observed differences in hepta-nona PBDE homologue profiles between peregrine eggs and either commercial mixtures or environmental matrices pointed to the metabolic debromination of BDE-209 in peregrine falcons or in their diet.

Ospreys (*Pandion haliaetus*) are piscivorous birds that inhabit some of the most contaminated coastal and estuarine waters of the US and Canada. In the late 1960s and 1970s, several populations were nearly decimated by organochlorine pesticide-related eggshell thinning and reproductive impairment /188/. Following restrictions, populations rebounded in the mid-1990s, but in some areas, legacy contaminants (PCBs and DDE) are still present at levels comparable to the pre-DDT era /189/, hence, the occurrence of emerging contaminants (PBDEs and perfluorinated surfactants) whose environmental concentration appear to be steadily

increasing is of concern. In 2000-2001, Rattner et al /188/ investigated PBDEs and the reproductive success of ospreys nesting at sites throughout the Chesapeake Bay. PBDE (sum of eight congeners) concentrations in eggs from regions of concern ranged from 195 to 725 ng g^{-1} ww, whereas average concentrations at reference sites ranged from 176 to 253 ng g^{-1} ww. Fledgling success among nestling ospreys was marginally adequate to sustain local populations in highly impacted areas (for example, Baltimore Harbor, the Patapsco River, and in the Anacostia and middle Potomac Rivers); this study, however, did not find conclusive evidence of a contaminant-related impact on osprey reproduction. A study by Toschik et al /189/ measured PBDEs in ospreys breeding in the Delaware Bay area. The PBDE concentrations in osprey eggs collected from the Delaware River and Bay were similar to those observed in the Chesapeake Bay osprey eggs /188/ and exhibited a north- to south gradient. The highest PBDE concentrations (sum of BDEs -47, -99, -100, -153, -154, and unidentified congeners hexa-a and hexa-c) were found in eggs from sites in the Delaware estuary (mean 572; range 442-820 ng g^{-1} ww), followed by those in Delaware Bay (mean 206; range 141-429 ng g^{-1} ww) /189/. Lower concentrations were found in eggs at coastal sites (mean 82.2; range 70.9-93.5 ng g^{-1} ww). These latitudinal trends show a decrease in PBDE concentrations in marine waters and reflect the distance from pollution sources, as well as the differences in congener mobility. BDE-47 was the predominant congener in osprey eggs, contributing 31% to 69% of the total PBDE content, followed by penta- and hexa-BDEs. BDE-47 was proportionally higher in eggs collected along the coast (further from sources), reflecting its higher volatility and greater mobility in the marine environment.

Henny et al /73/ measured PBDEs in osprey eggs collected between 2002 and 2007 along three rivers in the states of Oregon and Washington and in Puget Sound. The highest concentration of ΣPBDEs (sum of 12 BDE congeners) (geometric mean 897; range 507-1880 ng g^{-1} ww) was found in osprey eggs from the middle Willamette River (Oregon). A negative relation was observed between reproductivity and ΣPBDE concentrations in excess of 1000 ng g^{-1} ww in eggs at two river locations, suggesting that concentrations of ~1000 ng g^{-1} ww may reduce reproductive performance in ospreys. Interestingly, the ΣPBDE concentration in osprey eggs from Everett, Washington was nearly twice that of eggs from double-crested cormorants (*Phalacrocorax auritus*) that were nested side-by-side, likely owing to dietary differences (and contaminant exposure) between the species.

A monitoring study by Goodale et al /190/ measured PBDEs (tri- through deca-BDEs) in eggs collected in 2007 from 23 bird species at locations across the state of Maine. PBDE concentrations were higher in coastal birds in embayments receiving large river outflows (Penobscot Bay) and areas of high population density (Portland) than elsewhere. The highest concentrations were detected in bald eagles (8627 ng g^{-1} lw), herring gulls (2340 ng g^{-1} lw), osprey (4204 ng g^{-1} lw), and peregrine falcons (2551 ng g^{-1} lw). Relatively low levels were found in eiders (*Somateria mollissima*) (30 ng g^{-1} lw), Arctic terns (*Sterna paradisaea*) (143 ng g^{-1} lw), common terns (*Sterna hirundo*) (134 ng g^{-1} lw), least terns (*Sternula antillarum*) (110 ng g^{-1} lw), and Leach's storm petrels (*Oceanodroma leucorhoa*) (110 ng g^{-1} lw), whereas black guillemot eggs (*Cepphus grille*) had intermediate levels (439 ng g^{-1} lw). Compared with the results of other studies, the PBDE levels in Maine bald eagles (*Haliaeetus leucocephalus*) were higher than levels reported for white-tailed sea eagles (*Haliaeetus albicilla*) from Norway /191/, whereas levels in Maine herring gull eggs were three-fold lower than those reported in herring gull eggs from the Great Lakes /192/. The PBDE levels in osprey eggs were similar to those reported for osprey from British Columbia /27/, whereas PBDEs in the Maine peregrine eggs were slightly lower than those reported in /182/ for eggs from various locations in the northeastern US. PBDE concentrations in tern eggs (lipid basis) were an order of magnitude

lower than those in terns from the San Francisco Bay area /193/. Congener patterns varied widely among the 23 species, reflecting differences in trophic level, habitat and prey selectivity, and/or metabolic capacity for PBDEs /190/. In piscivorous birds (e.g., common terns and ospreys), the congener profiles showed a penta-BDE signature, whereas the eggs of peregrine falcons and herring gulls were enriched in BDE-183, -197, -203, -207, and -209. BDE-47 contributed < 5% of the total PBDE content in the Maine peregrines. Similarly, the eggs of eiders, which feed on benthic organisms (primarily mussels), were enriched in hepta-BDE 183 and nona-BDE 207. Interestingly, bald eagles are scavengers that have a varied diet of fish, seabirds, and terrestrial/marine mammals, but the congener profiles of the Maine bald eagles were similar to those of the piscivorous species, indicating a predominantly fish-based diet. In Leach's storm petrels, the congener profiles were dominated by tetra- and penta-BDEs.. As storm petrels feed 100-200 km offshore on mesopelagic fish and crustaceans, their profiles represent a global signal of PBDEs with higher volatility and mobility.

She et al /194/ investigated PBDEs in seabird eggs collected between 2000 and 2003 from various locations in the San Francisco Bay and Grays Bay, Washington. The median concentrations of PBDEs (sum of tri- through deca-BDEs) in eggs of Caspian terns (*Sterna caspia*) for 2000-2003 were 2410, 4730, 3720, and 2880 ng g^{-1} lw, respectively; in Forster's terns (*Sterna forsteri*) 1820, 4380, 5460, and 3600 ng g^{-1} lw, respectively; and in California Least terns (*Sterna antillarum brownii*) (for 2001 and 2002) 5060 and 5170 ng g^{-1} lw, respectively. In contrast, the median PBDE concentration in California Clapper rail eggs for 2001 was low (379 ng g^{-1} lw). Both the California Least terns and the Clapper rails are currently protected under the Federal Endangered Species Act of 1973. The higher PBDE concentrations in the piscivorous terns compared with the omnivorous Clapper rails suggest that fish consumption is the primary exposure pathway for PBDEs in the San Francisco aquatic food web. Although the PBDE congener profiles in seabirds were similar to those reported earlier in fish from the San Francisco Bay area /195/, low levels of BDE-183 and -209 were also detected in these egg samples. The mean PBDE levels in bird eggs (6200 ng g^{-1} lw) and fish (1000 ng g^{-1} lw) were orders of magnitude higher than the levels detected in breast milk (86 ng mL^{-1} lw) of women from the San Francisco Bay area /106,195/. The two highest concentrations (62,000 and 63,000 ng g^{-1} lw) found in two Forster's tern eggs from the upper South Bay exceeded those in the eggs of peregrine falcons from Sweden (up to 39,000 ng g^{-1} lw) /196/ and were comparable to the concentrations recently reported in peregrines from the north-eastern US (72,000 ng g^{-1} lw) /182/.

Glaucous gull (*Larus hyperboreus*) and glaucous-winged gull *(Larus glaucescens)* eggs collected in 2005 from seven colonies in Alaska were recently monitored for PBDEs /197/. The median PBDE concentrations (sum of tri- through octa-BDEs) in eggs of glaucous gulls from the Chukchi and Bering Sea were 76.8 and 92.8 ng g^{-1} lw, respectively. The median concentrations (348 ng g^{-1} lw) detected in glaucous-winged gulls from the Gulf of Alaska were an order of magnitude higher than those (61.4 ng g^{-1} lw) detected in gull eggs from the Bering Sea. The highest PBDE concentration (4130 ng g^{-1} lw) was found in a glaucous-winged gull from the Gulf of Alaska, reflecting proximity to urban sources in the southern region. Much higher PBDE concentrations (mean 3690-11131 ng g^{-1} lw) were reported in herring gull eggs from the Laurentian Great Lakes region /198/. The PBDE congener profiles in the Alaskan gulls were highly variable, both among the colonies and among the individuals, reflecting the gulls' opportunistic feeding habits. BDE-47 was the dominant congener, contributing 10% to 65% of the total PBDE content, followed by BDE-100 (7%-58%), -99 (5%-40%), -154 (2%-27%), and -49 (0.2%-21%) /197/. Gulls are scavengers

that feed on a broad range of items varying from fish to invertebrates to marine mammal carcasses and human refuse in dumps. Thus, the proximity to urbanized coastal areas of Alaska results in PBDE congener profiles in gulls that are different from those in gulls feeding in remote open ocean environments. Higher PBDE levels were found in gull eggs at a site that was historically used as a US aviation and aerospace station and is currently a Federal Aviation Agency flight service station, as well as a research station for the National Weather Service. A recent study has shown that military and scientific research stations are a significant point source of PBDE contamination in Antarctica /23/.

Elliott et al /27/ investigated trends in PBDE contamination in marine and freshwater bird eggs collected during 1979-2002 in British Columbia, Canada. The mean PBDE concentrations (sum of tri- through octa-BDEs) were highest in the eggs of great blue herons (*Ardea herodias*) (455 ng g^{-1} ww) collected in 2002 from the Fraser estuary, located downstream from an urban area of approximately 2 million people near Vancouver. The concentrations in eggs of double-crested cormorants and ospreys collected from sites of varying urban influence tended to be about half that value. The eggs of Leach's storm petrels collected from a colony on the Queen Charlotte Islands had only trace amounts of PBDEs (3.39 ng g^{-1} ww), despite having similar concentrations of PCBs and organochlorine pesticides relative to the other species. PBDE profiles in eggs were in the order: BDE-47 > -100 > -99, > -153 > -154 > - 28 > -183. High-resolution analysis of a subset of heron eggs revealed the presence of up to nine other congeners, including BDE-181, -207, and -209 (range 0.9 -1.8 ng g^{-1} ww).

McKinney et al /199/ analyzed PBDEs, hydroxylated (OH-) and methoxylated (MeO-) BDEs, in plasma of bald eagle nestlings collected in 2001-2003 from western coastal areas of North America. The mean PBDE concentrations (sum of 8 congeners) (1.78-8.49 ng g^{-1} ww) were similar in eaglets from southwestern British Columbia (BC) but lower than those from Santa Carolina Island, California (SCI), an area highly impacted by DDT and PCB contamination, and significantly higher than levels in eaglets from a reference site in northern BC. The mean concentrations of OH-PBDEs in the eaglets (sum of 14 congeners; 0.31-0.87 ng g^{-1} ww) were similarly distributed. The dominant PBDE congeners in the eaglet plasma samples were BDE-47, -99, and -100, but the SCI eaglets also contained low levels of highly brominated congeners. Higher concentrations of PBDEs (as well as a greater proportion of BDE-153) in the southwestern BC eaglets compared with those from the reference site may be related to the proximity of the southern birds to urban/ industrial source regions. The authors postulate, however, that the spatial variability is more likely related to dietary differences (namely, the predominance of piscivorous birds versus fish in the diet of the southern population). A recent study /200/ examined the relation between contaminant concentrations and bald eagle diets (trophic level), using stable isotope ratios. The results suggested that the trophic level influences the accumulation of PCBs but not that of PBDEs and OH-PBDEs in the eagles, which is possibly due to a greater metabolic depletion of PBDE congeners in eagles. In the SCI eaglets, the mean PBDE levels (30.9 ng g^{-1} ww) were 4- to 77-fold higher than in BC nestlings, indicating more recently released contaminants in the California population /199/. In contrast to the BC eaglets, the mean PBDE concentrations in the SCI eaglets were 2.5-fold higher than their PCB concentrations. Low levels of BDE-138, -154, and -183 were also found in the SCI eaglets, which may reflect their proximity to sources of release (namely, the industrialized and urbanized areas of Los Angeles). Interestingly, BDE-209 was not detected in SCI or BC eaglets, possibly due to rapid debromination in the eagles or in their prey. Ostensibly of biogenic origin, 6′-OH-BDE-49 and 6-OH-BDE-47 were found in BC nestlings, whereas only 4′-OH-BDE49 was found

in SCI eaglets. Regardless of origin, estrogenic and thyroidogenic dysfunction have been reported in laboratory rats exposed to OH-PBDEs /201,202/. MeO-PBDEs and HBCD were not found in any bird, but the polybrominated biphenyl, PBB-101, was detected in southwestern BC birds, likely due to the historic release of PBBs in this coastal area.

2.5. Marine Mammals

Marine mammals are long-lived, apex predators in marine ecosystems that accumulate relatively high concentrations of complex mixtures of POPs through the marine food chain. High PBDE levels (> 1000 ng g^{-1} lw) have been reported in marine mammals from urbanized areas of North and South American waters, and, akin to the pattern in their fish prey, congener patterns exhibit a penta-BDE signature /3/. Recent studies indicate that highly brominated BDEs are also accumulating in marine mammals, indicating exposure to the octa- and deca-BDE formulations. In certain areas, PBDEs in marine mammals surpass PCBs as the top contaminant in tissues. Given their high body burdens of complex mixtures of contaminants, the rising levels of PBDEs in marine mammals are of concern.

