Polybrominated diphenyl ethers in pinnipeds stranded along the southern California coast

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Abstract

Little to no information exists for polybrominated diphenyl ethers (PBDEs) in marine mammals frequenting the highly urbanized southern California (USA) coast. Fourteen PBDE congeners were determined by GC-ECNI-MS in blubber of pinnipeds stranded locally between 1994 and 2006. Total PBDE concentrations (ΣPBDE) in California sea lion (*n* = 63) ranged from 0.04 to 33.7 µg/g wet weight (mean: 5.24 µg/g). To the authors' knowledge, these are the highest reported PBDE levels in marine mammals to date. By comparison, mean ΣPBDE in Pacific harbor seals (*n* = 9) and northern elephant seals (*n* = 16) were 0.96 and 0.09 µg/g, respectively. PBDEs in adult males were higher than for adult females, however, no age class differences or temporal trends were observed. As the first PBDE data reported for marine mammals in this region, the elevated levels underscore the need for additional studies on the sources, temporal trends, and potential effects of PBDEs in highly urbanized coastal zones.

Introduction

Polybrominated diphenyl ethers are a class of flame retardants added in bulk to consumer and commercial products, including: electronics, automobiles, paints, furniture, clothing, and plastics (de Wit 2002). For example, polyurethane foam can be treated with as much as 10 to 30 wt % PBDEs to comply with federal and state safety regulations (Hites 2004). A high-volume use chemical, approximately 67,400 metric tons of formulations consisting predominantly of tetra- and higher brominated congeners were used worldwide in 2001 (Law *et al*. 2006). United States (US) demand for PBDEs has accounted for more than one-third of global production, and the State of California has among the most stringent fire retarding regulations for consumer products in the world. In 2006, California became the first US state to ban the use of penta- and octa-BDE formulations in commerce (California Environmental Protection Agency 2006).

Polybrominated diphenyl ethers can be released into the environment prior to product manufacture, as well as after such products have been discarded as waste (de Wit 2002). Structurally similar to polychlorinated biphenyls (PCBs), PBDEs are very hydrophobic (log K_{ow} values 4 - 10), resistant to degradation, and biomagnify in aquatic and terrestrial food webs (Boon *et al*. 2002, Muir *et al*. 2006, Sørmo *et al*. 2006, Wolkers *et al*. 2006). They have been reported in air, water, sediment, fish, birds, marine mammals, food, and human tissue from around the globe in recent years and environmental levels continue to increase over time (Hites 2004, Law *et al*. 2006, Wang *et al*. 2007). Selected congeners are thought to be endocrine disrupters (Darnerud 2008) with evidence of interaction between thyroid hormones reported in certain marine mammal species during their first year of life (Hall *et al*. 2003).

Several studies have found elevated levels of PBDEs in marine mammals as top predators in aquatic food webs (Haglund *et al*. 1997, Lindström *et al*. 1999, Ikonomou *et al*. 2002, Lebeuf *et al*. 2004, Rayne *et al*. 2004, Kajiwara *et al*. 2004, Thron *et al*. 2004, Tuerk *et al*. 2005, Johnson-Restrepo *et al*. 2005, Kannan *et al*. 2005a, Ramu *et al*. 2005, Rigét *et al*. 2006, Litz *et al*. 2007, Vorkamp *et al*. 2008). Polybrominated diphenyl ethers concentration in marine mammals from the Canadian Arctic

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increased exponentially with a doubling time of approximately seven years, similar to that reported in other parts of the world (Hites 2004). Levels approaching 10 μg/g wet weight have been reported in urbanized coastal areas (Law *et al*. 2002, Fair *et al*. 2007). Similar levels have been found in blubber of pinnipeds stranded in San Francisco Bay (She *et al*. 2002) and along the central California coast (Stapleton *et al*. 2006). Kannan *et al*. (2007) recently reported maximum PBDE levels in liver of southern sea otters (*Enhydra lutris nereis*) from the central California coast of 27 μg/g lipid, the highest lipid-normalized PBDE levels reported for marine mammals to date.

In contrast, no studies on PBDEs in marine mammals frequenting the southern California coast have been conducted to date, even though this region is one of the most heavily populated in the world. The goals of this study were to determine and compare PBDE levels among regionally abundant pinniped species, examine the influence of gender and age, and evaluate for temporal trends in levels during the study period (1994-2006). To accomplish this, blubber samples from three species of pinnipeds stranded along the southern California coast during this period were collected, processed, and subjected to congener-specific PBDE analysis.

Methods

Target Species and Sample Collection

The California sea lion (*Zalophus californianus*) is a carnivore that ranges from western Canada to southern Baja California (Mexico) in the eastern Pacific ocean. *Z. californianus* inhabits coastal areas and feeds opportunistically on fish and squid, making it a good sentinel of nearshore environmental contamination (Stapleton *et al*. 2006). The Pacific harbor seal (*Phoca vitulina*) ranges from Alaska to Baja California and also favors nearshore coastal waters, often seen at sandy beaches, mudflats, bays, and estuaries (She *et al*. 2002). Largest of the three study species, the northern elephant seal (*Mirounga angustirostris*) is found throughout the northeastern Pacific from Baja California to the Gulf of Alaska. In contrast to the other target species, elephant seals remain well offshore (up to 8000 km) for the majority of their lives, using selected islands off the southern California coast as seasonal breeding and molting grounds (Bartholomew and Boolootian 1960).

Blubber samples from 63 sea lions, 9 harbor seals, and 16 elephant seals found stranded along a 150 km stretch of the southern California coast (comprising Los Angeles and Orange counties) during 1994-2006 were obtained from the Fort MacArthur (San Pedro, CA), and Pacific Marine Mammal Centers (Laguna Beach, CA) as described in Blasius and Goodmanlowe (2008). Briefly, full depth blubber samples were taken from animals alive at the time of stranding but who subsequently died at the Centers, wrapped in clean aluminum foil and kept frozen at -20°C. Detailed information (species, sex, age class, cause of death and body condition) was compiled for each stranded animal by Center personnel. Samples from sea lions of both gender and at various life stages were included in this study; in contrast, the majority of samples for harbor and elephant seals were from pups (<1 year-old) of both gender (Table 1).

Extraction and Chemical Analysis

Blubber samples were originally extracted for analysis of organochlorines including PCBs, DDTs and chlordanes (Blasius and Goodmanlowe 2008). Approximately 1.0 g of blubber was extracted three times with 25 ml dichloromethane (DCM) at 100°C for 15 minutes in a Teflon vessel under microwave energy. Lipid content was determined gravimetrically after evaporation of DCM. Extracts were purified on a SiO_2/Al_2O_3 column packed with 3% water deactivated silica gel and basic alumina. Target organochlorines were eluted with 15 ml of hexane and 30 ml of 30% DCM in hexane (v:v). Recoveries of tetrachloro-m-xylene and PCBs 30, 112 and 198 spiked into each sample prior to microwave extraction ranged between 70 and 130%. For determination of PBDEs, the extracts (stored at -20°C) were spiked with BDEs 71 and 172 as cleanup recovery surrogates and applied to 40 g of SX-3 “Bio-Beads” (Bio-Rad Laboratories, Hercules, CA, USA) packed into a 50 cm x 2.5 cm id glass column. Target PBDEs were eluted in the 100 to 220 ml fraction of 50% DCM in hexane (v:v), which was then concentrated to 1 ml using a TurboVap 500 evaporator (Zymark, Hopkinton, MA, USA).

