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Contaminants still high in top-level carnivores in the Southern California Bight: Levels of DDT and PCBs in resident and transient pinnipeds

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ABSTRACT

Highly industrialized areas, such as the Southern California Bight, often have high levels of contaminants in marine sediments, which can cause chronic exposure to organisms long after their use has ceased. tDDT and tPCB were analyzed in the blubber of 145 stranded pinnipeds that died at local marine mammal centers between 1994 and 2006. Resident species (California sea lion and Pacific harbor seal) had significantly higher concentrations of tDDT and tPCB than the transient species (northern elephant seal). Adult female California sea lions had significantly lower concentrations of tDDT and tPCB than pups, yearlings, and adult males. Concentrations of both tDDT and tPCB in California sea lions significantly declined over time, but did not change in northern elephant seals. Current concentrations of tDDT and tPCB in California sea lions and harbor seals are among the highest values reported worldwide for marine mammals and exceed those reported to cause adverse health effects.

1. Introduction

Many highly industrialized areas have repositories of organochlorines (OCs), such as dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCBs), which through bioturbation and resuspension of sediments act as chronic sources of these compounds long after their use has ceased (Zeng and Venkatesan, 1999). The coast adjacent to the Southern California Bight (SCB; Fig. 1) is one such example where large amounts organic pollutants were discharged into the Pacific Ocean off the Palos Verdes Shelf (PVS) from 1949 to 1970s. By 1993 it was determined that over 11 tons of PCBs and 110 tons of DDT remained in the SCB (Lee et al., 2002) and these high levels of contaminants have been shown to have deleterious effects on local marine life (Anderson et al., 1975; Gilmartin et al., 1976).

One of the biological problems associated with OCs is that they are lipophilic, are resistant to metabolic processes, and therefore, are bioaccumulated. Because pinnipeds have a high body fat content, occur high trophically, and are long lived, they are useful sentinel species for monitoring lipid-soluble contaminants (Kannan et al., 2004; Fossi et al., 2003). The accumulation of these contaminants also makes pinnipeds highly susceptible to a variety of cancers, and the impairment of immune, reproductive, developmental, and endocrine systems (de Swart et al., 1996).

Three species of pinnipeds commonly inhabit SCB waters: the California sea lion (CA sea lion; *Zalophus californianus californianus*), the Pacific harbor seal (*Phoca vitulina richardii*), and the northern elephant seal (*Mirounga angustirostris*). Each species differs in their residency patterns within the SCB, differs in their foraging locations (nearshore vs. offshore) and foraging latitude (mid vs. high), and specializes on different prey (Bartholomew and Boolootian, 1960; Antonelis and Fiscus, 1980; Le Boeuf et al., 2000). Pacific harbor seals are the most resident of the three species in the SCB, making only localized movements in search of food (Antonelis and Fiscus, 1980), while northern elephant seals are the most transient, mainly using the SCB for breeding and molting, but feeding offshore and as far north as Alaska (Stewart and DeLong, 1995; Le Boeuf et al., 2000). California sea lions have the most variable residency pattern in the SCB: juveniles and adult males travel as far north as British Columbia in the winter to feed but return to the SCB in the spring to breed, while adult females remain near their natal rookeries in the SCB to nurse their pups for 6–11 months (Bartholomew and Boolootian, 1960), and consequently spend most of their time feeding in the SCB. Whereas both CA sea lions and Pacific harbor seals are known to be opportunistic feeders preying on locally abundant fish and cephalopods (Antonelis and Fiscus, 1980; Lowry et al., 1990), northern elephant seals appear to prey mainly on cephalopods along with a few fish and elasmobranch species (Condit and Le Boeuf, 1984).