2.5.1. *Pinnipeds, sea otters, polar bears*. The reported concentrations of PBDEs in pinnipeds, sea otters, and polar bears from Canada, the US, and Ecuador are presented in Table 6a. A study by Ikonomou and Addison /203/ examined the factors controlling distribution of PBDEs in two populations of harbor seals (*Phoca vitulina*) from British Columbia (BC) sampled in 1991-1992 and in mother-pup pairs of gray seals (*Halichoerus grypus*) from Sable Island, Nova Scotia, sampled in 1995. For adults, the main route of exposure is through diet, reflecting various inputs (local point and non-point sources and/or long-range atmospheric transport), whereas young animals are exposed to PBDEs through placental and lactational transfer, which is selective because of differences in lipophilicity and molecular weight between compounds. The highest mean concentrations of PBDEs (mono- to hepta-BDEs) were found in harbor seals from the Strait of Georgia, BC (319 ng g^{-1} lw), reflecting industrial and municipal discharges from the greater Vancouver area, whereas the Sable Island gray seals had lower concentrations (112 ng g^{-1} lw). Although relatively remote, Sable Island is downstream and downwind of the large urbanized and industrialized St. Lawrence Basin. Harbor seals from Quatsino Sound, a remote inlet in western Vancouver Island had the lowest mean PBDE concentrations (7.8 ng g^{-1} lw). Congener profiles of all three populations were dominated by BDE-47 and were suggestive of the influence of PBDE sources in contrast to those of Arctic ringed seals (*Phoca hispida*), in which the enrichment of the less brominated congeners suggested the influence of atmospheric transport of more volatile congeners to the Arctic /24/. In the Sable Island gray seals, the PBDE transfer rate from mother to pup was 44.4% (n = 5). The transfer efficiency during lactation was shown to decline with increasing degree of bromination, as a function of increasing octanol/water partitioning (K_{ow}) values; this decline may be a consequence of the molecular size (> 600 Da), which may limit diffusion and partitioning into biological membranes /203/. Thus, hepta-BDEs and the higher molecular weight congeners appear not to be transferred efficiently from gray seal mothers to pups during lactation.

Similarly, Wolkers et al /204/ examined the accumulation and lactational transfer of PBDEs (tetra- to hexa-BDEs) in the blubber, liver, plasma, and milk samples of hooded seals (*Cistophora cristata*) collected in 2001 from the Gulf of St. Lawrence, Canada. The PBDE concentrations in hooded seal blubber were much lower than those reported in the blubber of harbor seals and beluga whales (*Delphinapterus leucas*) from the Gulf sampled in 2000 /157/, possibly owing to a short residence time in the Gulf, and/or the lag time for new contaminants to reach the deeper waters where hooded seals feed. PBDE body burdens (calculated for blubber and liver as a percentage of

body weight) were correlated with concentrations in the blubber, liver and milk, but not with blood plasma levels, indicating that plasma may not be a reliable tissue for monitoring contaminants in hooded seals. Consistent with previous observations /203/, a clear selective transfer from female blubber into the milk favored the less brominated PBDEs (up to penta-BDEs) and was associated with relatively high plasma concentrations in the females. The pups had a higher proportion of less brominated PBDEs than the females, confirming the preferential transfer of these compounds into milk. Similarly, a study by She et al /98/ compared PBDE profiles in a harbor seal mother-fetus pair and suggested that placental transfer from female blubber into cord blood was selective for the less brominated PBDEs. Another interesting finding was the substantially higher PBDE liver burden in the pups, which may reflect the relatively large liver of pups (3.8% of body weight) compared with adults (1.9% of body weight), as well as the higher lipid content in pup liver /204/. The liver burden in the pups, however, was 5-25 times higher than that of the females, suggesting a selective retention of PBDEs in pup liver. Despite lactational transfer, blubber PBDE concentrations were not significantly lower in females than in males, possibly because hooded seal females lose relatively little weight compared with other seal species, and thus excrete little of their accumulated contaminant burden during lactation.

The PBDE levels reported in marine mammals inhabiting the California coast are among the highest in the world, presumably owing to the state's stringent fire-retarding regulations /205/. She et al /98/ investigated PBDEs in harbor seals (*Phoca vitulina richardsi*) from San Francisco Bay collected between 1989 and 1998; this population has suffered significant habitat loss and degradation, including decades of environmental contamination. Average PBDE concentrations (sum of tetra- to hexa-BDEs) in the blubber of adult males and females were 2049 and 1251 ng g^{-1} lw, respectively. The congener profiles in the seals—BDE-47 (64%) > -99> -100> 153> -154—differed from those in adipose tissue of San Francisco Bay area residents /98/, which were more enriched in hexa-BDE congeners. The differences likely reflect the different PBDE exposure pathways for seals (primarily diet) and humans (diet and inhalation/ ingestion of contaminated dust). A study by Neale et al /206/ measured PBDEs (BDEs-47, -99, and -153) in blood of free-ranging San Francisco Bay harbor seals collected in 2001-2002. The mean PBDE concentration in whole blood of 33 seals was 760 ng g^{-1} lw, which is at the low end of the concentration range reported by She et al /98/ in the blubber of stranded San Francisco Bay harbor seals. The ΣPBDE concentrations in whole blood of the free-ranging seals were positively correlated with leukocyte counts and inversely correlated with erythrocyte counts. The authors proposed that these relations between hematological parameters and PBDEs could be indicative of inflammatory disease in the highly exposed seals.

Stapleton et al /207/ analyzed PBDEs (tri- to deca-BDEs) in adult male California sea lions (*Zalophus californianus*) collected between 1993 and 2003 along the California coast. The concentrations of PBDEs (sum of tri- to hepta-BDEs) detected in sea lion blubber (overall mean of 5778 ng g^{-1} lw; range: 569 to 24320 ng g^{-1} lw) were comparable to those reported in San Francisco Bay harbor seals in 1997-1998 /98/. BDE-47 contributed 55% of the total PBDEs in blubber. A higher proportion of BDE-100 relative to BDE-99 in blubber suggested a metabolic depletion of BDE-99 and a corresponding enrichment of BDE-47 in the sea lions and/ or their prey, similar to that previously demonstrated in laboratory exposure studies of carp /30/. BDE-209 was not detected in any sample. Of the 14 MeO-BDE congeners analyzed in the sea lions, 6-MeO-BDE-47 was detected in blubber at low concentrations /207/. In addition, a bromo-, chloro-cyclo heterocyclic compound 1,1′-dimethyl-tetrabromodichloro-2,2′-bipyrrole (DBP-Br_4Cl_2) was detected at concentrations rivaling the dominant congener BDE-47.

Table 6a. *Concentrations of individual PBDE congeners (ng g^{-1} lw) and mean and range of ΣPBDEs in pinnipeds, sea otters, and polar bears. Reference numbers refer to the corresponding references in the manuscript.*

Species	Tissue	Location			Age	#		PBDE congener								ΣPBDEs			Ref
Pinnipeds		**Country**	**State**		**Sex**	**Cong**	**N**	**47**	**99**	**100**	**153**	**154**	**155**	**183**	**209**	**Mean**	**Min**	**Max**	**#**
Gray seal	Blubber	Canada	Nova Scotia-Sable	1995	AF	13+	5	70	11	8.7	9.4	8.4	2.0	nr	nm	112			203
	Blubber	Canada	Nova Scotia-Sable Isld	1995	Pups	13+	5	40	3.9	2.7	1.2	1.3	0.3	nr	nm	50			203
Hooded seal	Blubber	Canada	Gulf of St. Lawrence	2001	AM	8	6	22	1.6	1.7	3.5	0.7	nm	nm	nm	33			204
	Blubber	Canada	Gulf of St. Lawrence	2001	AF	8	6	31	2.9	2.7	3.7	0.5	nm	nm	nm	44			204
	Blubber	Canada	Gulf of St. Lawrence	2001	Pups	8	6	25	1.2	1.9	1.6	0.1	nm	nm	nm	32			204
	Milk	Canada	Gulf of St. Lawrence	2001	AF	8	6	37	1.3	2.2	0.6	1.9	nm	nm	nm	45			204
	Liver	Canada	Gulf of St. Lawrence	2001	AM	8	6	nr	nr	nr	nr	nr	nm	nm	nm	69			204
	Liver	Canada	Gulf of St. Lawrence	2001	AF	8	6	nr	nr	nr	nr	nr	nm	nm	nm	83			204
	Liver	Canada	Gulf of St. Lawrence	2001	Pups	8	6	nr	nr	nr	nr	nr	nm	nm	nm	82			204
	Blood	Canada	Gulf of St. Lawrence	2001	AM	8	6	nr	nr	nr	nr	nr	nm	nm	nm	50			204
	Blood	Canada	Gulf of St. Lawrence	2001	AF	8	6	nr	nr	nr	nr	nr	nm	nm	nm	200			204
	Blood	Canada	Gulf of St. Lawrence	2001	Pups	8	6	nr	nr	nr	nr	nr	nm	nm	nm	149			204
Harbor seal	Blubber	Canada	BC-Quatsino Sound	1991-92	Mixed	13+	7	19	3.5	1.8	1.8	0.69	0.38	0.09	nm	28			203
	Blubber	Canada	BC-Strait of Georgia	1991-92	Mixed	13+	13	225	54	17	15	1.6	0.71	0.14	nm	319			203
	Blubber	US	Gulf of Maine	2000-05	Pups	41	13	2974	308	146	99	42	30	14	0.8	3645	80	25720	213
	Blubber	US	Gulf of Maine	1991-05	Y	41	14	1730	335	124	558	95	85	20	1.9	2945	250	23513	213
	Blubber	US	Gulf of Maine	2000-05	AM	41	7	904	134	49	210	31	45	20	1.1	1385	454	3827	213
	Blubber	US	Gulf of Maine	2000-05	AF	41	8	190	46	18	51	6.0	6.3	20	0.5	326	131	713	213
Gray seal	CSF [b]	US	Massa.-Cape Cod	2004-05	Pup M	35 [a]	1 [a]	nd	0.17 [a]	nd	nd	nd	nd	nd	nd	0.17 [a]			214
Harbor seal	Blubber	US	SF Bay California	1989-98	AM	5	6	1617	100	89	137	106	nm	nm	nm	2049	88	8325	98
	Blubber	US	SF Bay California	1989-98	AF	5	4	833	131	85	63	139	nm	nm	nm	1251	473	2985	98
	Blood	US	SF Bay California	2001-02	Mixed	3	33	nr	nr	nm	nr	nm	nm	nm	nm	760			206
	Blubber	US	So. California Coast	1994-06	Pup	14	8	1784	373	119	98	17	7	nd	nm	2430	320	7200	209
	Blubber	US	So. California Coast	1994-06	AM	14	1	203	101	36	29	15	1	nd	nm	370			209
No. elephant seal	Blubber	US	So. California Coast	1994-06	Pup	14	16	214	56	29	5	5	2	nd	nm	360	40	2010	209
California sea lion	Blubber	US	So. California Coast	1994-06	Pup	14	12	37276	5760	7327	997	1327	287	15	nm	51100	2900	236000	209
	Blubber	US	So. California Coast	1994-06	Y	14	16	25057	3659	4396	549	789	169	8	nm	36500	60	184000	209
	Blubber	US	So. California Coast	1994-06	SA	14	15	11624	1759	2224	362	515	92	9	nm	16900	2150	84600	209
	Blubber	US	So. California Coast	1994-06	AM	14	5	29147	9868	10399	3042	3400	618	104	nm	55300	3430	194000	209
	Blubber	US	So. California Coast	1994-06	AF	14	15	3153	607	813	152	210	42	7	nm	5150	270	16000	209

Species	Tissue	Country	Location/State	Year	Age	#		PBDE congener								ΣPBDEs			Ref
					Sex	Cong	N	47	99	100	153	154	155	183	209	Mean	Min	Max	#
California sea lion	Blubber	US	California Coast	1993-03	M	27	26	nr	nr	nr	nr	nr	nr	nr	nd	5778	569	24343	207
Spotted seal	Blood	US	Alaska-Bristol Bay	2000-01	M	3	2	nr	nr	nm	nd	nm	nm	nm	nm	0.45	0.03	0.89	210
	Blood	US	Alaska-Bristol Bay	2000-01	F	3	4	nr	nr	nm	nd	nm	nm	nm	nm	0.35	0.10	0.71	210
	Blubber	US	Alaska-Bering Sea	2003	M	11	1	nd	nd	nd	nd	nd	nd	nd	nm	5.5 [a]			211
	Blubber	US	Alaska-Bering Sea	2003	F	11	2	2.8 [a]	nd	nd	nd	nd	nd	nd	nm	16 [a]	11	21	211
Bearded seal	Blubber	US	Alaska-Bering Sea	2003	M	11	3	nd	nd	nd	nd	nd	nd	nd	nm	3.2 [a]	1.7	4.9	211
	Blubber	US	Alaska-Bering Sea	2003	F	11	2	nd	nd	nd	nd	nd	nd	nd	nm	3.7 [a]	3.3	4.1	211
Ringed seal	Blubber	US	Alaska-Bering Sea	2003	M	11	3	nd	nd	nd	nd	nd	nd	nd	nm	3.6 [a]	1.40	6.8	211
	Blubber	US	Alaska-Bering Sea	2003	F	11	3	nd	nd	nd	nd	nd	nd	nd	nm	8.1 [a]	6	10.5	211
Ribbon seal	Blubber	US	Alaska-Bering Sea	2003	M	11	4	nd	nd	nd	nd	nd	nd	nd	nm	17 [a]	6	39	211
	Blubber	US	Alaska-Bering Sea	2003	F	11	2	nd	nd	nd	nd	nd	nd	nd	nm	15 [a]	13	17	211
Galapagos sea lion	Blubber-muscle	Ecuador	Galapago Islands	2005	Pup	4	1	33.3						0.63		35			215
Polar Bear, Otter																			
Polar bear	Adipose	Canada	West Hudson Bay	1999-02	F	36	15	9.9	1.9	0.49	1.2	nr	nr	nr	nm	14	4.3	46	219
	Adipose	US	Alaska-Bering Chukchi Sea	1999-02	F	36	8	4.9	0.78	0.20	0.11	nr	nr	nr	nm	6.7	4.6	11	219
	Adipose	US	Alaska-Bering Chukchi Sea	1999-02	M	36	7	5.2	1.4	0.22	<0.08	nr	nr	nr	nm	6.8	2.9	17	219
	Liver	US	Alaska-Beaufort Sea	1993-00	AM	8	7	12	nd	nd	nd	nd	nm	nm	nm	12	<2	23	217
	Liver	US	Alaska-Beaufort Sea	1999	AF	8	1	16	nd	nd	nd	nd	nm	nm	nm	16			217
	Liver	US	Alaska-Chukchi Sea	1994-02	AM	8	14	10	nd	nd	nd	nd	nm	nm	nm	10	<2	32	217
Alaskan sea otter	Liver	US	Alaska-Prince William Snd	1995-98	A	?	3									1027			216
	Liver	US	Washington-Clallam County	1995-98	A	?	6									909			216
California sea otter	Liver	US	California-Monterey Bay	1995-98	A	?	6									2423			216
Southern sea otter	Liver	US	California Coast	1992-02	AF	5	80	Nr	nr	nr	nr	nr	nm	nm	nm	2170	10	26800	77

[a] Concentrations expressed in ng g^{-1} wet weight; [b] CSF=cerebrospinal fluid; A = adult, SA = subadult, Y = yearling, M = male, F = female; nd = not detected, nr = not reported, nm = not measured, SF = San Francisco

The concentrations of DBP-Br_4Cl_2 were positively correlated with 6-MeO-BDE-47, but not with BDE-47, suggesting that these compounds originated from such natural sources as marine algae and sponges /208/.