To confirm quantitative recovery of target PBDEs retrospectively, three blubber samples were re-processed using a second protocol. After thawing at room temperature, ~1.0 g of blubber was ground with kiln-fired (~500°C for 5 hours) Na_2SO_4 and extracted with DCM using a Dionex 300 ASE system (Salt Lake City, UT, USA). After addition of

Table 1. Lipid and total PBDE concentrations (ΣPBDE) in blubber of three pinniped species stranded along the southern California coast from 1994 to 2006.

California Sea Lion (*Zalophus californianus*)

Age Class (years)	*n*	% Lipid		ΣPBDE (µg/g ww[a])		ΣPBDE (µg/g lw[a])	
		Mean ±sd	Range	Mean ±sd	Range	Mean ±sd	Range
Pup (< 1)	12	46.5 ±36.5	0.2 - 96.4	6.23 ±4.26[b]	0.42 - 13.9	51.10 ±75.2[b]	2.90 - 236
Yearling (1 - 2)	16	56.7 ±35.3	0.7 - 95.4	6.17 ±8.29[b c]	0.04 - 33.7	36.50 ±63.3[b c]	0.06 - 184
Sub-adult (2 - 8)	15	60.1 ±28.7	5.0 - 97.1	5.73 ±4.50[b]	1.73 - 15.5	16.90 ±21.7[b]	2.15 - 84.6
Adult M (>8)	5	38.3 ±40.6	2.4 - 100.0	6.19 ±3.86[b]	0.97 - 10.6	55.30 ±79.3[b]	3.43 - 194
Adult F (>5)	15	66.4 ±22.3	7.2 - 98.9	2.65 ±2.52[c]	0.22 - 8.08	5.15 ±5.52[c]	0.27 - 16.0
All	63	56.4 ±31.9	0.2 - 100.0	5.24 ±5.38	0.04 - 33.7	28.70 ±53.2	0.06 - 236

Pacific Harbor Seal (*Phoca vitulina*)

Age Class (years)	*n*	% Lipid		ΣPBDE (µg/g ww[a])		ΣPBDE (µg/g lw[a])	
		Mean ±sd	Range	Mean ±sd	Range	Mean ±sd	Range
Pup (<0.5)	8	57.4 ±25.2	22.3 - 89.8	1.08 ±0.73	0.16 - 2.41	2.43 ±2.25	0.32 - 7.20
Adult M (>4)	1	7.3		0.03		0.37	
All	9	51.8 ±28.9	7.3 - 89.8	0.96 ±0.76	0.03 - 2.41	2.20 ±2.22	0.32 - 7.20

Northern Elephant Seal (*Mirounga angustirostris*)

Age Class (years)	*n*	% Lipid		ΣPBDE (mg/g ww[a])		ΣPBDE (µg/g lw[a])	
		Mean ±sd	Range	Mean ±sd	Range	Mean ±sd	Range
Pup (<1)	16	53.0 ±31.0	0.5 - 95.6	0.09 ±0.11	0.01 - 0.45	0.36 ±0.57	0.04 - 2.01

[a]Wet weight = ww; Lipid weight = lw

[b]Male (M) and female (F) animals included unless otherwise specified

[c]Mean concentrations significantly different (p = 0 05)

recovery surrogates BDE 71 and 172, each sample was extracted sequentially (four cycles) at 100°C and 1500 psi followed by a nitrogen gas purge (100 seconds). After gravimetric determination of lipid content, lipid in the remaining extract was removed using Bio-Beads as described above. The collected fraction was concentrated and further purified on a 10 mm id glass column packed (from top to bottom) with neutral alumina (6 cm, 3% deactivated) and silica gel (12 cm, 3% deactivated). Target PBDEs were eluted with 50 ml of 3:7 DCM/ hexane (v:v) and resulting eluates were reduced to ~1 ml and stored at -20°C until analysis.

Extracts were analyzed on an Agilent 7890A gas chromatograph/5975C quadrupole mass spectrometer (Palo Alto, CA, USA) outfitted with a 30 m x 0.25 mm id x 0.25 µm DB-XLB fused silica column (J&W Scientific, Folsom, CA, USA). Splitless mode injections of 1 µL were performed with an injector temperature of 280°C. The GC oven was programmed as follows: 60°C (3-minute hold); increase to 250°C at 20°C/minute; increase to 300°C at 2°C/minute (5-minute hold) for a total run time of 43 minutes. Ultrahigh purity helium was used as the carrier gas. The transfer line, ion source and quadrupole temperatures were maintained at 280, 250, and 300°C, respectively. The MS was operated in the negative chemical ionization and selected ion monitoring modes, with *m/z* 79 and 81 serving as the quantitation ions. Methane (>99.999%) as the CI moderating gas was maintained at a pressure of ~1.0 x 10^{-4} torr.

Congener-specific quantitation of BDE 15, 28, 33, 47, 49, 66, 75, 99, 100, 119, 153, 154, 155, and 183 and two surrogates (BDE 71 and 172) was based on a nine point calibration curve (1 to 500 ng/ml). The nominal congener-specific method detection limit (MDL) was 1.0 ng/g wet weight (1.0 g sample mass; 1 µL GC injection volume). Extracts with BDE concentrations that exceeded the linear calibration upper limit were diluted and re-analyzed. Several target BDEs were detected in procedural blanks ($n = 6$); however, only BDE 47 was consistently detected above 1.0 ng/ml and thus its MDL was set at its average blank level of 2.3 ng/g ww. The mean, ± standard deviation (sd), recoveries of BDE 71 and 172 were 105 ±8.8% and 93 ±11%, respectively. Using the ASE protocol, the mean (±sd) recovery of the 14 target PBDEs plus two additional congeners spiked into elephant seal blubber samples with low organochlorine levels (M. Blasius, unpublished data) was 92 ±3.6%, and the mean recovery of PBDE congeners in SRM1945 (pilot whale homogenate; National Institute of Standards and Technology, Gaithersburg, MD, USA) was 82 ±8.2%. The mean difference in PBDE congener concentration for the three blubber samples processed using the two procedures was 8.6%, indicating that the extraction of PBDEs using the original microwave protocol was quantitative.

Data and Statistical Analysis

Statistical analyses were performed using SPSS version 11.5 (SPSS Inc., Chicago, IL, USA) with statistical significance for all tests set at $\alpha = 0.05$. Total PBDE concentrations (ΣPBDE) for wet weight (ww) and lipid weight (lw) were represented as the sum of the 14 target congeners. For statistical evaluation of ΣPBDE, only congeners with concentrations greater than the MDL were included. Concentrations were log-transformed prior to the statistical analyses if they were not normally distributed (Kolmogorov-Smirnov Z test). Student's t-test and one-way analysis of variance (ANOVA) were used to examine the difference between means. Tukey's multiple comparison test was used when a difference by ANOVA was observed.