Despite the highly publicized dumping of DDT off the PVS from the late 1940s to its subsequent agricultural ban in 1972, relatively few studies have been conducted on the levels of DDT and PCBs in the pinnipeds specifically residing and feeding in the SCB where the highest dumping in CA occurred, even though many papers have been published on contaminants in pinnipeds found in other areas of California (Le Boeuf and Bonnell, 1971; DeLong et al., 1973; Gilmartin et al., 1976; Bacon et al., 1992; Lieberg-Clark

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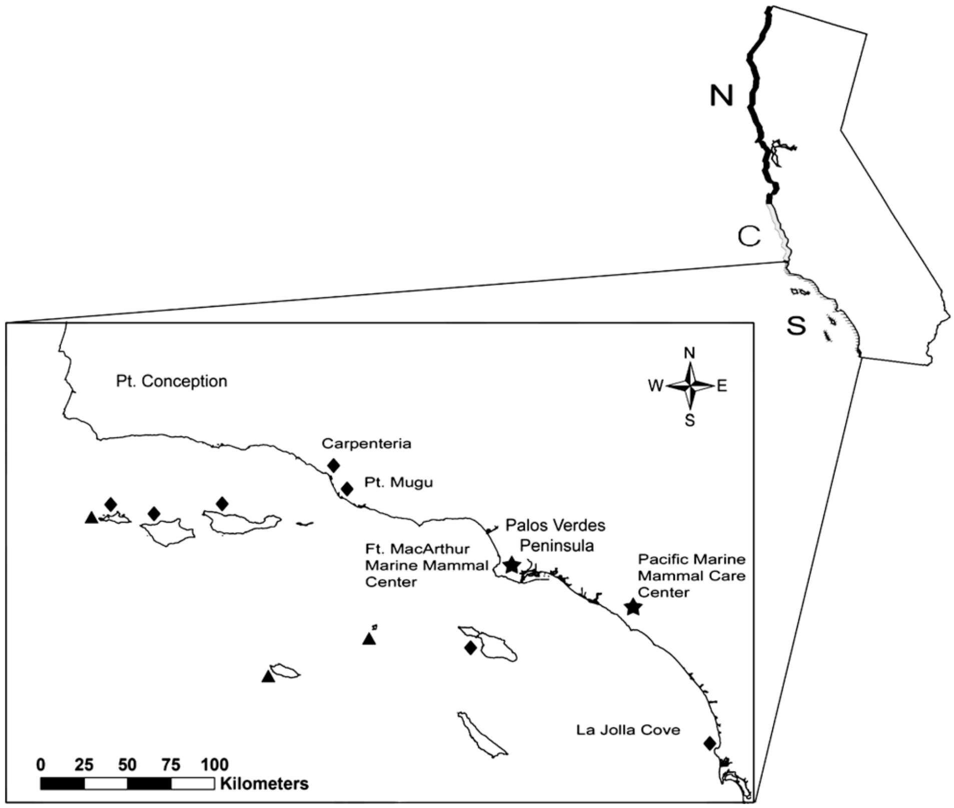

Fig. 1. The map of the Southern California Bight. *N* = north, *C* = central, and *S* = south. ▲ Indicates major California sea lion rookeries (Bartholomew and Boolootian, 1960), and ♦ indicates major Pacific harbor seal haul-out sites (Bartholomew and Boolootian, 1960; Joe Cordaro, personal communication.). ★ Indicates locations of the two marine mammal centers where samples were collected: Fort MacArthur Marine Mammal Center located in San Pedro, CA, and Pacific Marine Mammal Center located in Laguna Beach, CA.

et al., 1995; Beckman et al., 1997; Kajiwara et al., 2001; Le Boeuf et al., 2002; Kannan et al., 2004; Debier et al., 2005; Ylitalo et al., 2005; Debier et al. 2006; Greig et al., 2007). Furthermore, while both Le Boeuf et al. (2002) and Kannan et al. (2004) examined contaminants in CA sea lions from the SCB, these studies were based on the same seven animals that stranded off the Channel Islands, which is located several miles from the PVS and the contaminant hotspot, and no studies to date have examined contaminants found in either Pacific harbor seals or northern elephant seals within the SCB. Our study is the first to examine these three pinniped species concurrently, exclusively from the SCB, and over a 13 year time period.

The goals of this study were to investigate: (i) the levels of OCs within CA sea lions, Pacific harbor seals, and northern elephant seals, which may be expected to contain differing levels of contaminants due to differences in preferred prey, residency, and habitat use; (ii) the variation in the levels of these compounds that may occur with age and sex, since this trend has been shown in other species; and (iii) the temporal trends in tDDT and tPCB concentrations over a 13 year period (1994–2006), since the production of DDT and PCBs ended in the 1970s.