A study by Meng et al /209/ measured PBDEs in California sea lions, Pacific harbor seals, and northern elephant seals (*Mirounga angustirostris*) collected between 1994 and 2006 from the southern California coast. The highest mean concentrations (sum of 14 BDE congeners) were detected in adult male California sea lions (55300 ng g^{-1} lw, range 3430-194000 ng g^{-1} lw), followed by sea lion pups (51100 ng g^{-1} lw; range 2900-236000). These concentrations are an order of magnitude higher than those reported in California sea lions by Stapleton et al /207/ and are the highest reported PBDE levels in marine mammals to date. By comparison, the respective mean PBDE concentrations in the harbor seals were 2,430 and 370 ng g^{-1} lw for pups and adult males /209/. The lowest concentrations were found in northern elephant seal pups (360 ng g^{-1} lw). Whereas the three species have a similar diet, this concentration gradient likely reflects differences in home range, and thus exposure to contaminant sources.

California sea lions are near-shore feeders inhabiting industrialized coastal embayments having elevated contaminant levels (for example, the Los Angeles/ Long Beach Harbor complex). Harbor seals also inhabit the coastal zone but are known to spend days feeding in the open ocean, whereas northern elephant seals are highly pelagic feeders in deeper waters and may range as far north as Alaska. The significantly lower concentrations in adult females compared with adult males suggested reproductive offloading of PBDEs in mature females. Although BDE-47 dominated the congener profiles, the proportion of BDE-100 in the sea lions exceeded that of BDE-99, whereas the harbor seals contained twice as much BDE-99 as -100, implying species differences in metabolism/ elimination capacity for BDE-99.

A pilot study by Neale et al /210/ analyzed the concentrations of BDEs-47, -99, and -153 in the blood of free-ranging spotted seals (*Phoca largha*) collected in 2000-2001 from Bristol Bay, Alaska. Only BDEs-47 and -99 were detected in seal blood. The average PBDE concentrations ranged from 0.35 to 0.45 ng g^{-1} lw in females and males, respectively. That these levels were three orders of magnitude lower than those in blood of San Francisco Bay harbor seals may reflect open-ocean foraging and feeding at a lower trophic level in this species. Whereas harbor seals feed in coastal waters, Alaskan spotted seals are pelagic feeders that are strongly associated with pack ice in the Bering Sea and inhabit the shoreline only in summer months.

Quakenbush /211/ measured PBDEs in the blubber of four species of seals that are consumed by humans and polar bears in the Alaskan Bering Sea: ringed, spotted, bearded (*Erignathus barbatus*), and ribbon seals (*Phoca fasciata*). Of the 38 congeners analyzed, only 11 mono- to tetra-BDEs were detected in blubber, and the average concentrations were low, ranging from 3.4 ng g^{-1} ww in bearded seals to 16.5 ng g^{-1} ww in ribbon seals. Interestingly, the dominant congeners in blubber were BDE-30 and -2; penta- to hexa-BDEs were not detected. BDE-47 was detected only in spotted seals, a species that forages at a higher trophic level compared with the other species. The low concentrations and the predominance of mono- to tri-BDEs in these seals suggest that PBDEs have not yet reached the pelagic waters of the Bering Sea.

Along the US northwestern Atlantic, coastal urban development has produced some of the densest concentrations of human populations in North America, and POP contamination has been a concern since the 1950s. In the 1980s, Massachusetts Bay and Long Island Sound, New York were ranked as having more highly contaminated sites than any other coastal state or region in the US /212/. Few studies, however, have examined PBDE contamination in marine mammals from this region.

A study by Shaw et al /213/ reported the occurrence of PBDEs in harbor seals (*Phoca vitulina concolor*) inhabiting the northwest Atlantic (from the Gulf of Maine to Long Island, New York. PBDEs (mono- to hexa-BDEs) were measured in blubber samples collected between 1991 and 2005. The ΣPBDE concentrations detected in blubber ranged from 80 to 25720 ng g^{-1} lw, (overall mean 2403 ± 5406 ng g^{-1} lw; n = 42). By age, the mean ΣPBDE concentrations were: 3645, 2945, 1385, and 326 ng g^{-1} lw in pups, yearlings, adult males, and adult females, respectively. Although not mother-pup pairs, the higher levels in the pups suggested reproductive offloading of PBDE burdens from females. Unlike the spatial trend previously observed for PCBs, no decreasing south-north gradient from urban to rural/remote areas was observed for PBDEs, likely reflecting inputs from diffuse local sources across the harbor seal range. BDE-47 was the dominant congener in blubber, but the profiles varied by age. In adult males, hexa-BDEs (BDE-153, -154, and -155) contributed more to the total PBDEs (22%) than did the penta-BDEs-99 and -100 (14%), whereas the pattern in the pups suggested a selective placental and lactational transfer of BDE-47 and to a lesser degree, BDEs-99 and -100, but very limited transfer of hexa-BDEs. Hepta-BDE-183, the marker for the octa-BDE mixture, and octa-BDE 197 were detected in blubber, along with several unidentified hepta- and octa- congeners. BDE-209 was detected in blubber at measurable (above trace) concentrations (range 1.1 to 8 ng g^{-1} lw). The observed PBDE accumulation pattern suggested exposure to all three commercial PBDE mixtures and/or BDE-209 debromination processes in harbor seals or their prey fishes.

Montie et al /214/ measured PBDEs and OH-PBDEs in the cerebrospinal fluid (CSF) of a male gray seal pup collected in 2004-2005 from Cape Cod, Massachusetts. Of the PBDE congeners analyzed (tri- to deca-BDEs), only BDE-99 was detected in CFS of the gray seal pup at very low concentrations (0.17 ng g^{-1} ww). In addition, trace levels of 6'-OH-BDE-49 were detected in CSF. Given the developmental neurotoxic potential of PBDEs and their OH-metabolites in laboratory studies (reviewed in /44/), the presence of these compounds in the central nervous system of marine mammals is of concern.

Few studies have investigated PBDEs in marine mammals from Central or South America. A recent study by Alava et al /215/ investigated PBDEs in muscle-blubber biopsy samples of sea lion pups (*Zalophus wollebaeki*) live captured in the Galapagos Islands, off the coast of Ecuador. Of 21 pups, only one male pup exhibited detectable concentrations of four congeners, including BDE -47, -49, -66, and -183 (sum PBDE 35 ng g^{-1} lw), with BDE-47 representing the dominant congener. Similarly, the study found low concentrations of PCBs in the pups, dominated by low molecular weight PCB congeners, suggesting that global transport is the main source of POPs in Galapagos sea lions. The results suggest that at present, local sources are relatively small and thus, PCB and PBDE concentrations in the pups may largely reflect global rather than local contamination. The study also indicates that despite their remote location, the Galapagos Islands are not immune from the threat of global pollutants.

Sea otters. Sea otters (*Enhydra lutris*) are considered important indicators of local contamination in coastal marine environments because they are non-migratory and their diet is composed of sessile and slow-moving invertebrates. Kannan et al /216/ measured PBDE levels in the liver of two sea otter subspecies collected during 1995-1998 from the coastal waters of California, Washington, and Alaska. The concentrations of PBDEs in sea otter livers varied widely (88-26800 ng g^{-1} lw). The highest concentrations of PBDEs were found in southern sea otters (*Enhydra lutris nereis*) from urbanized areas of central California (mean 2423 ng g^{-1} lw); these concentrations were two-fold greater than those in northern sea otters (*Enhydra lutris kenyonis*) from Washington and Prince Island Sound, Alaska (mean 909, and 1027

ng g^{-1} lw, respectively). The spatial trends in PBDE concentrations in sea otters were correlated with the human population density in these areas. PBDE concentrations in the Alaskan sea otters were two orders of magnitude greater than those reported in livers of Alaskan polar bears *(Ursus maritimus)* /217/, reflecting the sea otters' proximity to sources and a diet dominated by benthic organisms (for example, clams, mussels). BDE-47 was the dominant congener in sea otters from all locations. Sea otters from Prince William Sound contained a high proportion of BDE-99, suggesting current exposure to penta-BDE mixtures in this region of Alaska. Another study /77/ examined the relations between infectious disease in southern sea otters and exposure to PBDEs and other halogenated contaminants. The results suggested that elevated PCBs and perfluorochemicals (but not PBDEs) were strongly associated with disease status in the sea otters. Additive effects caused by exposure to complex mixture of toxicants, including PCBs and PBDEs, on sea otter health have been suggested.

Polar bears. Polar bears are apex predators of the Alaskan Arctic marine ecosystem and feed primarily on ringed and bearded seals, but occasionally on larger prey such as walruses (*Odobenus rosmarus*) and beluga whales. Because of their high trophic level in the food chain and long life span (25-30 years), polar bears are exposed to elevated levels of POPs.

Kannan et al /217/ examined the concentrations and profiles of PBDEs in the livers of two subpopulations of polar bears collected by native subsistence hunters between 1993 and 2002 from the (northern) Beaufort Sea and (western) Chukchi Sea, Alaska. The PBDE concentrations in livers of Alaskan polar bears ranged from < 2 to 32 ng g^{-1} lw; mean range 10-16 ng g^{-1} lw. These concentrations were 10-fold lower than those reported in plasma of Norwegian polar bears /218/. In contrast to the distribution of other organohalogenated compounds (for example, PCBs), the mean PBDE concentrations in the northern (12 ng g^{-1} lw) and western populations (10 ng g^{-1} lw) were not significantly different /217/. Age and gender were not significant covariates for PBDE concentrations. PBDE levels decreased with age in both male and female polar bears, a pattern similar to that observed in humans /93/, likely reflecting the relatively recent introduction of PBDEs (less than 30 years) and the metabolism and elimination of PBDEs by polar bears. Of the eight PBDE congeners analyzed (BDE-28, -30, -47, -85, -99, -100, -153, and -154), only BDE-47 was found in polar bear livers. Recent studies /218,219/ reported that the PBDE pattern in polar bears is dominated by BDE-47, presumably due to a greater accumulation of this congener and/or to BDE-99 debromination /30/. The metabolic transformation of PBDEs by polar bears has been suggested by the occurrence of hydroxy- and methoxy-PBDEs in polar bear plasma /218/.

Muir et al /219/ determined PBDE concentrations in the adipose tissue of polar bears collected by subsistence hunters between 1994 and 2002 in several Arctic regions, including the Bering-Chukchi Sea, northwestern Alaska. The respective mean PBDE concentrations in female and male Alaskan bears were 6.7 and 6.8 ng g^{-1} lw. These levels were similar to those in bears from the Canadian Arctic but much lower than levels in bears from Svalbard, Norway or East Greenland. Only four congeners (BDE-47, -99, -100, and -153) were consistently identified in polar bear fat. BDE-47 was the dominant congener, contributing 65%-82% of the total PBDE content in fat. Higher proportions of BDE- 99, -100, and -153 were found in polar bears from the Canadian Arctic than in those from Svalbard or the Bering-Chukchi Sea, possibly owing to the greater use of penta-BDE formulations in North America. The congener patterns in polar bears, however, are subject to discrimination among isomers during long-range transport, as well as during bioaccumulation and metabolism, thus, source signatures could be lost /219/.

2.5.2 *Cetaceans*. The reported concentrations of PBDEs in various cetacean species from the US, Canada, and Brazil are shown in Table 6b. The first study to report PBDE concentrations in marine

mammals from US coastal waters examined the carcasses of bottlenose dolphins (*Tursiops truncatus*) that stranded during a mass mortality event in 1987-1988 along the US Atlantic coast from New Jersey to Florida /220/. The PBDE levels in the blubber of three female dolphins ranged from 180 to 220 ng g^{-1} lw (mean 200 ng g^{-1} lw). In 1990, a second mortality event occurred among bottlenose dolphins in the Gulf of Mexico /221/. The mean PBDE concentrations were 3110 and 190 ng g^{-1} lw in the blubber of adult males and females, respectively. The highest PBDE concentration (20000 ng g^{-1} lw) was detected in an adult female.

Ikonomou et al /222/ measured the PBDE concentrations in blubber samples collected in 1991-1993 from stranded male harbor porpoises (*Phocoena phocoena*) from British Columbia, Canada. The concentrations of PBDEs (sum of 13 congeners) ranged from 360 to 2300 ng g^{-1} lw (mean 868 ng g^{-1} lw). Congener profiles in blubber reflected the proximity of the harbor porpoise range to urban and industrial centers (for example, Vancouver) on the west coast of British Columbia that are source areas for the penta-BDE formulation.

Compared with populations in the Canadian Arctic, beluga whales in the St. Lawrence Estuary, Quebec, Canada are relatively remote and more highly contaminated by POPs owing to their higher trophic level and their habitat, which is located downstream of the St. Lawrence River and Lake Ontario, a highly populated and industrialized area. Lebeuf et al /223/ analyzed PBDEs (tri- to hepta-BDEs) in the blubber of adult beluga whales found on the shores of the St. Lawrence Estuary between 1988 and 1999. The respective mean concentrations of ΣPBDEs (sum of 10 congeners) in St. Lawrence Estuary belugas whales collected between 1997 and 1999 were 430 and 540 ng g^{-1} lw in adult males and females. Three predominant congeners (BDE-47, -99, and -100) contributed, on average, 75% of the total PBDE content in blubber. Several congeners were proportionally higher in females (BDE-49, -66, and -99), whereas BDEs-155 and -183 were proportionally higher in males. The lack of a significant difference in PBDE contamination between males and females was attributed to the rapidly increasing PBDE levels in this population. The PBDEs eliminated through reproductive off-loading in females may be negligible relative to their recent intake.

Another study /224/ determined the concentrations of PBDEs, OH-PBDEs, and MeO-PBDEs in liver samples collected in 2000 to 2003 from beluga whales in the St. Lawrence Estuary. The PBDE concentrations (sum of 9 congeners) in the liver of the St. Lawrence Estuary belugas (mean 2210 ng g^{-1} lw) were two orders of magnitude higher than those in belugas from the Western Hudson Bay in the Canadian Arctic. The dominant PBDEs (BDEs-47, -99, -100) in liver were similar to those reported in blubber /223/, comprising 40%, 20%, and 20% of the total PBDE concentrations in beluga liver, respectively. Two OH-PBDEs were detected at trace levels in the St. Lawrence Estuary belugas, whereas two MeO-PBDE isomers (4′-MeO-BDE-17 and 6-MeO-BDE-47) were present at concentrations ranging from 20 to 100 ng g^{-1} lw /224/. The results suggest that these metabolites are likely from a natural origin and are accumulating the St. Lawrence Estuary beluga whale food web. Given the endocrine-disrupting properties of PBDEs and their metabolites, the authors conclude that PBDE exposure may be having a negative impact on the health of the St. Lawrence Estuary beluga whale population.