Results

Congener-specific concentrations, ΣPBDE, lipid content and animal body condition for individual samples are shown by species in Tables 2 through 4. Average ΣPBDE for sea lions (5.24 ±5.38 µg/g ww; 28.7 ±53.2 µg/g lw) were one to two orders of magnitude higher than for harbor or elephant seals (Table 1). ΣPBDE for the latter two species averaged 0.96 ±0.76 µg/g ww (2.20 ±2.22 µg/g lw) and 0.09 ±0.11 µg/g ww (0.36 ± 0.57 µg/g lw), respectively. Lipid content averaged 56 ±32% for all species, but was highly variable, ranging from as little as 0.2 to 100% (Table 1). Blubber lipid content below 10% is unusual but not unprecedented for stranded pinnipeds (Stapleton *et al*. 2006); in this study, 11 of the 13 animals with lipid <10% were classified in poor (emaciated) to fair condition (Table 2). Although lipid was relatively low (18%) for the animal with the highest ΣPBDE (33.7 µg/g ww), lipid content for the 10 highest ranked samples averaged 61 ±27%, suggesting that poor animal condition was not associated with elevated wet weight PBDE levels.

In contrast, if the 13 samples with <10% lipid (Table 2) were removed from consideration, mean lipid content for sea lions in Table 1 would increase by ~10% (66 ±24%) and mean lipid-normalized ΣPBDE for all sea lion samples would decrease by a factor of two (13.3 ± 27.0 µg/g lw). However, the change in mean wet weight ΣPBDE by eliminating the 13 "low lipid" samples would be small (<14%) for all species and age classes, and the overall mean for sea lions would not change appreciably (e.g., 5.71 ±5.67 µg/g ww vs. 5.24 ±5.38 µg/g ww). Thus, statistical comparisons among species, age classes and by gender were performed on a wet weight basis, and caution should be exercised when comparing lipid normalized data from this study.

Brominated diphenyl ether (BDE) 47 was the most abundant congener in all blubber samples, with mean (range) contributions of 67% (47 - 90%), 76% (66 - 85%), and 69% (43 - 81%) for sea lions, harbor and elephant seals, respectively (Figure 1). Moreover, the sum contribution of BDE 47, 99, and 100 to ΣPBDE was greater than 90% for all three species, which allows for direct comparison of ΣPBDE reported herein with several previous studies (e.g., Ikonomou *et al*. 2002, She *et al*. 2002, Rayne *et al*. 2004, Kalazanti *et al*. 2005, Law *et al*. 2005, Ramu *et al*. 2005, Rigét *et al*. 2006, Stapleton *et al*. 2006, Vorkamp *et al*. 2008).

Considering only pups, mean ΣPBDE for sea lion, harbor and elephant seals were 6.23, 1.08 and 0.09 µg/g ww (51, 2.2, and 0.36 µg/g lw), respectively (Table 1), and were significantly different among the three pinniped species (ANOVA; both $p < 0.001$; Figure 2). Mean ΣPBDE in sea lion pups were significantly higher than those in harbor and elephant seals (both $p < 0.001$). Mean PBDE concentrations in harbor seals were also higher than those in elephant seals, but this difference was statistically significant only for lipid weight concentrations. The influence of age on PBDE levels was examined only for California sea lions due to insufficient sample size for the other study species. Classifying sea lions into four age groups -- pups (0 - 1 year), yearlings (1 - 2 years), subadults (2 - 8 years for males and 2 - 5 years for females), and adults (>8 yr for males;Table 1) -- no significant difference by age class ($p = 0.996$) as observed.

To facilitate comparison in PBDEs by gender, sea lion pups, yearlings and subadults (Table 1) were classified as juveniles. Total PBDE concentrations for juvenile males (mean ±sd: 6.30 ±8.17 µg/g ww; $n = 18$) and females (5.84 ±3.98 µg/g ww; $n = 25$) were not statistically different ($p = 0.81$; Figure 3). Similarly, no significant differences were found in ΣPBDE between male and female pups for harbor or elephant seals ($p = 0.32$ and $p = 0.35$, respectively; Figure 3). In contrast, a significant difference was observed in ΣPBDE between adult male (mean ±sd: 6.19 ±3.86 µg/g ww; $n = 5$) and adult female sea lions (2.65 ±2.52 µg/g ww; $n = 15$; Figure 3). Moreover, PBDEs in female adults were significantly lower than those in male and female juveniles combined (mean ±sd: 6.03 ±6.01 µg/g ww; $p = 0.04$).

Because significant differences in ΣPBDE were only observed by gender for adult sea lions, data for male and female juveniles and adult males were combined to investigate for trends between 1994 and 2006. For harbor and elephant seals, only data for pups were available. As sample sizes for a given year were relatively small and uneven, the power of trend analysis is limited. For sea lions, a significant difference was found over the study period using ANOVA ($p = 0.04$); however, no consistent increasing or declining trend was observed (Figure 4). The single highest (33.7 µg/g ww) and peak mean PBDE concentration (12.9 µg/g ww; $n = 6$) were observed in animals stranded in 1997, whereas the lowest level was found in the previous year, when only one sample (0.04 µg/g ww) was analyzed. Peak concentrations were also observed for harbor and elephant seals, occurring in 2001 (mean: 1.98 µg/g ww; $n = 2$) and 2000 (mean: 0.18 µg/g ww; $n = 3$), respectively.

Discussion

To the authors' knowledge, the mean (5.24 µg/g ww) and maximum (33.7 µg/g ww) concentrations of PBDEs in blubber of California sea lions reported herein are the highest in pinnipeds to date. Furthermore, these levels were higher than any marine mammal species expressed on a wet weight basis. Prior to this study, the highest single animal ΣPBDE reported was 16.2 µg/g ww for a killer whale stranded in the UK (Law *et al*. 2005), less than half the maximum concentration (33.7 µg/g ww) for a sea lion reported herein.

Like other persistent organic pollutants (POPs), a significant fraction of PBDEs found in juvenile marine mammals are transferred from their mothers via milk during lactation (Hall *et al*. 2003, Sørmo *et al*. 2003). For pinnipeds, PBDE transfer from mother to pup was reported to be 44.4 ±9.9% ($n = 5$; Ikonomou and Addison, 2008). Thus, it is likely that the difference in PBDE levels for pups in this study was due in large part to the difference in PBDE contamination in their adult mothers. Studies employing stable isotope ratios indicated that the three study species represent the same trophic level and have similar food preferences, which include fish, squid, and larger benthic invertebrates (Pauly *et al*. 1998, Burton and Koch 1999). In contrast, their home ranges differ substantially with California sea lions staying closest to shore, feeding throughout the water column out to the edge of the continental shelf. Adult females, in particular, remain within the Southern California Bight to nurse their pups (Stewart and DeLong 1995). Pacific harbor seals also inhabit the coastal zone, but may also spend several days in the open ocean. In the present study region, sea lions in particular are commonly sighted within highly industrialized coastal embayments with elevated contamination levels (e.g., the Los Angeles/Long Beach Harbor complex). Although virtually no data exists on the occurrence, sources and inputs of PBDEs in this system, a recent pilot study by NOAA has indicated elevated levels of PBDEs in coastal bivalves collected within 20 km of the Harbor complex (G. Lauenstein, pers. comm.). Elephant seals, on the other hand, are rarely seen in these populated and/or industrialized areas. A pelagic feeder that frequents deeper waters, the adult elephant seal ranges as far north as Alaska. When present in the study area, they are largely confined to the northern Channel Islands (Bartholomew and Boolootian 1960), well offshore and some 100 km from known contamination hotspots.