2. Materials and methods

2.1. Sample collection

The blubber of 145 pinnipeds (92 CA sea lions, 11 Pacific harbor seals, and 42 northern elephant seals; Table 1) was acquired from the Fort MacArthur Marine Mammal Center in San Pedro ($n = 125$), CA, and the Pacific Marine Mammal Center in Laguna Beach, CA ($n = 20$). Blubber samples were collected from 1994 to 2006 by Center personnel from pinnipeds that died at the Centers after stranding events. Necropsies were performed at the marine mammal centers, and blubber samples were collected and frozen at −20 °C for preservation. Detailed information about each animal, including collection date, species, sex, age class, and cause of death (postmortem diagnosis) was catalogued by the Center personnel. Causes of death for the pinnipeds in this study included respiratory disease (11.7%), trauma (11.0%), domoic acid toxicity (5.8%), infectious disease (4.4%), carcinoma (3.6%), metabolic disorder (0.7%), and unknown or unidentified (62.8%). Animals were assigned to age classes based on the approximate length and maturity of the animal, as pup, yearling, subadult, and adult (Jefferson et al., 1993).

2.2. Chemical analysis

Sample extracts were obtained from blubber using the microwave extraction procedure of EPA Method 3546 (2007), Hummert et al. (1996). Approximately 1.0 g of blubber and 25 mL of dichloromethane (DCM) were placed in a double-walled teflon vessel and the extraction was performed in a microwave at 100 °C with a heating time of 15 min, and repeated three times to ensure 99.9% extraction efficiency (Sun et al., 2004). Lipid content was determined gravimetrically from split aliquots of the extracts after removing the DCM (Bligh and Dyer, 1959). Sample cleanup of the extracts was accomplished by elution through a chromatographic column packed with 3% water deactivated silica gel capped with basic alumina. The target compounds were eluted from the column with 15 mL of hexane and 30 mL of 30% DCM/hexane.

Each extract was analyzed and concentrations of individually resolved peaks of OCs were summed to obtain total PCB (tPCB) concentrations (47 PCB congeners: −18, −28, −52, −49, −44,

Table 1
Sample sizes and years of blubber collection from the stranding centers in southern California by species and age of pinniped ($n = 145$)

Species	Pup (M&F) $n = 57$	Yearling (M&F) $n = 30$	Sub-adult (M&F) $n = 23$	Adult female $n = 24$	Adult male $n = 6$	Age class unknown $n = 5$	Year $n = 145$
California sea lion (*Zalophus californianus californianus*) $n = 92$	$n = 14$ 1997 ($n = 1$) 1998 ($n = 2$) 2000 ($n = 3$) 2001 ($n = 1$) 2003 ($n = 4$) 2004 ($n = 2$) 2005 ($n = 1$)	$n = 26$ 1996 ($n = 1$) 1997 ($n = 5$) 1998 ($n = 1$) 2000 ($n = 1$) 2001 ($n = 1$) 2002 ($n = 4$) 2003 ($n = 3$) 2004 ($n = 4$) 2005 ($n = 6$)	$n = 23$ 1997 ($n = 2$) 1998 ($n = 3$) 1999 ($n = 4$) 2000 ($n = 5$) 2001 ($n = 1$) 2002 ($n = 1$) 2003 ($n = 1$) 2004 ($n = 4$) 2005 ($n = 2$)	$n = 24$ 1997 ($n = 3$) 2000 ($n = 4$) 2001 ($n = 9$) 2003 ($n = 2$) 2004 ($n = 5$) 2005 ($n = 1$)	$n = 5$ 1994 ($n = 2$) 1998 ($n = 1$) 2004 ($n = 1$) 2006 ($n = 1$)	$n = 0$	1994 ($n = 2$) 1995 ($n = 4$) 1996 ($n = 1$) 1997 ($n = 14$) 1998 ($n = 9$) 1999 ($n = 5$) 2000 ($n = 23$) 2001 ($n = 24$) 2002 ($n = 9$) 2003 ($n = 13$)
Pacific harbor seal (*Phoca vitulina richardsii*) $n = 11$	$n = 9$ 1995 ($n = 4$) 1997 ($n = 1$) 2001 ($n = 2$) 2003 ($n = 2$)	$n = 1$ 2003 ($n = 1$)	$n = 0$	$n = 0$	$n = 1$ 1998 ($n = 1$)	$n = 0$	2004 ($n = 20$) 2005 ($n = 20$) 2006 ($n = 1$)
Northern elephant seal (*Mirounga angustirostris*) $n = 42$	$n = 34$ 1997 ($n = 2$) 1998 ($n = 1$) 1999 ($n = 1$) 2000 ($n = 7$) 2001 ($n = 9$) 2004 ($n = 4$) 2005 ($n = 10$) 2000 ($n = 3$) 2001 ($n = 1$) 2002 ($n = 4$)	$n = 3$	$n = 0$	$n = 0$	$n = 0$	$n = 5$	