In the northeastern Pacific, three ecotypes of killer whales (*Orcinus orca*) have been studied in the coastal waters of Canada and the US—residents, transients, and offshore killer whales /225/. Resident killer whales are further divided into northern and southern groups and forage primarily on a combination of Chinook salmon and local fishes. Transient killer whales range widely along the coast and primarily consume marine mammals. Offshore killer whales are widely distributed from Alaska to California, and their preferred prey species are higher trophic level fish /226/. Rayne et al /227/ investigated PBDEs in blubber biopsy

Table 6b. *Concentrations of individual PBDE congeners (ng g^{-1} lw) and mean and range of ΣPBDEs in cetaceans. Reference numbers refer to the corresponding references in the manuscript.*

Species	Tissue	Country	Location / State	Age		#		PBDE congener								Sum			
				Year	Sex	N	Cong.	47	99	100	153	154	155	183	209	Mean	Min	Max	#
Beluga whale	Blubber	Canada	St. Lawrence Estuary	1988-99	AF	26	10	174	73	40	19	22	2.2	1.5	nm	540	300	1060	223
	Blubber	Canada	St. Lawrence Estuary	1988-99	AM	28	10	151	53	33	16	25	3.1	2.4	nm	430	170	780	223
	Liver	Canada	St. Lawrence Estuary	2000-03	Mixed	6	9	nr	Nr	nr	nr	nr	nm	nr	nm	2210	246	3030	224
	Liver	Canada	West Hudson Bay	2000-03	Mixed	11	9	nr	Nr	nr	nr	nr	nm	nd	nm	53	2	183	224
Harbor porpoise	Blubber	Canada	BC .-Vancouver	1991-93	M	5	34	470	163	125	15	26	3	nm	nm	868	350	2300	222
Killer whale-North. Res	Blubber	Canada	Northeast Pacific	1993-96	M	13	12	136	21	25	3.5	7.7	0.98	nr	nr	203	27	758	227
	Blubber	Canada	Northeast Pacific	1993-96	F	8	12	253	71	57	11	15	1.6	nr	nr	415	23	2828	227
Killer whale-South. Res	Blubber	Canada	Northeast Pacific	1993-96	M	5	12	597	108	168	15	41	2.1	nr	nr	942	242	1812	227
Killer whale-Transient	Blubber	Canada	Northeast Pacific	1993-96	M	6	12	766	76	117	15	27	2.8	nr	nr	1014	283	2250	227
	Blubber	Canada	Northeast Pacific	1993-96	F	7	12	623	74	117	20	38	4.1	nr	nr	885	162	2896	227
Killer whale-Resident	Blubber	US	Alaska-Cent. Aleutian Isl.	2003-04	AM	3	10	nr	nr	nr	nr	nr	nm	nr	nm	36	nd	57	226
	Blubber	US	Alaska-East. Aleutian Isl.	2003-04	AM	20	10	nr	nr	nr	nr	nr	nm	nr	nm	50	nd	110	226
	Blubber	US	Gulf of Alaska	2003-04	AM	17	10	nr	nr	nr	nr	nr	nm	nr	nm	120	nd	270	226
Killer whale-Offshore	Blubber	US	Alaska	2003-04	AM	4	10	nr	nr	nr	nr	nr	nm	nr	nm	3300	2200	4400	226
Killer whale-Transient	Blubber	US	Alaska-East. Aleutian Isl.	2003-04	AM	15	10	nr	nr	nr	nr	nr	nm	nr	nm	790	93	2100	226
	Blubber	US	California Coast	2003-04	AM	1	10	nr	nr	nr	nr	nr	nm	nr	nm	12600			226
Killer whale-South. Res	Blubber	US	Puget Snd-Georgia Basin	2006	AF	1	10	nr	nr	nr	nr	nr	nm	nr	nm	7500			225
	Blubber	US	Puget Snd-Georgia Basin	2006	AM	8	10	nr	nr	nr	nr	nr	nm	nr	nm	6800	3300	15000	225
Short-beak. common dol.	CSF[b]	US	Massachusetts-Cape Cod	2004-05	AF	2	35	0.58[a]	nd	0.06[a]	nd	nd	nd	nd	nd	0.64[a]	0.34	0.94	214
White-sided dolphin	CSF[b]	US	Massachusetts-Cape Cod	2004-05	AM	2	35	0.77[a]	0.04[a]	0.24[a]	nd	0.2[a]	nd	nd	nd	1.1[a]	0.74	1.6	214
White-sided dolphin	CSF[b]	US	Massachusetts-Cape Cod	2004-05	AF	6	35	13[a]	nr	nr	nd	nd	nd	nd	nd	13[a]	nd	37	214
	Brain	US	Massachusetts-Cape Cod	2004-05	AF	3	35	27[a]	7[a]	11[a]	2[a]	4[a]	nd	nr	nd	nr			214
	Blubber	US	Massachusetts-Cape Cod	1993-00	Juv	23	6	1460[a]	290[a]	390[a]	120[a]	150[a]	nm	nd	nm	2410[a]			232
	Blubber	US	Massachusetts-Cape Cod	1993-00	AM	15	6	990[a]	220[a]	350[a]	120[a]	150[a]	nm	nd	nm	1820[a]			232
	Blubber	US	Massachusetts-Cape Cod	1993-00	AF	9	6	260[a]	85[a]	110[a]	74[a]	83[a]	nm	nd	nm	610[a]			232
Rough-toothed dolphin	Blubber	US	Florida-Gulf of Mexico	1997	Juv	7	6	690[a]	140[a]	260[a]	67[a]	210[a]	nm	nd	nm	1360[a]			232
	Blubber	US	Florida-Gulf of Mexico	1997	AF	6	6	260[a]	56[a]	100[a]	25[a]	77[a]	nm	nd	nm	510[a]			232
Bottlenose dolphin	Blubber	US	South Atlantic Coast	1987	F	3	?									200	180	220	220
	Plasma	US	South Carolina-Charleston	2004	Mixed	20	12	11[a]	1.7[a]	4.4[a]	0.9[a]	1.7[a]	nm	nr	nm	21[a]	16	26	230
	Plasma	US	Florida-Indian Riv. Lagoon	2004	Mixed	33	12	1.7[a]	0.14[a]	0.85[a]	0.1[a]	0.4[a]	nm	nd	nm	3.9[a]	2.2	5.6	230
	Blubber	US	South Carolina-Charleston	2003	AM	31	13	4110	341	1380	268	687	nm	nd	nm	6830	1711	13167	229

Bottlenose dolphin	Blubber	US	South Carolina-Charleston	2003	AF	5	13	246	62	66	61	106	nm	nd	nm	565	429	904	229
	Blubber	US	South Carolina-Charleston	2003	Juv M	5	13	5120	480	1628	178	394	nm	nd	nm	7850	2680	22800	229
	Blubber	US	South Carolina-Charleston	2003	Juv F	8	13	4220	430	1070	126	355	nm	nd	nm	6260	1756	15600	229
	Blubber	US	Florida-Indian Riv. Lagoon	2004	AM	25	13	1040	89	353	59	125	nm	nd	nm	1690	463	3790	229
	Blubber	US	Florida-Indian Riv. Lagoon	2004	AF	14	13	367	68	118	47	64	nm	nd	nm	696	196	1410	229
	Blubber	US	Florida-Indian Riv. Lagoon	2004	Juv M	11	13	617	67	178	38	58	nm	nd	nm	979	594	1920	229
	Blubber	US	Florida-Indian Riv. Lagoon	2004	Juv F	6	13	945	87	287	43	94	nm	nd	nm	1480	224	3560	229
	Blubber	US	Florida-N. Biscayne Bay	2002-04	AM/Juv	16	14	1338	165	563	91	147	nr	nr	nr	2385			231
	Blubber	US	Florida-N. Biscayne Bay	2002-04	AF	3	14	48	8	17	12	15				102			231
	Blubber	US	Florida-S. Biscayne Bay	2002-04	AM/Juv	15	14	527	56	839	128	217	nr	nr	nr	873			231
	Blubber	US	Florida-S. Biscayne Bay	2002-04	AF	3	14	35	1.8	10	1.7	4.1				60			231
	Blubber	US	Florida-Indian Riv. Lagoon	2001-04	Mixed	5	12	712	73	189	34	81	nr	28	<.02	1130	30	4500	25
	Blubber	US	Florida-Gulf of Mexico	1991-96	Mixed	8	12	175	16	41	11	29	nr	9	0	363	86	585	25
	Blubber	US	Florida-Gulf of Mexico	2000-01	Mixed	6	12	678	86	258	75	164	nr	<0.01	<.02	1270	200	4160	25
	Blubber	US	Texas-Gulf of Mexico	1990	Calf	6	?									313	35	771	221
	Blubber	US	Texas-Gulf of Mexico	1990	Juv	5	?									1900	560	3410	221
	Blubber	US	Texas-Gulf of Mexico	1990	AF	6	?									190	36	432	221
	Blubber	US	Texas-Gulf of Mexico	1990	AM	10	?									3110	126	16300	221
Striped dolphin	Blubber	US	Florida Coast	1994	AF	1	12	250	67	89	53	155	nr	4.7	<.02	660			25
Tucuxi dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	13	9	nr	nr	nr	nr	nr	nr	nd	nr	670	260	1620	18
	Liver	Brazil	Rio de Janeiro	1994-06	AF	6	9	nr	nr	nr	nr	nr	nr	nd	nr	160	13	450	18
	Liver	Brazil	Rio de Janeiro	1994-06	Newb	1	9	nr	nr	nr	nr	nr	nr	nd	nr	242			18
Atlantic spotted dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	6	9	nr	nr	nr	nr	nr	nr	nd	nr	1150	360	2440	18
	Liver	Brazil	Rio de Janeiro	1994-06	AF	1	9	nr	nr	nr	nr	nr	nr	nd	nr	96			18
False killer whale	Liver	Brazil	Rio de Janeiro	1994-06	Newb	2	9	nr	nr	nr	nr	nr	nr	nd	nr	3600	1210	5960	18
Bottlenose dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	3	9	nr	nr	nr	nr	nr	nr	nd	nr	960	270	1350	18
Rough-toothed dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	1	9	nr	nr	nr	nr	nr	nr	nd	nr	360			18
	Liver	Brazil	Rio de Janeiro	1994-06	AF	2	9	nr	nr	nr	nr	nr	nr	nd	nr	1150	700	1600	18
Common dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	1	9	nr	nr	nr	nr	nr	nr	nd	nr	240			18
	Liver	Brazil	Rio de Janeiro	1994-06	AF	1	9	nr	nr	nr	nr	nr	nr	nd	nr	125			18
Pan-tropical spotted dol.	Liver	Brazil	Rio de Janeiro	1994-06	AM	1	9	nr	nr	nr	nr	nr	nr	nd	nr	1215			18
Spinner dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AF	1	9	nr	nr	nr	nr	nr	nr	nd	nr	150			18
Striped dolphin	Liver	Brazil	Rio de Janeiro	1994-06	?	1	9	nr	nr	nr	nr	nr	nr	nd	nr	210			18
Fraser's dolphin	Liver	Brazil	Rio de Janeiro	1994-06	AM	2	9	nr	nr	nr	nr	nr	nr	nd	nr	22	15	28	18
	Liver	Brazil	Rio de Janeiro	1994-06	AF	7	9	nr	nr	nr	nr	nr	nr	nd	nr	7	3	14	18

[a] Concentrations expressed as ng g^{-1} wet weight; [b] CSF=Cerebrospinal Fluid; A = adult, Juv = juvenile, Newb = newborn, M = male, F = female; nd = not detected, nr = not reported, nm = not measured

samples collected between 1993 and 1996 from free-ranging killer whales belonging to three communities: southern residents from the industrialized Puget Sound-Georgia Basin area of British Columbia and Washington State; northern residents off northern Vancouver Island; and transients ranging from the south of Puget Sound-Georgia Basin to Alaska. The mean PBDE concentrations (sum of tri- to hexa-BDEs) in the male southern resident killer whales (942 ng g^{-1} lw) were similar to those in the male and female transients (1015 and 885 ng g^{-1} lw, respectively), but were two- to five-fold higher than the levels in the male and female northern residents (203 and 415 ng g^{-1} lw, respectively) /227/ The differences in PBDE concentrations between northern and southern residents can be attributed to the influence of localized contaminant sources, as southern residents spend more time in the industrialized areas of the Georgia Basin and Puget Sound, and part of their diet consists of contaminated local fishes.

In addition to proximity to source, the PBDE concentrations in transient killer whales are influenced by their high trophic level diet. Male transients, which consume seals and porpoises and occasionally spend time near urbanized areas, had ΣPBDE concentrations approximately equal to those of the southern residents /227/. BDE-47 was proportionally higher in the transients, akin to patterns in their marine mammal prey, whereas BDE-100 and -154 were proportionally higher in the southern residents. The lack of gender-based differences in PBDE concentrations in the transients and northern residents may reflect a masking of reproductive offloading by females as PBDEs continue to increase in the northeastern Pacific marine ecosystem /223,228/. Krahn et al /226/ measured PBDE concentrations in blubber/ epidermis biopsy samples collected in 2003-2004 from free-ranging adult male killer whales from Alaska. The respective mean PBDE concentrations were 6695, 3300, and 76 ng g^{-1} lw in transients, offshore whales, and northern residents. The higher PBDE concentrations in the transients can be explained by their consumption of marine mammals, whereas the levels in offshore whales may reflect exposure to contaminated prey along the urbanized California coast. Compared with PBDE concentrations reported in killer whales by Rayne et al /227/, the levels reported in this study were 6.5 times higher in transients and 2.5 times lower in northern residents /226/.

A study by Fair et al /229/ measured PBDEs (13 tri- to hepta-BDEs) in blubber biopsy samples collected in 2003 and 2004 from free-ranging bottlenose dolphins from the Charleston Harbor estuary, North Carolina, and the Indian River Lagoon on the east coast of Florida. The mean ΣPBDE concentrations (sum of six congeners) were significantly higher in the Charleston (CHS) dolphins (5860 ng g^{-1} lw) than in the Indian River Lagoon (IRL) dolphins (1260 ng g^{-1} lw). The highest PBDE concentration (22780 ng g^{-1} lw) was found in a male CHS dolphin. Charleston is a highly urbanized estuary with multiple inputs from such possible PBDE sources as wastewater treatment facilities, whereas the Indian River Lagoon is an expansive 156-mile estuary along the Florida coast that has experienced a reduction in wastewater treatment discharges since 1996 /229/. The PBDE concentrations in the adult male CHS dolphins were comparable to the levels detected in resident adult male killer whales from the Puget Sound-Georgia Basin of the northeastern Pacific in 2004/2006 /225/. PBDE levels in the male CHS dolphins are an order of magnitude higher than those reported in the blubber of bottle-nose dolphins that stranded during a mass mortality in 1987-1988 /220/ and also exceed the blubber concentrations reported during the 1990 dolphin mortality event in the Gulf of Mexico /221/. In dolphins from both sites, PBDE congener profiles (BDE-47> -100> -154> -99> -153> -28) were suggestive of penta-BDE exposure /229/. Gender differences were observed in concentrations at both sites, with significantly lower levels in females that were indicative of maternal transfer of PBDE to

their calves. For the CHS dolphins, juveniles also had significantly higher levels than the females, highlighting the importance of maternal offloading of PBDEs. Unlike the trend for lipophilic POPs, age was not a significant influence on PBDE levels, which may relate to the relatively recent introduction of PBDEs and/or to metabolism/ excretion in adult dolphins. The lack of an increasing PBDE trend with age was also observed in polar bears /217,219/.