Non-gender differentiation in PBDE levels for juveniles were previously reported, e.g., for belugas; harbor, grey, and ringed seals; polar bears; and killer whales (She *et al*. 2002, Lebeuf *et al*. 2004, Rayne *et al*. 2004, Kalantzi *et al*. 2005, Kannan *et al*. 2005b, Vorkamp *et al*. 2008). It follows that male and female juveniles for a given species/population are

Table 2. Congener-specific and total PBDE concentrations (ng/g wet weight) in blubber of California sea lions (*Zalophus californianus*) stranded along the southern California (USA) coast from 1994 to 2006 (n = 63). Body condition was assessed based on blubber thickness during the necropsy using the following scale: 1 = emaciated (2 mm or less); 2 = thin; 3 = fair (1 - 2 cm); 4, 5 = good (2 - 3 cm); 6 = excellent (3 - 4 cm); NR = not recorded; and nd = not detected.

Sample ID	Collection Year	Sex	Age Class	% Lipid	15	28	33	47	49	66	75	99	100	119	153	154	155	183	∑PBDE	Body Condition
00-101	2000	M	Pup	43.5	<=0.9	91.3	<=0.7	1265.0	12.0	35.3	2.7	151.0	256.0	<=0.2	18.5	30.6	4.9	<=0.3	1870	1 - 2
01-146	2001	M	Pup	7.7	<=0.5	53.5	2.4	1908.0	4.8	35.1	3.7	331.0	472.0	<=0.2	51.0	80.9	11.7	<=0.8	2950	3
03-002	2003	M	Pup	1.7	<=0.8	103.0	2.1	2783.0	3.6	44.2	8.2	439.0	568.0	<=0.1	50.0	74.6	13.7	<=0.8	4090	2
04-111	2004	M	Pup	32.6	1.1	128.0	2.1	3990.0	10.9	63.9	1.3	522.0	240.0	<=0.3	58.7	84.9	24.2	<=0.8	5130	2
97-136	1997	F	Pup	79.6	2.8	347.0	3.5	7940.0	21.6	144.0	7.6	1144.0	1558.0	<=0.2	167.0	241.0	36.8	3.5	11,620	NR
98-071	1998	F	Pup	96.4	1.1	81.4	3.9	7432.0	11.3	34.0	10.8	750.0	1707.0	<=0.5	202.0	347.0	74.4	3.8	10,660	NR
00-46	2000	F	Pup	39.8	1.7	169.0	3.2	5445.0	16.4	69.3	4.9	639.0	1033.0	<=0.8	105.0	148.0	34.6	1.7	7670	3 - 4
00-52	2000	F	Pup	89.5	1.6	110.0	4.1	9304.0	21.9	108.0	3.7	3239.0	516.0	1.6	262.0	243.0	38.6	6.7	13,860	5
03-007	2003	F	Pup	0.2	<=0.2	10.2	1.3	279.0	<=0.5	5.1	<=0.3	46.5	53.1	<=0.1	12.2	14.0	3.4	<=0.5	424	2
03-028	2003	F	Pup	10.7	1.4	145.0	3.3	5926.0	3.7	80.0	5.8	583.0	1304.0	<=0.6	101.0	178.0	44.1	<=0.6	8380	2
04-010	2004	F	Pup	71.5	1.8	89.4	1.8	1402.0	15.1	47.4	4.3	185.0	262.0	nd	24.1	37.2	7.0	<=0.3	2080	5
05-399	2005	F	Pup	84.3	1.1	109.0	2.0	4274.0	10.9	51.6	6.7	552.0	886.0	nd	73.8	111.0	20.1	1.0	6100	NR
97-127	1997	M	Yearling	86.5	2.5	239.0	10.0	9466.0	28.0	85.0	10.2	1185.0	1059.0	<=0.7	227.0	265.0	65.2	4.3	12,650	NR
97-132	1997	M	Yearling	18.4	3.3	629.0	5.0	23718.0	28.1	346.0	18.0	3428.0	4052.0	1.1	541.0	778.0	140.0	11.7	33,700	NR
03-297	2003	M	Yearling	0.8	<=0.2	4.8	<=0.6	114.0	1.1	3.7	<=0.4	25.7	33.4	nd	6.9	8.4	2.3	<=0.3	200	1
03-069	2003	M	Yearling	0.7	<=0.3	42.3	<=0.8	889.0	1.9	16.9	1.4	98.3	157.0	<=0.2	15.4	23.9	6.2	<=0.2	1250	2
03-378	2003	M	Yearling	95.1	1.3	58.2	2.9	1464.0	15.1	46.9	5.1	352.0	293.0	<=0.3	36.2	55.0	10.0	<=0.5	2340	4
04-089	2004	M	Yearling	62.5	3.1	206.0	2.9	5522.0	16.2	99.5	11.4	887.0	1170.0	<=0.7	122.0	194.0	36.2	<=0.2	8270	1
04-121	2004	M	Yearling	19.2	<=0.7	19.7	3.0	381.0	5.4	14.7	1.2	63.1	76.2	<=0.3	15.5	19.7	4.5	<=0.6	604	2
05-405	2005	M	Yearling	79.9	<=0.4	22.8	1.3	806.0	3.8	10.8	<=0.7	86.5	68.2	<=0.4	15.8	21.7	7.2	<=0.2	1040	NR
96-076	1996	F	Yearling	66.0	<=0.1	<=0.6	1.6	28.6	<=0.2	1.7	<=0.1	4.2	3.7	nd	<=0.7	<=0.9	<=0.1	<=0.1	39.8	NR
97-080	1997	F	Yearling	67.9	2.3	278.0	3.1	6739.0	29.8	244.0	8.1	1373.0	1243.0	<=0.7	163.0	248.0	58.3	1.7	10,390	2
98-178	1998	F	Yearling	3.8	1.5	137.0	1.7	2901.0	10.5	94.0	3.6	508.0	467.0	1.2	57.3	80.8	15.2	<=0.7	4280	6
00-091	2000	F	Yearling	78.3	2.7	259.0	4.9	6216.0	20.8	133.0	8.4	930.0	1415.0	<=0.6	125.0	187.0	33.1	2.0	9340	2
01-080	2001	F	Yearling	95.4	1.1	110.0	1.4	2959.0	19.8	71.4	3.1	468.0	387.0	<=0.2	63.4	86.5	14.6	<=0.6	4180	4
04-254	2004	F	Yearling	64.5	1.7	63.9	3.0	2563.0	12.0	41.0	3.7	449.0	280.0	<=0.3	51.0	73.7	15.4	<=0.9	3560	NR
05-071	2005	F	Yearling	78.0	<=0.4	43.1	1.3	1708.0	7.6	60.2	2.7	633.0	358.0	<=0.5	51.9	78.3	14.4	1.0	2960	3
05-404	2005	F	Yearling	91.3	2.2	55.7	1.4	2627.0	6.2	34.5	2.9	435.0	538.0	<=0.2	61.2	79.7	12.5	<=0.6	3660	4 - 5
97-010	1997	M	Sub-adult	52.9	<=0.9	96.3	1.2	2136.0	11.8	57.1	3.4	317.0	355.0	<=0.2	36.5	59.6	10.0	1.1	3080	6
98-129	1998	M	Sub-adult	64.7	<=0.9	119.0	5.6	12862.0	21.8	44.0	13.4	642.0	1247.0	<=0.5	169.0	331.0	56.5	1.4	15,510	NR

Table 2. Continued

Sample ID	Collection Year	Sex	Age Class	% Lipid	15	28	33	47	49	66	75	99	100	119	153	154	155	183	∑PBDE	Body Condition
00-010	2000	M	Sub-adult	80.4	<=0.6	32.7	1.5	1186.0	7.4	11.4	1.9	189.0	187.0	<=0.3	47.5	51.0	11.8	1.5	1730	5
01-103	2001	M	Sub-adult	87.0	<=0.8	66.7	2.2	3012.0	6.6	29.7	2.5	428.0	550.0	<=0.2	89.4	110.0	21.1	2.3	4320	3 - 4
03-335	2003	M	Sub-adult	43.6	<=0.9	81.3	2.5	7647.0	5.4	54.3	6.3	1410.0	1964.0	<=0.2	310.0	411.0	67.6	4.9	11,960	1
04-098	2004	M	Sub-adult	61.5	<=0.8	46.6	1.8	2177.0	5.7	16.7	<=0.9	218.0	119.0	<=0.1	30.5	37.2	15.9	<=0.4	2670	5
97-149	1997	F	Sub-adult	38.0	1.6	119.0	3.3	4223.0	10.9	43.5	4.9	774.0	627.0	<=0.4	168.0	207.0	34.6	5.2	6220	NR
98-193	1998	F	Sub-adult	72.7	<=0.9	113.0	2.3	5218.0	9.7	45.8	5.7	662.0	832.0	<=0.6	177.0	231.0	44.8	5.9	7350	NR
98-250	1998	F	Sub-adult	34.5	2.8	332.0	4.2	8761.0	12.0	109.0	7.4	1067.0	2136.0	nd	197.0	319.0	59.3	3.7	13,010	NR
00-050	2000	F	Sub-adult	10.2	<=0.5	49.9	1.0	1692.0	1.8	15.9	<=0.3	201.0	50.2	<=0.3	33.9	22.0	10.8	<=0.7	2080	2
00-056	2000	F	Sub-adult	5.0	<=0.4	23.6	<=0.9	2639.0	2.0	12.6	1.7	517.0	722.0	<=0.2	110.0	173.0	27.1	3.9	4230	3
04-005	2004	F	Sub-adult	83.0	<=0.9	42.4	2.2	5273.0	10.2	43.2	2.2	1103.0	557.0	<=0.2	180.0	225.0	36.7	2.0	7480	4
04-064	2004	F	Sub-adult	97.1	<=0.6	27.8	1.1	1845.0	7.5	18.6	2.8	328.0	114.0	<=0.2	60.4	59.0	13.4	<=0.6	2480	6
05-015	2005	F	Sub-adult	91.2	<=0.6	28.3	1.4	1505.0	6.2	13.6	1.5	227.0	115.0	<=0.4	39.0	38.3	8.7	<=0.7	1980	3
05-381	2005	F	Sub-adult	80.0	<=0.3	19.0	<=0.9	1178.0	4.1	7.1	1.2	169.0	295.0	<=0.1	49.1	52.1	12.6	<=0.9	1790	NR
179415	1994	M	Adult	2.4	<=0.1	4.4	1.9	546.0	<=0.2	4.5	<=0.4	47.7	223.0	nd	35.6	77.3	25.3	2.1	968	NR
178934	1994	M	Adult	100.0	<=0.9	59.3	2.4	2116.0	7.6	38.0	3.5	537.0	523.0	<=0.1	107.0	142.0	28.8	9.5	3570	NR
98-253	1998	M	Adult	4.2	<=0.2	19.0	1.6	3782.0	3.1	15.6	2.8	1724.0	1400.0	<=0.4	518.0	509.0	72.9	16.8	8060	4
04-229	2004	M	Adult	30.2	<=0.4	50.9	2.0	6530.0	10.6	38.4	6.5	1468.0	1789.0	<=0.2	301.0	335.0	54.6	6.0	10,590	1
06-001	2006	M	Adult	54.9	1.7	109.0	4.4	5049.0	11.9	44.4	11.1	497.0	1606.0	<=0.4	160.0	224.0	49.0	2.7	7770	NR
97-009	1997	F	Adult	45.6	<=0.2	25.4	1.1	1104.0	2.3	19.0	1.0	215.0	300.0	<=0.5	63.2	70.0	13.5	4.4	1820	5 - 6
97-001	1997	F	Adult	72.7	1.0	46.5	1.6	827.0	13.6	31.2	1.9	155.0	202.0	<=0.2	42.5	63.0	13.6	4.0	1400	3
97-082	1997	F	Adult	86.3	1.8	81.6	2.8	2700.0	13.1	40.4	7.0	542.0	675.0	<=0.4	122.0	159.0	26.5	4.3	4360	NR
00-054	2000	F	Adult	49.9	<=0.1	2.8	<=0.7	302.0	<=0.3	1.5	<=0.1	7.1	16.0	nd	3.5	1.1	<=0.8	<=0.1	334	NR
00-068	2000	F	Adult	82.1	1.0	137.0	1.5	2624.0	10.5	66.1	4.8	414.0	493.0	<=0.2	57.2	76.6	13.8	2.9	3900	NR
00-106	2000	F	Adult	7.2	<=0.3	17.8	<=0.8	574.0	4.0	18.9	<=0.8	173.0	226.0	<=0.2	45.8	73.8	13.1	1.7	1150	2
01-141	2001	F	Adult	56.9	2.2	143.0	6.5	5125.0	12.4	55.0	7.6	758.0	1402.0	<=0.4	252.0	246.0	56.4	10.0	8080	NR
01-122	2001	F	Adult	70.9	<=0.6	18.9	<=0.8	734.0	8.6	15.2	1.5	133.0	195.0	<=0.2	38.8	73.9	12.9	2.3	1240	2 - 3
01-101	2001	F	Adult	71.6	1.1	32.0	1.1	1559.0	16.3	34.7	5.3	348.0	320.0	<=0.3	64.5	95.6	13.5	6.0	2500	1 - 2
03-166	2003	F	Adult	98.9	<=0.5	29.5	2.4	415.0	10.9	14.1	1.4	86.6	143.0	<=0.2	31.1	37.3	7.1	2.0	777	NR
03-191	2003	F	Adult	69.4	<=0.8	16.5	<=0.9	376.0	9.2	10.9	1.4	103.0	141.0	<=0.1	25.5	45.7	7.0	3.0	738	NR
04-216	2004	F	Adult	48.5	<=0.8	87.4	2.9	4701.0	8.7	44.4	8.7	918.0	1099.0	<=0.3	205.0	308.0	73.4	6.1	7460	3 - 4
04-109	2004	F	Adult	80.5	<=0.3	4.4	2.6	118.0	5.7	4.5	<=0.5	21.9	36.7	<=0.2	8.4	12.1	3.1	1.1	217	6
04-238	2004	F	Adult	70.2	1.1	60.4	2.0	3527.0	7.8	26.4	3.4	425.0	437.0	<=0.1	77.1	102.0	13.8	1.0	4680	4
05-398	2005	F	Adult	84.7	2.3	24.9	1.5	673.0	4.1	11.0	1.6	88.7	201.0	<=0.1	21.2	28.2	21.2	<=0.7	1080	NR