A recent study /230/ determined PBDEs and OH-PBDEs in the plasma of free-ranging bottlenose dolphins from the same sites along the southeastern Atlantic. The plasma PBDE concentrations (sum of 12 congeners) of resident CHS dolphins (arithmetic mean 30 ng g^{-1} ww) were almost six times higher than those in the IRL dolphins (5.45 ng g^{-1} ww). The congener profiles in plasma were similar to those found in blubber /229/. Whereas an age-related trend in blubber was lacking, the mean PBDE concentrations in plasma were negatively correlated with age at both locations /230/. The mean concentrations of OH-PBDE congeners were almost two-fold higher in the CHS dolphins. 6-OH-BDE-47 was the predominant congener at both locations. The lack of correlations between OH-PBDEs and the five major PBDE congeners implied natural sources (for example, marine sponges and algae) and/or combined contributions from natural and anthropogenic (biotransformation of parent PBDEs) sources. The results suggested that, compared with OH-PCBs, OH-PBDEs in dolphin plasma are minor products, and a significant proportion may result from the dietary uptake of naturally produced MeO- and OH-PBDEs.

Johnson-Restrepo et al /25/ investigated PBDE contamination in blubber samples collected between 2000 and 2004 from bottlenose dolphins that stranded on the western (Gulf of Mexico) and eastern (Indian River Lagoon) coasts of Florida, and a striped dolphin (*Stenella coeruleoalba*) from the Florida west coast (Sarasota). The respective mean concentrations of ΣPBDEs were 1190 and 660 ng g^{-1} lw in bottlenose and striped dolphin blubber. The PBDE concentrations and congener profiles were similar in dolphins from the east and west coasts and to the levels and patterns reported by Fair et al /229/ in blubber biopsy samples of free-ranging bottlenose dolphins from the Indian River Lagoon. A study by Litz et al /231/ examined the fine-scale spatial variation of PBDEs (BDE-47, -85, -99, -100, -153, and -154) in blubber biopsy samples collected between 2002 and 2004 from free-ranging bottlenose dolphins in Biscayne Bay, Florida. Adult males and juveniles (combined) had significantly higher mean PBDE concentrations (1465 ng g^{-1} lw) than females (76 ng g^{-1} lw). The concentrations in the males were slightly higher than those reported in bottlenose dolphins from the Indian River Lagoon /25,229/, but lower than the PBDE levels in dolphins from the Charleston Harbor estuary, North Carolina. The PBDE congener profiles were similar between north and south Biscayne Bay dolphins, but the PBDE concentrations in the male northern dolphins (2385 ng g^{-1} lw) were almost three-fold higher than those in the southern portions of the Bay (873 ng g^{-1} lw), likely due to the high levels of urban and industrial development around the city of Miami.

Tuerk et al /232/ investigated PBDEs in blubber samples of Atlantic white-sided dolphins (*Lagenorhynchus acutus*) collected between 1993 and 2000 during mass stranding events along the Massachusetts coast, near Cape Cod, and rough-toothed dolphins (*Steno bredanensis*) collected in 1997 during a single mass stranding along the Gulf coast of Florida. In both species, the respective mean PBDE concentrations (sum of BDE-47, -99, -100, -153, and -154) were highest in juveniles (2410 and 1360 ng g^{-1} lw in white-sided dolphins and rough-toothed dolphins), followed by the white-sided dolphins adult males (1820 ng g^{-1} lw). Females had significantly lower PBDE levels than males and juveniles. PBDE congener profiles differed between the species. BDE-47 dominated the profiles but was proportionally higher in white-sided dolphins, whereas BDE-154 was proportionally higher in rough-toothed dolphins, likely

reflecting species differences in the metabolism/ elimination of PBDEs. Montie et al /214/ measured PBDEs and OH-PBDEs in the cerebrospinal fluid (CSF) of stranded short-beaked common dolphins (*Delphinus delphis*) and in CSF and cerebellum gray matter (GM) (of Atlantic white-sided dolphins collected in 2004-2005 from a similar geographic location (Cape Cod, Massachusetts). In two female short-beaked common dolphins, only trace levels of BDE-47 and BDE-100 were detected in CSF. The CSF of the Atlantic white-sided dolphins contained BDE-47, -99, -100, and -153, with BDE-47 as the dominant congener. In the three white-sided dolphins in which both CSF and cerebellum GM were analyzed, the levels of all detected PBDE congeners were higher in the cerebellum GM, including BDE-28, -47, -99, -100, -153, -154, and -183. The higher concentrations in cerebellum GM likely reflect the higher lipid content of cerebellum GM compared with CSF. OH-PBDEs were also detected in dolphin brain tissues. Trace levels of 6′-OH-BDE 49 were detected in the CSF of both species; in addition, 6-OH-BDE-47 was present in dolphin CSF. In Atlantic white-sided dolphins, cerebellum GM contained detectable levels of 6′-OH-BDE-49 and 4′-OH-BDE-49 but not 6-OH-BDE-47. Interestingly, in cerebellum GM the concentration of 4′-OH-BDE-49 (which was not detected in CSF) was ~50 times greater than that of 6′-OH-BDE-49. PBDE concentrations reported in the blubber of white-sided dolphins /232/ were orders of magnitude higher than the levels found in CSF and cerebellum GM, which may be a function of the decreased lipid content of brain tissue and the restricted accumulation of PBDEs across the blood-brain barrier. Given the neurodevelopmental toxicity of PBDEs, the detection of these compounds in dolphin CSF and cerebellum GM is of concern.

Published reports on PBDE contamination in cetaceans from Central and South America are scarce. A recent study by Dorneles et al /18/ measured PBDEs and MeO-PBDEs in the liver of various stranded cetacean species, including Guiana dolphins (*Sotalia guianensis*), Atlantic spotted dolphins (*Stenella frontallis*), false killer whales (*Pseudo crassidens*), bottlenose dolphins, rough-toothed dolphins, common dolphins, pan-tropical spotted dolphins (*Stenella attenuata*), spinner dolphins (*Stenella longirostris*), striped dolphins, and Fraser's dolphins (*Lagenodelphis hosei*) collected between 1994 and 2006 from waters near Rio de Janeiro, southeastern Brazil. Guiana dolphins inhabit the Guanabara Bay estuary located in a highly industrialized coastal area bordered by four cities (Rio de Janeiro metropolitan area) with a total population of about 11 million people. The remaining nine dolphin species inhabit either the continental shelf (CS) or oceanic environments frontal to Guanabara Bay. A PBDE concentration range of 3 to 5960 ng g^{-1} lw was found in dolphin liver, similar to that observed in Northern Hemisphere cetaceans. The highest mean PBDE concentrations were found in false killer whale calves (3600 ng g^{-1} lw), adult male pan-tropical spotted dolphins (1215 ng g^{-1} lw), adult male Atlantic spotted dolphins (1150 ng g^{-1} lw) and adult female rough-toothed dolphins (1150 ng g^{-1} lw), whereas the lowest PBDE concentrations were detected in male and female Fraser's dolphins (22 and 7 ng g^{-1} lw, respectively). Three of the species with elevated PBDE concentrations (false killer whales, Atlantic spotted dolphins, and rough-toothed dolphins) inhabit the continental shelf, while pan-tropical spotted dolphins are oceanic.

Interestingly, Guiana dolphins from the Guanabara Bay estuary had intermediate mean PBDE levels (670 and 160 ng g^{-1} lw in males and females, respectively). BDE-47 was the dominant congener in all dolphin profiles, contributing 32%-80% of the total PBDE content, with the exception of the Fraser's dolphin, a species of extreme oceanic habitat. BDE-47 was the only congener detected in two of the nine Fraser's dolphins analyzed. Tri-BDE-28 was the second most abundant congener in these dolphins, suggesting a significant influence of atmospheric transport of the less-brominated PBDEs to open ocean food webs. In the (estuarine)

Guiana dolphins, the congener pattern–BDE-47 > -100 > -99 > -154 > -153 > -28 > -85–was indicative of the possible use of penta-BDE commercial mixtures in Brazil. The differences in PBDE congener profiles among estuarine, CS, and oceanic dolphins could be attributed to a skewing of PBDEs in oceanic waters in favor of more volatile congeners. A relatively high BDE-28/-47 ratio, however, was observed in the Guiana (estuarine) dolphins, suggesting the influence of species-specific metabolism for individual PBDE congeners.

The MeO-PBDE concentrations in the liver of CS dolphins were among the highest detected to date in cetaceans (up to 250 μg g^{-1} lw). ΣMeO-PBDEs were significantly higher in the CS and oceanic dolphins than in estuarine dolphins. Further, the ratio of two naturally-produced MeO-PBDEs (2′-MeO-BDE-68 and 6-MeO-BDE-47) was also higher in CS dolphins than in estuarine dolphins, indicating that the continental shelf dolphins receive MeO-PBDEs predominantly from sponges or associated organisms. Interestingly, 65% of the CS delphinids were residing in a coastal region strongly influenced by upwelling phenomenon, which may contribute to the transport of MeO-PBDEs from the benthic to the pelagic food chain. Squids, which constitute important prey for CS dolphins, execute vertical diel migrations and may also play a role in the benthic-pelagic transport of MeO-PBDEs in the food web.

High concentrations of MeO-PBDEs were also reported in cetaceans from Queensland, Australia /233/, suggesting there may be more intense biosynthesis of MeO-PBDEs and/or greater bioavailability of these compounds to nektonic invertebrates in tropical regions. The ratio of ΣPBDEs/ΣMeO-PBDEs also varied significantly by habitat, with mean values of 7.12, 0.08, and 0.01 for estuarine, CS, and oceanic dolphins, respectively, indicating a clear shift in the contribution of anthropogenic PBDEs and naturally produced MeO-PBDEs to the total PBDEs in dolphin liver /18/. In two Guiana dolphin pairs, the fetus/ mother ratios calculated for BDE congeners revealed a reduced transplacental transfer for higher-brominated compounds. Further, the mean values of the fetus/mother ratios were negatively correlated with the degree of bromination of the congeners, indicating that placental transfer in the dolphins may be selective due to the K_{ow} value and molecular size of the congener /203/. PBDE concentrations in female Guiana dolphins were significantly lower than in males and decreased with female age, reflecting the effect of reproductive offloading to offspring.

2.6 Biomagnification of PBDEs in Marine Food Webs

PBDEs are often elevated in species at the top of food webs, which clearly points to biomagnification. Only a few studies, however, have specifically examined the transfer of PBDEs through aquatic/marine food chains. Increasing concentrations of ΣPBDEs (on a lipid-weight basis) were observed in a Florida coastal food web in the order: forage fish (silver perch, striped mullet, small spotted seatrout) and Atlantic stingrays < predator fish (red drum, hardhead catfish, spiny dogfish) < Atlantic sharpnose sharks < bull sharks and bottlenose dolphins /25/. The ΣPBDE concentrations in sharks and dolphins were one to two orders of magnitude greater than those in the lower trophic level fishes. The biomagnification factors (BMFs) for ΣPBDEs, calculated as the ratio between lipid normalized concentrations in predator and prey, ranged, on average, from 3 to 85, indicating a high potential for biomagnification in this food web. The highest BMFs of ΣPBDE were measured from forage fish (silver perch) to bottlenose dolphins (150) and bull sharks (204). Bull sharks are apex predators that inhabit estuarine, near-shore, and offshore waters of both the Gulf and the Atlantic coasts of Florida /25/. These sharks are the only shark species to penetrate far into fresh-water habitats. BDE-209 was biomagnified in the sharks, whereas only trace levels of BDE-209 were detected in the bottlenose dolphins. The respective mean concentrations of

BDE-209 in spiny dogfish, Atlantic sharpnose sharks, and bull sharks (17514, and 778 ng g^{-1} lw) were several orders of magnitude higher than those in the teleost fishes, and accounted for 60% of the total PBDE content in shark species.

A recent study /32/ examined biomagnification of ΣPBDEs and individual BDE congeners in a northwest Atlantic marine food web (from the Gulf of Maine to the New York coast). The PBDE concentrations in adult male harbor seals were two orders of magnitude higher than those detected in teleost fishes, indicating a high potential for biomagnification. The BMFs calculated for ΣPBDEs between seven species of teleost fishes and harbor seal blubber ranged, on average, from 17 to 76. For three fish species comprising the majority (70%) of the harbor seal diet (silver hake, white hake, herring), the BMFs for ΣPBDEs averaged 36, 33, and 17, respectively. Comparable BMFs for ΣPBDEs were reported between prey fishes and predators (bottle-nose dolphins and bull sharks) in Florida coastal waters /25/, between teleost fishes and harbor seals in the North Sea (38, 30, and 27 for silver hake (whiting), herring, and Atlantic cod, respectively) /164/, between prey fishes (sole and whiting) and harbor seals and harbor porpoises from the southern North Sea (range 0.6 to 53) /234/, and between polar cod and harbor seals (12) and ringed seals (37) in Svalbard, Norway /16, 235/. Increasing BMFs for BDE congeners with degree of bromination (up to hepta-BDEs) were observed between fish and harbor seals /32/. Hexa-BDEs were highly biomagnified in seal blubber, with BMFs ranging from 148 to 677, 11 to 447, and 12 to 236 for BDE -153, -154, and -155, respectively. Similarly, Weijs et al /234/ reported increasing BMFs for hexa-BDEs in adult male harbor seals and porpoises, implying a low metabolic capacity for these bioaccumulative congeners. The BMFs for BDE-47 and BDE-99 from fish to harbor seals were highly variable among the seven fish species /32/. BDE-99 was highly biomagnified from white hake, winter flounder, and American plaice to harbor seals (BMFs 213, 53.2, and 33.9, respectively), and BDE-100 was relatively more abundant than BDE-99 in these species. Because BDE-99 is *meta-para*-substituted and can not be easily metabolized by seals /236/, the high biomagnification from white hake to seals could indicate a significant debromination of BDE-99 in this species, as previously described in teleost fish /30,187/.