Table 3. Congener-specific and total PBDE concentrations (ng/g wet weight) in blubber of Pacific harbor seal (*Phoca vitulina*) stranded along the southern California (USA) coast from 1994 to 2006 (n = 9). Body condition was assessed based on blubber thickness during the necropsy using the following scale: 1 = emaciated (2 mm or less); 2 = thin; 3 = fair (1 - 2 cm); 4, 5 = good (2 - 3 cm); 6 = excellent (3 - 4 cm); NR = not recorded; and nd = not detected.

Sample ID	Collection Year	Sex	Age Class	% Lipid	15	28	33	47	49	66	75	99	100	119	153	154	155	183	∑PBDE	Body Condition
95-070	1995	M	Pup	51.1	<=0.2	1.9	1.6	123.0	1.0	2.3	<=0.1	20.7	8.4	nd	4.0	<=0.8	<=0.6	<=0.2	162	2 -3
95-089	1995	M	Pup	22.3	<=0.3	5.7	1.6	1139.0	4.2	5.5	<=0.3	298.0	52.4	<=0.2	85.9	8.9	3.4	<=0.9	1610	2
01-094	2001	M	Pup	46.8	<=0.5	6.3	1.3	1161.0	5.0	4.2	<=0.7	253.0	72.1	<=0.3	43.8	7.1	4.1	<=0.7	1560	4
01-063	2001	M	Pup	76.2	<=0.7	11.1	1.6	1899.0	8.0	3.8	1.1	295.0	107.0	<=0.2	72.3	8.9	4.5	<=0.6	2410	5
03-308	2003	M	Pup	89.8	<=0.5	3.4	1.2	577.0	3.5	3.0	<=0.5	49.6	64.3	<=0.2	15.0	8.3	3.4	<=0.2	729	NR
95-073	1995	F	Pup	28.5	<=0.4	3.7	2.0	500.0	3.2	3.2	<=0.7	117.0	67.0	<=0.3	40.8	12.9	4.1	1.2	754	3 - 4
97-058	1997	F	Pup	86.2	1.1	6.4	1.7	739.0	3.5	2.6	<=0.6	62.0	37.1	<=0.2	14.3	3.3	1.9	<=0.5	873	2
03-328	2003	F	Pup	57.9	<=0.2	3.8	1.1	402.0	3.9	2.7	<=0.5	80.6	30.6	<=0.1	12.9	4.1	1.2	<=0.3	543	4 - 5
98-241	1998	M	Adult	7.3	nd	<=0.2	<=0.9	14.8	<=0.1	<=0.5	<=0.1	7.4	2.6	nd	2.1	1.1	<=0.1	<=0.4	26.9	1

Table 4. Congener-specific and total PBDE concentrations (ng/g wet weight) in blubber of Northern elephant seal pups (*Mirounga angustirostris*) stranded along the southern California (USA) coast from 1994 to 2006 (n = 16). Body condition was assessed based on blubber thickness during the necropsy using the following scale: 1 = emaciated (2 mm or less); 2 = thin; 3 = fair (1 - 2 cm); 4, 5 = good (2 - 3 cm); 6 = excellent (3 - 4 cm); NR = not recorded; and nd = not detected.

Sample ID	Collection Year	Sex	Age Class	% Lipid	15	28	33	47	49	66	75	99	100	119	153	154	155	183	∑PBDE	Body Condition
97-045	1997	M	Pup	88.6	<=0.2	3.6	3.1	117	1.6	3.3	<=0.8	15.2	15.6	<=0.1	2	3.4	1.2	<=0.4	166	1 - 2
97-032	1997	M	Pup	80.8	<=0.1	<=0.3	2.5	20.8	<=0.5	1.9	<=0.3	3.5	2.2	nd	<=0.6	<=0.7	<=0.2	<=0.4	30.9	NR
99-016	1999	M	Pup	53.5	<=0.3	<=0.7	2.8	42.1	<=0.6	1.7	<=0.4	11.1	4.1	<=0.1	1.3	1.5	1.4	<=0.2	65.9	NR
00-025	2000	M	Pup	0.8	<=0.1	<=0.3	1.4	9.5	<=0.1	1.5	<=0.3	2.9	1.3	1.5	<=0.4	<=0.4	nd	<=0.3	16.6	3
01-059	2001	M	Pup	95.6	<=0.1	<=0.6	2.4	38.4	<=0.4	2.1	<=0.2	9.5	4.1	<=0.1	<=0.9	1.2	<=0.8	<=0.3	56.4	6
05-084	2005	M	Pup	43.3	<=0.1	<=0.1	<=0.5	21.6	<=0.1	<=0.3	Nd	2.3	2.6	nd	<=0.3	<=0.6	1.2	<=0.2	27.7	NR
05-046	2005	M	Pup	21	nd	<=0.2	<=0.8	21.4	1.5	1	Nd	3	1.8	nd	<=0.3	<=0.7	<=0.5	<=0.1	27.7	NR
98-169	1998	F	Pup	90.4	<=0.1	4.2	1.3	69.1	1.5	3.1	<=0.5	15.5	13.1	nd	1.7	1.9	<=0.7	<=0.3	112	6
00-031	2000	F	Pup	72.2	<=0.4	19.7	2.3	322	5.5	8.6	<=0.9	29.1	53.8	<=0.1	3.7	5	2.1	<=0.5	452	5
00-012	2000	F	Pup	50.1	<=0.2	<=0.6	1.2	55.9	<=0.1	1.4	<=0.1	6.4	5	nd	<=0.6	1.1	<=0.4	<=0.3	70.9	3 - 4
01-099	2001	F	Pup	55.5	<=0.3	<=0.9	3.6	67.2	1	1.9	1	9.4	8.3	<=0.1	1.6	2.7	<=0.6	<=0.4	94.7	1
01-049	2001	F	Pup	63.7	<=0.3	1.1	3	76.5	<=0.6	2	<=0.3	11.5	8.5	<=0.5	1.4	2.2	<=0.7	<=0.4	105	4 - 5
04-057	2004	F	Pup	55.1	<=0.3	<=0.6	2.2	64.6	<=0.2	2	<=0.3	8.6	5.1	<=0.1	<=0.4	1.9	1.1	<=0.4	84.3	2 - 3
04-031	2004	F	Pup	0.5	<=0.1	<=0.1	1.2	3.1	<=0.2	1.4	nd	1.6	0.5	nd	<=0.3	<=0.2	<=0.1	<=0.3	7.2	NR
04-062	2004	F	Pup	67.2	<=0.2	2.2	<=0.5	20.6	<=0.3	1.6	<=0.9	3	2.1	<=0.1	<=0.4	<=0.9	<=0.4	<=0.1	29.5	2
05-031	2005	F	Pup	10.4	nd	<=0.1	<=0.6	16.6	<=0.1	<=0.9	nd	2.5	1.5	<=0.1	<=0.2	<=0.7	<=0.1	<=0.1	20.5	NR