A relative dominance of BDE-100 resulting from the metabolic conversion of BDE-99 to BDE-47 has also been reported in fish /30,237/. Biomagnification was not observed for tetra BDEs -49, -66, and -75, and was very low for the tri-BDE-28, suggesting that harbor seals may possess an efficient metabolism for these congeners /32/. The concentrations of BDE-183 and -197 were two to four-fold higher in the seals than in fish, implying trophic transfer in the food web. In contrast, the BDE-209 concentrations in harbor seal blubber (range: 1 to 8 ng g^{-1} lw) were similar to those in the fish (range: 0.2 to 4 ng g^{-1} lw), indicating a low biomagnification potential for this congener (BMF ≤ 1). Whether the lack of biomagnification of BDE-209 in marine mammals is a result of a low uptake rate for this large molecule (log K_{ow} 9-10) or debromination processes is unclear /5,16,157,164/. Uptake efficiencies of < 1% for BDE-209 have been reported in feeding studies on teleost fishes /30,187/; this value is several orders of magnitude lower than the accumulation efficiency of other PBDEs, and could explain the low concentrations of BDE-209 in fish, including species foraging on benthic organisms. BDE-209 debromination in fish may also account for a lack of biomagnification for BDE-209 and the trophic enrichment (higher BMFs) of the less brominated PBDE congeners /30,187/. A feeding study of juvenile lake trout /187/ demonstrated that the BMFs of PBDEs were likely to be much higher in fish that are exposed to the less brominated BDEs as well as BDE-209 than fish that are not exposed to BDE-209. In seals, a laboratory exposure study demonstrated that BDE-209 is slowly accumulated in blubber through diet /238/. Gray seals were fed deca-BDE spiked oil capsules (12 μg d^{-1}) for one

month. At the end of the study (after 29 days on a deca-free diet), 11%-15% of the ingested BDE-209 was stored in the blubber. Blubber concentrations in the gray seals (2 to 8 ng g^{-1} lw) continuously exposed to deca-BDE were similar to those observed in the blubber of wild harbor seals /213/, suggesting that more or less continuous exposure may be occurring through the marine food web. Also it is possible that BDE-209 may preferentially partition to tissues other than blubber in marine mammals. In the rat, BDE-209 is only marginally distributed to adipose tissue but may be associated with blood proteins and migrates to perfused tissues such as the liver /239/. Further studies are needed on the uptake, kinetics, and tissue distribution of BDE-209 in marine organisms.

Whereas most of the studies mentioned here have reported biomagnification of PBDEs as the ratio of lipid normalized concentrations in selected tissues of predators to prey items, future studies should measure whole body burdens of contaminants in predators and prey items. Nevertheless, this task is daunting, especially for large predatory species like dolphins and whales. For predatory species, we recommend that concentrations and burdens (concentration X mass of the tissue) in each tissue be measured and summed to obtain whole body burdens; such values will support a more accurate assessment of biomagnification factors.

Assessing the trophodynamics of PBDEs in aquatic/marine food webs is even more difficult, and discrepancies between studies are unavoidable owing to multiple factors that may influence biomagnification, including the degree of ambient contamination, environmental conditions (for example, water temperature), food web length, physiochemical properties of congeners (for example, octanol-water partition coefficients [log K_{ow}], volatility, solubility, and partitioning behavior) and species-selective biotransformation (biodegradation and bioformation).

Recent studies have examined the biomagnification of PBDEs for entire food webs using trophic magnification factors (TMFs) based on the relation between lipid-normalized PBDE concentrations and relative trophic level (TL) measured through stable isotope ($\delta^{15}N$) analysis. Whereas most of the recalcitrant PCBs were found to have TMFs > 1, a limited biomagnification of PBDEs was reported in a Lake Winnipeg (freshwater) food web /240/, a Bohai Bay marine food web (north China) /237/, a Tokyo Bay coastal marine food web (Japan) /166/, and two Canadian Arctic marine food webs /241, 242/. In most studies, only a few PBDE congeners (BDE-28, -47, -99, -100 and -154) were significantly biomagnified through the entire aquatic/marine food webs, although biomagnification was shown between individual feeding relationships /237,240,241/. TMFs > 1 for BDEs -47, -100, and -154 were measured in aquatic species from a highly contaminated freshwater food web near an electronic waste recycling site in south China /243/, whereas no biomagnification was observed for BDEs -99, -183, and -209, suggesting that an enhanced metabolic capacity of highly exposed fishes may result in trophic dilution of these congeners. In contrast, Law et al /240/ reported that only BDE-99, BDE-209, and decabromodiphenylethane (DBDPE) biomagnified throughout a (less contaminated) lower trophic-level Lake Winnipeg food web. In a coastal marine food web in Tokyo Bay, the bioconcentration factors for PBDEs increased as the octanol-water partition coefficient rose to log $K_{ow} = 7$, above which BMFs decreased as K_{ow} increased, presumably as a function of restriction of permeation through cell membranes caused by larger molecular sizes /166/. BDEs-47, -99, -100, -153, and -154 were biomagnified (up to fish), indicating that PBDE congeners biomagnify with increasing hydrophobicity up to hexa-BDEs, after which biomagnification may decrease. In contrast, Wu et al /243/ reported that TMFs were correlated with log K_{ow} of PCBs but not PBDEs in a highly contaminated south China food web, suggesting that enhanced metabolism in highly exposed species may play a greater role than octanol-water partition coefficients in the food-web transfer of PBDEs. Weijs et al /234/

reported significant species differences in the biomagnification of PBDEs between harbor seals and harbor porpoises, both apex predators in a southern North Sea food web, reflecting large species differences in the ability to metabolize PBDE congeners. Age was also found to significantly influence the biomagnification potential, whereas other factors, including the octanol-water partition coefficients of PBDEs and TL, were found to be of lesser importance in predicting biomagnification.

The influence of physiochemical properties on PBDE food-web transfer and geographical dispersal was investigated in a recent study of a near-shore estuary of the southern North Sea (Norway) /244/. Whole-body burdens of the most abundant PBDEs biomagnified with increasing trophic position in the food web and were higher in pelagic compared with benthic organisms (invertebrates and fish). Such differences were particularly evident for the less brominated and more volatile congeners, suggesting an atmospheric gas-water exchange of volatile compounds over the water surface. With the exception of zooplankton, the burdens of PBDEs including BDE-209 in aquatic marine biota from the high Arctic (Svalbard, Norway) were comparable to or exceeded those in the North Sea organisms, likely reflecting the significant accumulation of particle-associated PBDEs by sympagic (sea-ice associated) invertebrates and fish in pristine polar waters. The intake of sympagic zooplankton could also explain the high relative proportion of BDE-209 (10%-20%) in zooplankton and polar cod from Svalbard /244/. High body burdens of PBDEs, including BDE-209, were also observed in marine zooplankton and in polar cod from the Canadian Arctic /245/.

In certain aquatic/marine food webs, an apparent lack of biomagnification in top predator species has been reported, presumably owing to an increasing metabolic depletion of PBDE congeners with trophic level /200,242/. In nesting bald eagles from pelagic, freshwater, and estuarine sites in British Columbia and coastal California food webs, TL was correlated with an increase in PCBs, but not PBDEs, suggesting a greater rate of PBDE biotransformation in the eagles. Evidence of PBDE metabolism was supported by the finding of OH-PBDEs in the nestling plasma /199/. Similarly, PBDE concentrations decreased with increasing TL in seabirds and beluga whales in a Canadian Arctic food web, and MeO-BDEs were the predominant PBDEs in various organisms, contributing, for example, approximately 90% of the total PBDE burden in beluga whales /246/. In a polar bear food chain in Svalbard, Norway, most PBDEs biomagnified as a function of TL from zooplankton to polar cod to ringed seals /235/. Only BDE-153, however, was found to increase from the ringed seal to the polar bear, indicating that polar bears are able to metabolize and biodegrade most BDE congeners. In contrast, Muir et al /219/ reported the biomagnification of five PBDE congeners (BDE-47, -99, -100, -153, and -154) from ringed seal to polar bears (BMFs > 1) from four geographically distinct regions including Alaska, the Canadian Arctic, East Greenland, and Svalbard. BDE-153 exhibited the highest biomagnification (average BMF = 71), from ringed seal to polar bear, confirming that BDE-153 is a highly bioaccumulative compound in marine biota.

In the above mentioned studies, it is apparent that biotransformation and bioformation of PBDEs are variable among species and differences in source PBDEs are likely to vary between systems and populations. Whereas most studies show that the most abundant PBDEs readily biomagnify in aquatic/marine food webs, the magnification of PBDE congeners could be underestimated due to biotransformation or overestimated due to bioformation kinetics. Thus, all biomagnification parameters must be viewed with caution when comparing the food web dynamics of PBDEs. Possibly, in animal bodies, some PBDE congeners are more readily transformed or metabolized than others. The lack of a significant age-related increase in PBDE concentrations in certain species suggests that PBDEs are biotransformed more rapidly (relative to PCBs) in some organisms

2.7 Temporal Trends

Temporal trend studies have shown that PBDE levels in biota and humans in North America have been increasing exponentially over the last 30 years, with a doubling time of ~4-6 years /3,22, 95,105/. Following restrictions on penta-BDE production in the 1990s, PBDE levels began to decline in Europeans /247/ and may be leveling off in the US /87/. Current PBDE concentrations in tissues of North Americans, however, are ~20-100 times higher than levels in people anywhere else in the world. These higher levels reflect the use of 95% of the world's production of the penta-BDE product in the US and Canada.

Since the 1970s, PBDEs have increased substantially in marine biota from North America (Figure 2). In Dungeness crab collected near pulp and paper and urbanized sites in the coastal waters of British Columbia, total PBDE concentrations, particularly BDE-47, increased steadily between 1994 and 2002 /154/. This trend is consistent with earlier studies by this group /24/ indicating that PBDE levels increased more than an order of magnitude in ringed seals from the Canadian Arctic between 1981 and 2000. This trend was driven by an exponential increase in tetra-, penta-, and hexa-BDEs, with doubling times of 8.6, 4.7, and 4.3 years, respectively. PBDE levels were predicted to surpass those of PCBs in ringed seals

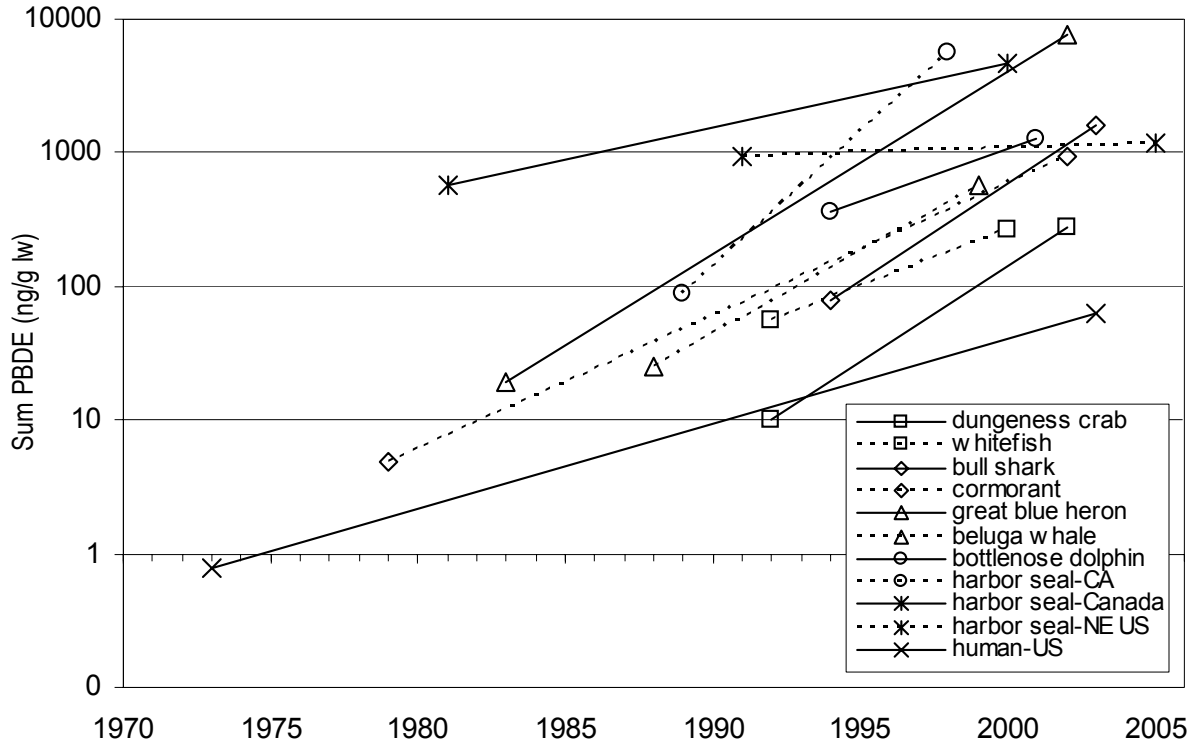

Fig. 2: Temporal trends of total PBDEs in humans and marine biota from North America 1970-2005. Trend lines show average PBDE concentrations reported for the first and last year of the studies. References: Humans US /105/; whitefish /158/; cormorant /27/; great blue heron /27/; Dungeness crab /154/; bull shark /25/; beluga whale /223/; bottlenose dolphin /25/; harbor seal-San Francisco Bay, CA /98/; harbor seal-British Columbia, Canada /24/; harbor seal- NW Atlantic US /213/

by 2050. A similar trend was observed in mountain whitefish samples from the Canadian sections of the Columbia River system, where the doubling period was only 1.6 years and PBDE concentrations were predicted to overtake PCBs by 2006 /158/. Elliott et al /27/ reported increasing levels of PBDEs in marine and freshwater birds from British Columbia. PBDEs increased exponentially between 1983 and 2002 in blue heron eggs from the Fraser River estuary and in double-crested cormorant eggs from the Strait of Georgia marine ecosystem between 1979 and 2002. A doubling time of 5.7 years was observed for both species.

Krahn et al /225/ investigated PBDE contamination in blubber/epidermis biopsy samples from three pods of free-ranging southern resident killer whales collected in 2004 and 2006 from the Puget Sound-Georgia Basin area. This population was listed as endangered in the US and Canada following a 20% decline between 1996 and 2001. The mean levels of PBDEs (sum of 10 tri- to hexa-BDEs) were 7500 and 6800 in females and males, respectively. These concentrations are an order of magnitude higher than those detected in male southern residents from this community 10 years earlier /227/. Overall, the data indicate that PBDEs are increasing in transients and southern residents from the industrialized PS-GB area, whereas the concentrations in northern residents are leveling off or declining.

Lebeuf et al /223/ reported that ΣPBDE concentrations increased exponentially between 1988 and 1999 in the blubber of both male and female beluga whales from the St. Lawrence Estuary, with a doubling time for the predominant congeners of ~ 3 years (Figure 2). This rapid increase could explain the similar PBDE concentrations measured in both males and females, thus, masking the effect of reproductive offloading in the females.

The PBDE concentrations in marine biota from the California coast are among the highest in the world and are doubling as rapidly as every 2-4 years in some species. Kimbrough et al /150/ reported significant increases in PBDEs in sediments and blue mussels *(Mytilus* spp*)* from various US coastal areas over the period 1994-2007. The highest increases were observed in sediments and mussel tissue from Anaheim, southern California. Lunder and Sharp /103/ reported that PBDE levels more than doubled in halibut and more than tripled in striped bass from the San Francisco Bay between 1997 and 2002. Marginal increases in PBDE concentrations were also found in other marine fishes, for example, white croakers and leopard sharks. She et al /98/ reported that PBDE concentrations increased by almost two orders of magnitude in adult harbor seals from the San Francisco Bay between 1989 and 1998, with a doubling time of 1.8 years.