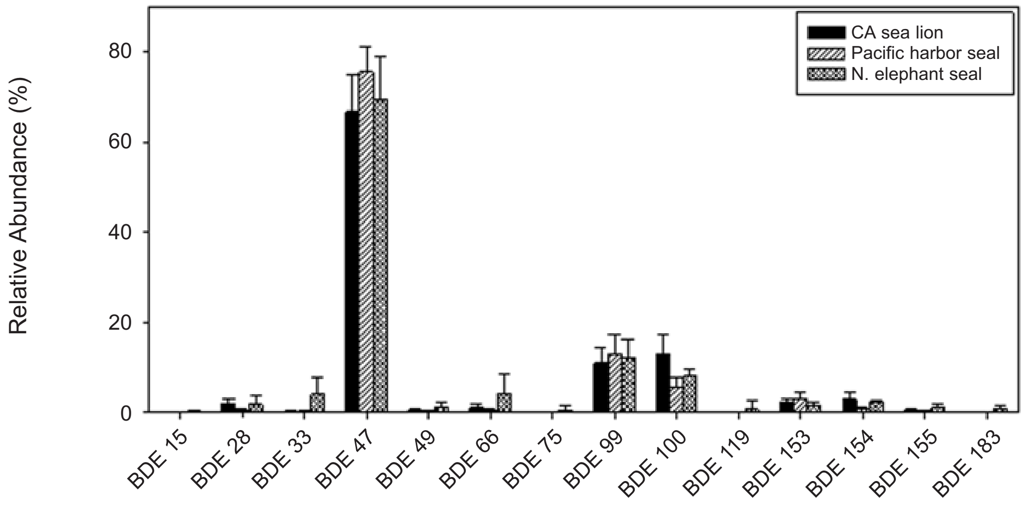

Figure 1. Mean relative abundance of selected PBDE congeners in blubber of stranded California sea lions, Pacific harbor seals and northern elephant seals (*n* = 88). Error bars represent standard deviation.

exposed to similar loads of PBDEs, and exhibit similar biotransformation potential and growth dilution rates. For adult sea lions, similar differences in levels of POPs have been reported for several marine mammal species (Lindström *et al*. 1999, Thron *et al*. 2004, Tuerk *et al*. 2005, Fair *et al*. 2007). That a gender difference in PBDE levels was only observed for adult sea lions is yet another example of a reduction in POP levels for reproductively mature female marine mammals, presumably due to reproductive offloading (Ikonomou *et al*. 2002). These investigators also found no difference in PBDE levels (both

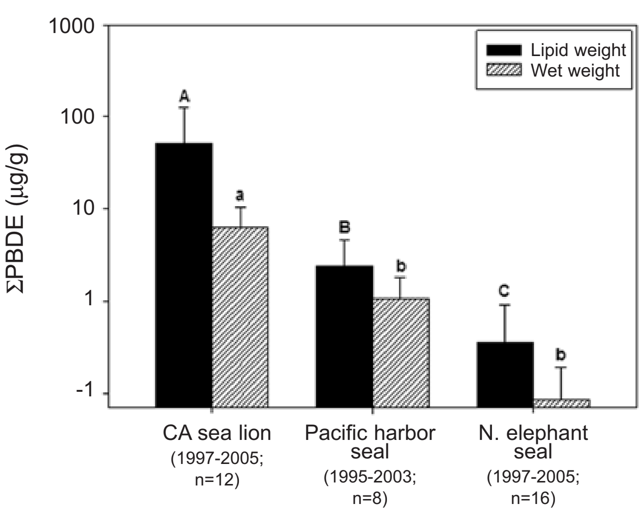

Figure 2. ΣPBDE (µg/g wet or lipid weight, mean ± standard deviation) in blubber of California sea lion, Pacific harbor seal, and northern elephant seal pups (<1 year old). Means with different letters are significantly different ($\rho \leq 0.05$).

total and individual congener) between younger (0 - 15 year-old) and older (16 - 35 year-old) male seals. Similarly, no age variation was found previously for harbor seals, dolphins, or polar bears (She *et al*. 2002, Muir *et al*. 2006, Fair *et al*. 2007). One possible explanation for the absence of age-related differences is the relatively recent introduction of PBDEs into the biosphere (Fair *et al*. 2007, Ikonomou and Addison 2008).

Temporal trends of POPs (including PBDEs) in biota may vary based on geography, climate and environmental conditions as well as from changing contaminant loads and accessibility. Increasing PBDE levels were observed in ringed seals from 1981 to 2000, and from 1982 to 2006; in harbor seals from 1989 to 1998; and in beluga whales collected between 1988 and 1999 (Ikonomou *et al*. 2002, She *et al*. 2002, Lebeuf *et al*. 2004, Vorkamp *et al*. 2008). In contrast, no temporal trends for PBDEs were observed for California sea lions or sea otters collected during a time period similar to the present study (Stapleton *et al*. 2006, Kannan *et al* 2007). Tuerk *et al*. (2005) also reported no discernable temporal trend for juvenile white-sided dolphins from the northwestern Atlantic. An 150-fold increase in PBDE in fur seals from 1972 to 1994 was followed by an apparent decrease from 1994 to 1998 (Kajiwara *et al*. 2004). A similar trend was recently reported using archived freeze-dried mussels from the Seine estuary, France, where PBDEs increased exponentially during 1982-1993, leveled off in 1999-2001, and then began to decline after 2002 (Law *et*