A 10-year time trend analysis (1993-2004) showed that PBDEs were exponentially increasing in Florida coastal marine predators, with an estimated doubling time of 2-3 years for bull sharks and 3-4 years for bottlenose dolphins (Figure 2) /25/. Temporal trends in dolphins from South America were recently reported by Dorneles et al /18/. PBDE concentrations in male Guiana (estuarine) dolphins from Guanabara Bay, Rio de Janeiro, southeastern Brazil, were positively correlated with the year of stranding, suggesting that concentrations were increasing in these dolphins between 1994 and 2006. The PBDE signature in the dolphins (BDE-47 > -99 > -100) suggested an unrestricted use of the commercial penta-BDE mixture in Brazil, which may explain the increasing PBDE concentrations in these dolphins.

Several recent studies have indicated a lack of a temporal trend in the PBDE concentrations found in marine biota, suggesting that the levels may have reached equilibrium in certain marine ecosystems. No temporal trends were observed in PBDE concentrations in aquatic bird eggs collected from the San Francisco Bay between 2000 and 2003 /193/. Chen et al /182/ reported a lack of significant change in the concentrations of PBDEs or individual congeners in peregrine falcon eggs from the northeastern US between 1996 and 2006, with the exception of BDE-209, which increased

with a doubling time of 5 years. The increase in BDE-209 in peregrine eggs may reflect recent inputs of the commercial deca-BDE mixture to the peregrine food web. Shaw et al /213/ reported a lack of a significant temporal trend in ΣPBDE concentrations in harbor seals from the northwest Atlantic between 1991 and 2005, although the congener compositions shifted in harbor seals over time. The percent contribution of BDE-47 increased while the proportion of BDE-153 decreased between 1991 and 2000; these trends leveled off between 2000 and 2005. BDE-99 concentrations decreased only slightly from 1991 to 2000. Similarly, Ikonomou et al /24/ reported that the concentrations of BDEs -47, -99, and -100 increased in male ringed seals from the Canadian Arctic between 1981 and 1996, but increases in the levels of BDE-99 were slowing considerably between 1996 and 2000. Similar changes were reported in gull egg samples from the Great Lakes collected between 1981 and 2000 /192/. Such changes probably reflect differences in the use or the composition of the various commercial PBDE formulations over the years.

Tuerk et al /232/ compared PBDE concentrations in juvenile Atlantic white-sided dolphins across the year of collection and found no significant temporal trend between 1993 and 2000, suggesting that a lag period may exist for higher concentrations to be detected in pelagic marine mammals, or that the concentrations may have peaked in this species before 1993. Stapleton et al /207/ reported the lack of any temporal trend in ΣPBDE concentrations in California sea lions between 1993 and 2003, although the levels of HBCD were increasing. Meng et al /209/ recently reported the lack of a significant change in ΣPBDE concentrations in pinnipeds from the southern California coast between 1994 and 2006. Similarly, a lack of a temporal trend in PBDEs was observed in polar bears from various regions (Alaska, the Canadian Arctic, East Greenland, Svalbard) between 1994 and 2002 /219/.

A study by Kannan et al /77/ reported the lack of an increase in total PBDE concentrations in California sea otters between 1992 and 2002. Yet, the ratio of PBDEs to PCBs concentrations increased significantly during this decade, suggesting that the rate of increase of PBDEs exceeded that for PCBs in sea otters. The predominance of PBDEs over PCBs has also been reported in human adipose tissue /93/. Kajiwara et al /248/ reported a 150-fold increase in ΣPBDEs in adult female northern fur seals (*Callorhinus ursinus*) collected from the Japanese coast between 1972 and 1994, but a 50% decrease in levels between 1994 and 1998. Collectively, the data suggest that PBDE levels were increasing in marine mammals between the 1970s and the mid-1990s, but may have stabilized or reached equilibrium over the past decade. It is noteworthy that none of the above-mentioned studies has shown a significant decrease in PBDE concentrations in North or South American marine biota. The lack of temporal increase in PBDE levels in certain species could also suggest faster elimination than uptake rates in some organisms.

Thus, the trend for PBDEs follows in the footsteps of that reported for PCBs and, to a lesser extent, for DDT/DDE in marine biota. In the northwestern Atlantic, PCB levels in marine mammals are reported to be consistently higher than DDT levels. This finding has been explained by a more rapid decline of DDT in the environment after these compounds were banned in the 1970s /249/, whereas PCBs are still being released from stockpiled residues /250/. This observation is consistent with temporal trends in many temperate areas of the northern hemisphere. In industrialized areas, the PCB levels in marine biota and sediments have remained constant or declined only slightly since the late 1980s, reflecting equilibrium in environmental cycling /38,251,252/. Tanabe /19/ calculated that in the mid 1980s, only 30% of all PCBs produced had thus far dispersed into the environment. Estimates were that by the late 1980s,

only about 1% of all PCBs had reached the oceans, whereas about 30% had accumulated in dumpsites and sediments of rivers, coastal zones, and estuaries /250/. Based on the likely future dispersal of PCBs into the oceans, Tanabe /19/ concluded that global PCB levels in marine biota are unlikely to decline in the near future, and not before 2010-2030.

Similarly for PBDEs, considering the existing stockpiles and growing indoor/outdoor reservoirs of the compounds in North America and the continued, substantial production and loading of deca-BDE into aquatic and coastal ecosystems, the possibility that PBDE levels in the ocean environment will decline for years, even decades, to come is unlikely.

3. NEW FLAME RETARDANTS AND NATURALLY OCCURRING BROMINATED COMPOUNDS

Before PBDEs came into use, PBBs were used as flame retardant additives in synthetic fibers and molded plastics. In 1973, when PBBs were inadvertently mixed into livestock feed in Michigan, thousands of animals either died or were destroyed. In 1973 and 1974, thousands of Michigan residents were exposed to meat, milk, butter, cheese, and eggs contaminated with PBBs. The production of PBBs was ceased by a manufacturer in 1974, although certain other formulations, such as octabromobiphenyl and decabromobiphenyl, were produced until the late 1970s (http://ntp.niehs.nih.gov/ntp/roc/eleventh/profiles/s148pbb.pdf). The occurrence of several hundreds to thousands of ng $^{-1}$ lw of PBBs in the blubber of bottlenose dolphins from the Florida coast was reported in the early 1990s /221/.

With the decline in PBB production, the brominated flame retardant industry turned to PBDEs as a replacement. PBDEs became a popular product in the 1990s. In the early 2000s, several governments banned the use of penta- and octa BDE products, and the major US manufacturer of these materials (Great Lakes Chemical) voluntarily stopped the production of these two products. The deca-BDE product is, however, still in use. The continuing use of deca-BDE is controversial as this form persists in the environment. A few studies have shown that deca-BDE is metabolically or photochemically degraded/ transformed to less brominated congeners /186, 253/. BDE-209 is widely distributed in sediments and indoor dusts, where it often dominates BFR profiles, and evidence is increasing of its accumulation in terrestrial and aquatic food chains. Future studies should examine the temporal trends of BDE-209 and its degradation products in marine organisms.

Several other non-regulated alternatives to PBDEs exist. A few studies have reported their environmental occurrence and, in some cases, an increase in concentrations over time. Tetrabromobisphenol A (TBBPA) and 1,2,5,6,9,10-hexabromocyclododecane (HBCD) are the two other most widely used flame retardants. The market demand and environmental levels for these two compounds have been increasing, although at concentrations much lower than those for PBDEs /254,255/. Studies have reported the occurrence of TBBPA and HBCD in US coastal organisms /32,256/. HBCD was detected in 87% of the fish samples collected from the Gulf of Maine at concentrations ranging from 2.4 to 38.1 ng g^{-1} lw (overall mean 17.2 ± 10.2 ng g^{-1} lw /32/). The concentrations of HBCD in the blubber (n = 57) and liver (n = 16) from Atlantic white-sided dolphins that stranded on the eastern coast of the US between 1993 and 2004, ranged from 19-380 ng g^{-1} lw and 2.9-140 ng g^{-1} lw, respectively /257/. In general, the concentrations of TBBPA and HBCD in marine mammals and fish from the US coast were below 50 ng g^{-1} lw. The highest concentrations of TBBPA and HBCD reported for bull shark from the Florida coast were 36 and 413 ng g^{-1}, lw, respectively. Temporal increases in the concentrations of HBCD have been reported for California sea lions /207/.

Studies have revealed increases in HBCD concentrations with time in marine mammals from the UK /258/. Tomy et al /63/ reported the biomagnification of HBCD in a Lake Ontario food chain. The biomagnification factors of HBCD varied

from 0.2 to 10. Overall, the concentrations of TBBPA and HBCD in marine organisms were 2 to 3 orders of magnitude lower than those of PBDEs. Recent review articles have examined the occurrence of TBBPA and HBCD /254,255,258/. House dust from the US contained HBCD at concentrations ranging from < 4.5 to 130,000 ng g^{-1} dry wt (median: 230 ng g^{-1} /113/). Overall, the residue levels of HBCD and TBBPA in marine organisms from North America are lower than those reported for European and Asian coastal waters, reflecting the geographic usage pattern of these compounds.

TBBPA is primarily (90%) used as a reactive flame retardant and is bound chemically to the polymer. Therefore, TBBPA is not readily leached from the products. However, when TBBPA is used as an additive component, the BFR molecules are not part of the structure of the polymer itself and can be released into the environment more readily. The annual consumption of TBBPA worldwide was estimated at 119,600 tonnes in 2001. HBCD is used more extensively in Europe than in the Americas, where it has been substituted for some of the non-foam applications for which PBDEs were formerly used. The total production of HBCD in 2001 was about 16,700 metric tonnes per year /14/.

3.1 1,2-*Bis*(2,4,6-tribromophenoxy)ethane and Decabromodiphenylethane

1,2-*Bis*(2,4,6-tribromophenoxy)ethane (BTBPE trade name FF-680) and decabromodiphenylethane (DBDPE) have been suggested as replacements for octa-BDE and deca-BDE mixtures, respectively. Both BTBPE and DBDPE have been reported in air, water, sewage sludge, sediment, mussels, fish, and birds /5,259-261/. The median concentrations of BTBPE and DBDPE in house dust from the US were 30 and 201 ng g^{-1} dry wt, respectively /113/. A significant positive correlation found between DBDPE concentrations (lw) and trophic level (based on δN-15) suggests that DBDPE bio-magnifies in the Lake Winnipeg food web /240/. The concentrations of DBDPE (1.3-288 ng g^{-1} ww) in eggs of herring gulls collected from the Great Lakes in 2005 and 2006 were similar to or higher than those of deca-BDE /261/. DBDPE was not detected in the eggs of herring gulls from the Great Lakes in 1996, whereas the concentrations did increase from 2004 to 2006 /261/.

A sediment core collected from Lake Michigan showed that the levels of BTBPE increased rapidly after 1973, with a doubling time of ~2 years until 1985, after which the BTBPE concentrations were relatively constant /259/. According to an US EPA inventory, Great Lakes Chemical produced 4500-22500 metric tonnes each year of BTBPE from 1986-1994, but the production decreased to 450-4500 metric tonnes per year after 1998 /259/. As Great Lakes Chemical plans to replace the octa-BDE product with BTBPE, however, the production of this compound may increase in the future.

3.2 2,3,4,5,6-Pentabromoethylbenzene

Pentabromoethylbenzene (PBEB) was used as an additive flame retardant for thermoset polyester and thermoplastic resins during the 1970s and 1980s. In 1977, the production of PBEB was 45-450 metric tonnes and declined to 5-225 metric tonnes in 1986. No ongoing or intended manufacture or processing of this substance occurred in 1988 /259/. PBEB, however, is listed as a low production volume chemical manufactured by Albemarle in France (according to the European Chemical Substance Information System), where PBEB production was listed as 10-1000 metric tonnes in Europe in 2002 /259/. Air samples collected in July 2003 in Chicago contained 520 pg PBEB/m^3 in the gas-phase and 29 pg PBEB/m^3 in the particle-phase /259/, although this compound was not found in sediment samples from Lake Michigan. PBEB was found in the eggs of herring gulls from the Great Lakes (0.03-1.4 ng g^{-1} ww) /262/, but at concentrations 2-3 orders of magnitude lower than those of PBDEs.

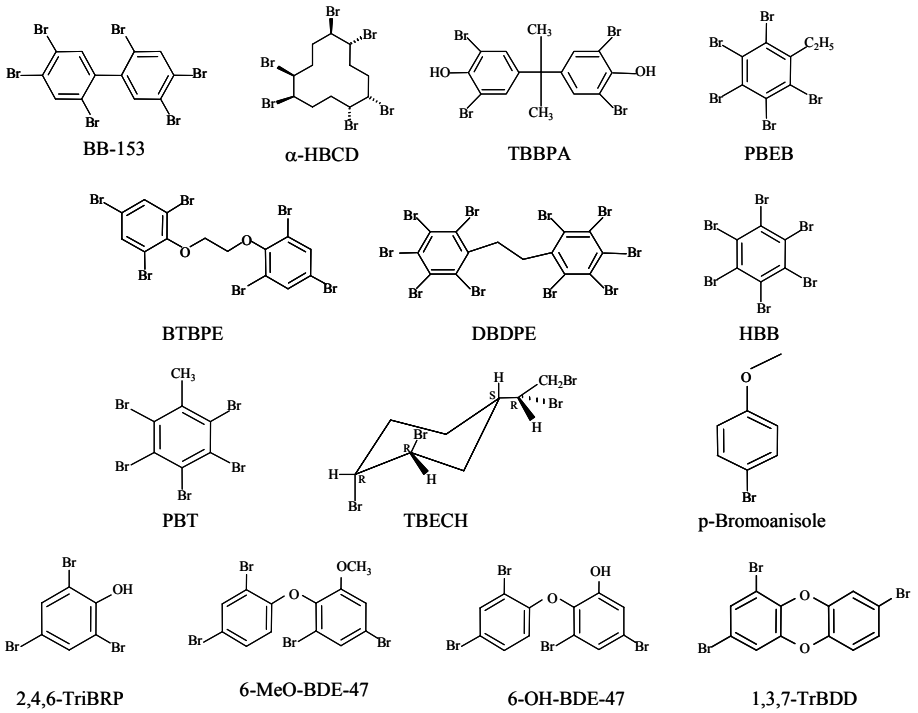

Fig. 3: Chemical structures of selected novel brominated flame retardants and naturally occurring brominated compounds in marine organisms. BB-153 – bromobiphenyl-153, HBCD – hexabromocyclododecane, TBBPA – tetrabromobisphenol A, PBEB – pentabromoethyl-benzene, BTBPE – 1,2-bis-(2,4,6-tribromohenxy)thane, DBDPE – deca bromodiphenylethane, HBB – hexabromobenzene, PBT – pentabromotoluene, TBECH – 1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane, TriBRP – tribromophenol, Meo-BDE-47 – methoxybromodiphenyl-ether-47, OH-BDE-47 – hydroxy bromodiphenyl ether-47, TrBDD – ribromodibeno-*p*-dioxin

3.3 Other BFRs

Due to the phase-out of penta- and octa-BDE mixtures, alternative flame retardants are expected to be introduced to comply with consumer product fire safety standards. Approximately 75 brominated compounds are in production for use as additives to polymers /20/. A few representative 'new' and naturally occurring organobromine compounds are shown in Figure 3. Hexabromobenzene (HBB), pentabromotoluene (PBT), and 1,2-dibromo-4-(1,2-dibromoethyl) cyclohexane (TBECH) have been reported to occur at trace levels (< 1 ng g^{-1} ww) in the eggs of herring gulls from the Great Lakes /261/. Several of the alternatives to PBDEs occur at trace levels in biota. Most of the replacements are large molecules and therefore they may be relatively less bioavailable for uptake by biota. Nevertheless, studies of the environmental degradation and transformation of these large brominated molecules are of interest.

3.4 Naturally Occurring Brominated Compounds

The occurrence of organobromine compounds including bromophenols, MeO-PBDEs, and poly-brominated hexahydroxanthenes has been reported in marine organisms, including marine mammals, fish, alga, sponges, and bacteria /263-266/. OH-PBDEs have been reported as metabolites of

PBDEs in fish and laboratory animals exposed to PBDEs. OH-PBDEs structurally resemble the thyroid hormone, thyroxine (T_4), and affect thyroid hormone homeostasis /201/. OH-PBDEs have also been shown to alter estradiol synthesis, elicit neurotoxic effects, and inhibit aromatase activity /202,267,268/. Both OH-PBDEs and MeO-PBDEs have been identified as natural products in marine sponges and red alga, including organisms from such remote marine locations as the Antarctic /269/. The structures of several OH-PBDEs and MeO-PBDEs present in marine organisms are different from those formed due to the metabolism of PBDEs and thus support the theory of a natural origin of hydoxylated and methoxylated PBDEs. For instance, the MeO-/OH- pairs 6-MeO-/OH-BDE-47 and 2'-MeO-/OH-BDE-68, all having a MeO- or OH- group in the *ortho* position of the diphenyl ether bond, have been isolated in polar bears, glaucous gulls, marine fish, marine sponges, ascidians, and algae /218,246,269/. In contrast, OH-PBDEs with an OH group in the *meta* or *para* position may be derived metabolically from PBDEs via a cytochrome P450-mediated biotransformation /266/. Findings by Teuten et al /264/, who isolated 6-MeO-BDE-47 and 2′-MeO-BDE-68 from the blubber of True's beaked whales from the North Atlantic, confirmed the natural origin of these compounds using radiocarbon (^{14}C) isotopic ratios.

Most studies describing the occurrence of OH- and MeO-PBDEs have not quantified the levels of these compounds in marine organisms. A few studies that reported the concentrations of MeO-PBDEs and OH-PBDEs showed higher levels of these compounds than concentrations of PBDEs. In fish oil dietary supplements purchased from local stores in several European countries and South Africa, the median concentrations of MeO-PBDEs and OH-PBDEs were 6.2 ng g^{-1} and 5.3 ng g^{-1} oil, respectively, which were 10-fold higher than the median concentrations of PBDEs /265/. The respective concentrations of MeO-PBDEs and OH-PBDEs, as high as 1670 ng g^{-1} and 200 ng g^{-1}, were found in fish oil dietary supplements. The abundance of OH-PBDEs and MeO-PBDEs relative to PBDEs in marine fish further supports the hypothesis of natural origin.

In contrast, fish from the Detroit River contained 2-3 orders of magnitude higher concentrations of PBDEs than OH-PBDEs /270/, whereas MeO-PBDEs were not detected in fish. The observation that OH-PBDEs in fish from the Detroit River were primarily due to the metabolism of PBDEs, and the low ratios of OH-PBDEs to PBDEs (0.02-0.002) suggested a slow rate of formation of OH-PBDEs. MeO-PBDEs were not found in Detroit River fish, although these compounds were abundant in marine mammals and marine fish, exceeding the concentrations of PBDEs. For example, in fish and marine mammals, including polar bears from the Arctic and seabirds from the Pacific Ocean, MeO-PBDEs were found at high levels /264,266,271,272/. MeO-PBDEs have been reported to biomagnify in the food chain of beluga whales in the Arctic Ocean and in dolphins feeding along the continental shelf and offshore from Rio de Janeiro, Brazil /18,224,246/. Two MeO-PBDEs, 2′-MeO-BDE-68 and 6-MeO-BDE-47, were found at the highest concentrations among the brominated compounds analyzed in beluga whale blubber and dolphin liver. A recent study measured the concentrations of PBDEs, OH-PBDEs, and MeO-PBDEs in bluefin tuna, albatross, and polar bears from several remote marine locations /272/. The concentrations of MeO-PBDEs in tuna and albatrosses were significantly higher than those of PBDEs. The demethoxylation of MeO-PBDEs to OH-PBDEs in vitro /272/ provided further evidence for the natural occurrence of OH-PBDEs in marine organisms from remote locations.

Bromophenols, bromoanisoles, and bromoindoles also occur naturally in marine ecosystems. At least 50 bromophenols have been known to occur naturally in marine plants and animals /263/. Some bromophenols, such as 2,4,6-tribromophenol,

are also produced and used as flame retardants and as a wood preservative. The worldwide production of 2,4,6-tribromophenol in 2001 was estimated to be 9500 tonnes. The ecological function of bromophenols in marine organisms is not clear, but these compounds may play a role in chemical defense and deterrence. Bromoindoles have been shown to act as antifungal and antioxidant compounds. Bromophenols have been shown to elicit estrogenic activity in vitro /273/, to affect thyroid hormone function, and to alter calcium homeostasis in endocrine cells /274/. Brominated phenols and brominated indoles have been sown to elicit lethal effects in zebrafish embryos at water concentrations on the order of a few mg L^{-1} /275/.

The most commonly reported bromophenols in marine organisms include the simple bromophenols such as, 2-bromophenol, 4-bromophenol, 2,4-dibromophenol, 2,6-dibromophenol, and 2,4,6-tribromophenol. Marine sponges from King George Island in Antarctica contained more than 35 brominated compounds of natural origin at concentrations ranging from ng kg^{-1} to mg kg^{-1} on a dry weight basis /269/. 2,4,6-tribromophenol was shown to be a major compound in sponges from Antarctica /269/, and in addition to dibromophenol, this compound has been reported to occur in seal blubber from the Arctic and Antarctica /269/. The concentrations of 2,4-dibromophenol and 2,4,6-tribromophenol in brown algae from coastal waters of Hong Kong were as high as 1280 and 7000 ng g^{-1} dry wt, respectively /276/. Tribromophenol was the most abundant bromophenol determined in marine algae from Australia and total bromophenol concentrations in alga ranged from 0.9 to 2590 ng g^{-1} ww /277/. The presence of naturally occurring organobromines, especially bromophenols in marine invertebrates and fish provides a link to their occurrence in marine mammals.

The natural formation of polybrominated dibenzo-*p*-dioxins (PBDD) in marine algae has been reported /278, 279/. Mussels from the Baltic Sea contained PBDD concentrations a high as 4.1 ng g^{-1} lw. The direct condensation of bromophenols has been suggested as the origin of PBDD, especially di-, tri- and tetra-bromodibenzo-*p*-dioxins in mussels and several species of fish from the Baltic Sea. A recent study showed the potential photochemical formation of polybrominated and mixed halogenated dibenzo-*p*-dioxins (PBDDs and PXDDs) from OH-PBDEs in aqueous solution /280/. In addition, polybrominated dibenzofurans (PBDFs) occur as impurities in technical PBDE mixtures /281/. Several of the naturally occurring organobromines, including PBDDs, have been shown to elicit toxic effects in laboratory tests.

4. CONCLUSIONS: FORESIGHT FROM CURRENT KNOWLEDGE

The BFRs are major industrial chemicals whose use has increased dramatically over the past few decades. These compounds are produced to retard fires and thus can have a direct benefit. Yet, the demonstrated persistence, bioaccumulation, and toxic potential of these compounds in animals and in humans are of increasing concern /20/. Since their introduction in the 1970s, PBDEs have been increasing in abiotic and biotic matrices in coastal marine environments of North America and are beginning to rival PCBs as the predominant contaminants in water and sediments in near-urban source areas. PBDEs, like structurally related PCBs, are strongly particle-reactive, especially the heavier congeners, and their presence in marine sediments assures their efficient delivery to aquatic/marine food webs. As a result, the deep oceans are global sinks for PBDEs and other hydrophobic POPs /8/, as evidenced by detection of PBDEs in a variety of deep-sea organisms.

Extrapolating from temporal trend studies, PBDEs are increasing exponentially in many North American fish, seabird, and marine mammal populations and are projected to overtake PCBs as the dominant contaminants in tissue within 10 years. Whereas a penta-BDE signature characterizes the accumulation patterns in fish and piscivorous

wildlife, exposure to the octa- and deca-BDE mixtures is indicated by the patterns of terrestrial food-web related species and in abiotic matrices. In certain aquatic/marine food webs, bioaccumulation and magnification of constituents of all three PBDE commercial mixtures is evident, presenting an increasing health risk to marine animals. The synergistic interactions and effects due to exposure to multiple contaminants (especially the mixture of PCBs and PBDEs) in marine organisms need further investigation.

Despite recent controls on the production and use of the penta- and octa-BDE commercial formulations in the US and Canada, penta-BDE-containing products will remain a reservoir for PBDE releases for years to come. For example, the average lifetime for foam-containing household furniture and automobile padding has been estimated at 10 years /34/. Of the overall estimated PBDE intake in Americans, 80% to 90% results from dust ingestion and inhalation, implying that only a fraction of the total PBDEs produced have reached the outdoor environment /88/. It is the entry of these chemicals into domestic dust through the aging/disposal of consumer products that supports the efficient transport of PBDEs to coastal waters by municipal wastewater systems and landfills.

Similar to our experience with PCBs (which have been banned from production and use in most developed countries for three decades), it is likely that regulatory actions aimed at controlling PBDEs by banning manufacturing and new uses will be insufficient to stop this trend. PBDE congeners from the penta-, octa-, and deca-BDE mixtures will continue to be released into the environment during the use, disposal, and recycling of existing fire-retardant-containing products for years to come. This situation is especially troublesome when one considers that large amounts of electronics and furniture, manufactured when PBDEs were most heavily used, are likely being recycled and disposed of at present or will be in the near future /282/. Similarly, Tanabe /19/ predicted that most of the PCBs (66%) were stockpiled in products (transformers and capactors) long after PCBs were banned from production in the 1970s. The indoor reservoir of PBDEs has been termed an environmental ‘time bomb’ /283/ as these chemicals are slowly leaching into coastal and marine waters and contaminating ocean food webs. Following the pattern observed for PCBs, the main exposure route for humans is predicted to shift eventually from the indoor environment to the food web /283/, signifying the delivery of significant quantities of PBDEs to the environment. The magnitude and timing of this shift cannot be predicted as both depend upon unknown factors like the amount of PBDEs yet to be released (from products containing such compounds and the ongoing usage of deca-BDE) and their uptake, incorporation, and magnification into marine and terrestrial food webs.

Concerted action is needed not only to ban the production and use of PBDEs but also to find ways of reducing the existing indoor reservoirs and managing the end-of-life of PBDE-containing products. However, even if the inputs of PBDEs can be controlled/reduced in the terrestrial environment, once these compounds have reached the oceans, levels are unlikely to decline any time soon. Thirty years after being banned, the amounts of PCBs cycling in marine food webs are gradually decreasing, as burial, metabolism, and degradation occur in marine sediments. PBDE emissions are repeating the experience with PCBs, such that we are now at the same point reached for PCBs in the late 1960s /38/. PBDE discharges continue to increase, and these compounds are loading into all compartments of the environment. The evidence suggests that if PBDEs were banned today, it will take decades after the end of discharge for marine sediment to bury them /38/.

Moreover, the environmental fate of PBDEs in various abiotic environmental compartments has not been adequately investigated. Although many authors have reported on the presence of PBDEs in air and sediments, comparatively little is known about PBDEs in North American surface waters

and soils. Perhaps the greatest source of uncertainty regarding the movement and fate of PBDEs in abiotic media is the extent of degradation in such environments. The degradation of BDE-209 is of particular interest as this congener is still in use, and its degradation may contribute to the environmental enrichment of lesser brominated, more toxic congeners.

Furthermore, a wide range of organobromine compounds occur in marine ecosystems and accumulate in marine organisms (such as algae, marine sponges). Many of these naturally occurring organobromines structurally resemble man-made brominated compounds (bromophenols, OH-PBDEs, MeO-PBDEs, and PBDDs). The toxicological significance of the naturally occurring brominated compounds in marine organisms is not understood. Studies are needed to evaluate the toxic effects of man-made and naturally occurring brominated compounds on marine organisms and humans.

Ample evidence indicates that the marine ecosystems of the North American continent are contaminated by PBDEs; yet, very little information exists for Central and South America. Recent data from Brazil indicate that PBDEs are becoming major contaminants, similar to PCBs and DDT, in marine biota inhabiting densely populated coastal areas. The PBDE burdens in several cetacean species are comparable to those in northern hemisphere cetaceans and are rapidly increasing.

Moreover, the naturally occurring MeO-PBDEs detected in cetaceans feeding along the continental shelf of South America are among the highest reported to date. The data from these reports, along with the observation of serum PBDE concentrations in Nicaraguan children working and living at a waste disposal site that are the highest ever reported, exceeding by 8-fold the current median concentrations in Americans /87/, highlight the need for a worldwide exposure assessment of PBDEs, not just in the developed countries /101/.

Few studies have addressed the effects of PBDE exposures in marine organisms, although relations between PBDE burdens and reproductive behavior, immunosuppression, and thyroid alterations have been observed in the field /68,74,77/. Mink are a particularly useful surrogate species for evaluating the toxic effects of pollutants in aquatic mammals, and new evidence suggests that exposure to environmentally relevant levels of PBDEs results in reproductive failure in mink /62/. Additional toxicological studies with sentinel species, such as fish, birds, seals, and mink/otter are needed to derive a threshold value for the toxic effects of PBDEs in marine organisms.

A further concern is the introduction of novel brominated flame retardant chemicals that are being marketed as replacements for the now-banned penta- and octa-PBDE mixtures. Recent studies have confirmed that such compounds as BTBPE and DBDPE have contaminated aquatic food webs. These non-regulated compounds are similar to their predecessors in structure and have not been adequately studied for their persistence and toxic properties.

Overall, the future trends for the PBDEs in the marine environments of the American continents are not clear, despite ongoing efforts to regulate/ban the production and use of these compounds. The current indication is that PBDEs will soon overtake PCBs as the dominant contaminants in marine biota, as large and growing reservoirs of these flame retardants exist and effective strategies for the reduction/end-of-life cycle management of PBDE-containing products are not in place. Moreover, the discharge of deca-BDE to marine ecosystems is increasing exponentially, ensuring the ongoing delivery of BDE-209 and its toxic, persistent breakdown products to the ocean environment for years or even decades after production ceases. Our past experience with PCBs strongly suggests that regulatory controls will do little to impede the continued cycling of these flame retardant chemicals through aquatic and marine ecosystems for the foreseeable future.

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