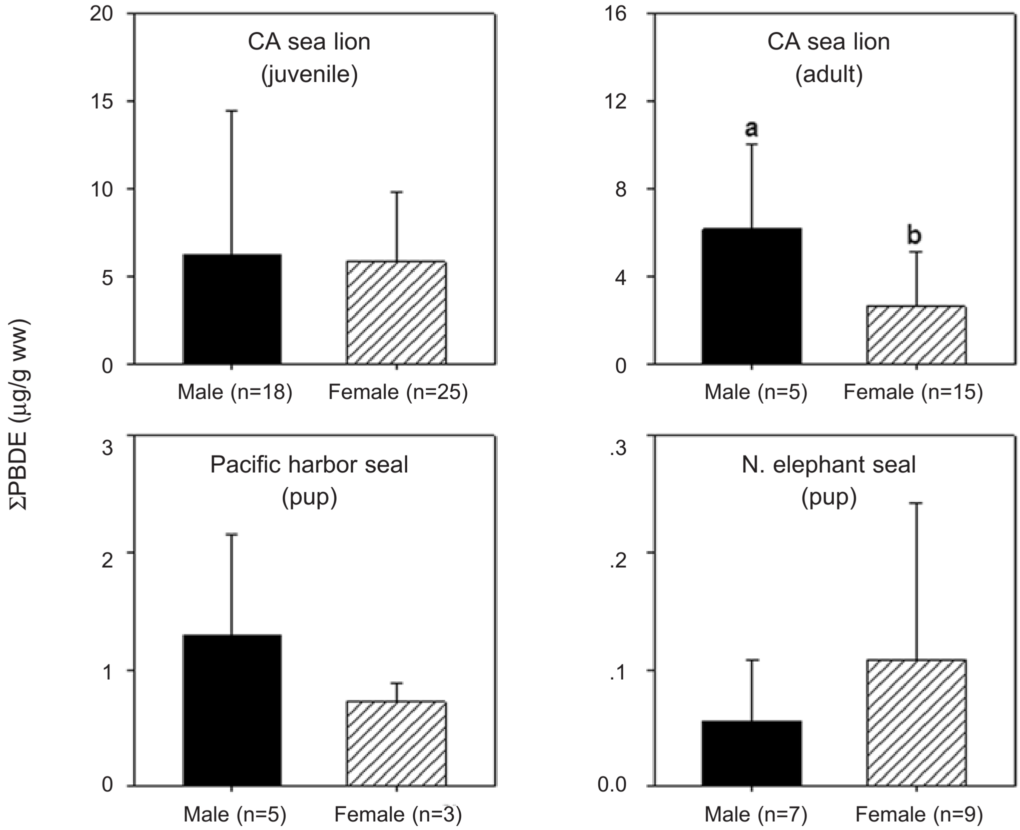

Figure 3. ΣPBDE (µg/g wet weight, mean ± standard deviation) for female and male California sea lions, Pacific harbor seal pups, and northern elephant seal pups. Means with different letters are significantly different ($\rho = 0.05$). See Table 1 for age classes (juvenile = pup, yearling and sub-adult categories).

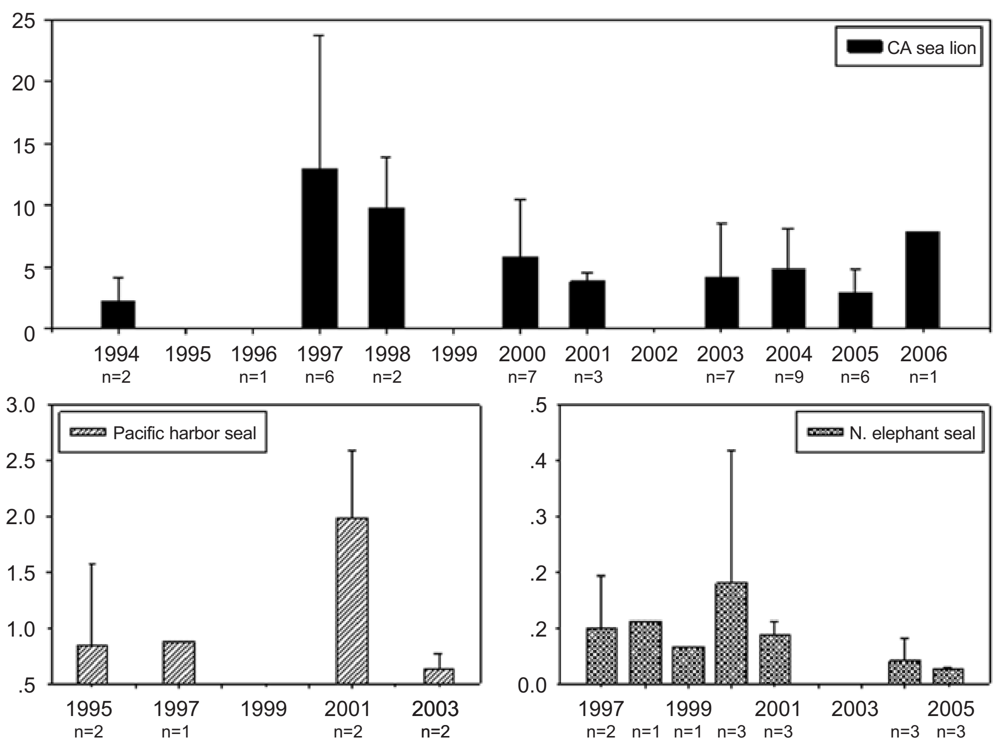

Figure 4. ΣPBDE (µg/g wet weight, mean ± standard deviation) by stranding year and pinniped species. Data are for pups (<1 year old) except for California sea lion (juveniles and adult males also included).

al. 2006). The present study's results are not inconsistent with the latter scenario in that levels do not appear to be rapidly increasing nor decreasing.

Although La Guardia *et al*. (2006) found BDE 99 (45 - 49%) to be the most abundant component in penta-BDE products DE-71 and 70-5DE used largely in North America, it was found in lower proportions (11 - 13%) in the present study's samples. As noted by others, this may be a consequence of preferential uptake of lower brominated congeners (e.g., BDE 47), or by its presence in lower proportions in the environment (She *et al*. 2002). Another possible reason for the present findings is that marine mammals may debrominate BDE 99 into BDE 47, as was reported for carp in laboratory exposures (Stapleton *et al*., 2004). The proportion of BDE 100 in the present study's blubber samples was similar to that reported in unmodified penta-BDE (~8 - 13%). Correspondingly, Stapleton *et al*. (2006) reported similar proportions of BDE 99 (12%) and BDE 100 (20%) in California sea lions. In contrast, harbor seals in the present study contained twice as much BDE 99 (13%) as BDE 100 (5.6%; Figure 1), consistent with previous data for the same species (12% vs 5%; She *et al*. 2002). The overall similarity in BDE congener pattern suggests a commonality in the exposure "profile" and elimination capability among the pinniped species targeted in the present study.

In the absence of differential biological uptake/elimination and knowledge of PBDE levels in prey species preferred by the three pinniped species, the difference in PBDE levels among species (Figure 2) indicates a clear gradient in the magnitude of exposure. By comparison, total PCBs were similar to ΣPBDEs, whereas DDTs were one to two orders of magnitude higher in these samples (Blasius and Goodmanlowe, 2008). In fact, ΣPBDE was positively correlated with matched total PCB and organochlorine pesticide levels ($r^2 = 0.61$ and 0.60, respectively). Moreover, the species-specific trend reported previously by these authors for organochlorines (e.g., sea lion > harbor seal > elephant seal) was identical to that observed herein for PBDEs. Coupled with knowledge of their life histories (similar diets, but different geographical range and local habitat), it is not suprising that sea lions stranded in the southern California region are exposed to the highest levels of PBDEs.

Although PBDE levels for California sea lions stranded along the southern California coast are the highest reported to date, there are currently no published or universally acknowledged concentration thresholds for marine (or terrestrial) mammals by which to gauge their potential for effects. Because PBDE concentrations and lipid content (one indicator of animal condition at the time of death) varied over many orders of magnitude in the present study, the role of PBDE contamination on regional strandings, and on pinniped health in general remains unclear. Superimposed onto this unresolved question is the potential for effects from (or in combination with) other stressors, physical, biological, ecological, and chemical in nature. Based on the results of this study, however, it is clear that the California sea lion would serve as an excellent indicator species for future studies to elucidate local or regional contamination sources (e.g., contaminated sediments), pathways for biomagnification, and long-term temporal trends in PBDE contamination throughout the region, as well as in highly urbanized coastal systems where they are locally abundant.

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