−37, −74, −70, −66, −101, −99, −119, −87, −77, −110, −81, −151, −149, −123, −118, −114, −153, −168, −105, −138, −158, −126, −187, −183, −128, −167, −177, −156, −200, −157, −180, −169, −170, −201, −194, −189, and −206; Schulz et al., 1989). Similarly total DDT (tDDT) was calculated from the sum of 2,4′-DDE, 4,4′-DDE, 2,4′-DDD, 4,4′-DDD, 2,4′-DDT, 4,4′-DDT; chlordanes (tCHL) refers to sum of heptachlor, heptachlor epoxide, chlordane-alpha, chlordane-gamma, *cis*-nonachlor, and *trans*-nonachlor; and hexachlorocyclohexane (tHCHs) refers to sum of α-, β-, γ-, and δ-isomers. Aldrin, dieldrin, endrin, endosulfan I, endosulfan sulfate, endosulfan II, endrin aldehyde, endrin ketone, methoxychlor, and mirex were additionally analyzed.

For identification and quantification of contaminants, each sample was injected using an autosampler (7683B series, Agilent Technologies, Santa Clara, CA, USA) onto an Agilent gas chromatograph (6890N series) equipped with a mass selective detector (GCMS; Agilent 5973 inert series). The GC column employed was a ZB-5 (Phenomenex; Torrance, CA) fused silica capillary (0.25 mm ID × 60 m) with 0.25 μm film thickness. The GCMS operating conditions of EPA Method 8270D (2007) were followed. The GCMS was used with a Hewlett Packard PC equipped with Agilent ChemStation (Rev. D.01.02.16 15 June 2004). Detection limits were defined as three times the standard deviation of the background noise, and were all in the range of 1.0 ng/g.

2.3. Quality assurance/Quality control (QA/QC)

To examine the quality of data obtained by the analytical procedure, 100 μl of PCB and pesticide surrogate recovery standards (tetrachloro-*m*-xylene, PCB30, PCB112, and PCB198; AccuStandard, New Haven, Connecticut) were spiked into one gram of blubber (Muir and Morita, 2003). The recovery rates of the surrogates were in the acceptable ranges 70–130% (EPA Method 8000B, 1996). Method blanks were analyzed simultaneously with each extraction (every 12 samples) to check for interferences or contamination from glassware and solvents (Muir and Morita, 2003). Analysis of duplicate samples was used to assess the variability introduced by the sampling process and sample matrix homogeneity. Sample duplicates were split during the extraction process. The sample duplicates were treated as independent samples during laboratory preparation processes and analysis. Approximately 15% of the samples were analyzed as duplicates, which exceeds the EPA standard performance recommendation of 5% of samples analyzed in duplicate (EPA Method 8000B, 1996). The duplicates' relative percent difference (RPD) yielded an average of 20%, which falls below the EPA standard performance recommendation of a RPD less than 30% (EPA Method 8000B, 1996).

2.4. Data analysis

Values for duplicate samples analyzed were averaged for data summaries and statistical analyses. Values below detection limits were treated as zero values. All OC concentration data were log-transformed to achieve normality. All statistical analyses were performed using Statistica 7.1 (StatSoft, Inc. 1984–2006; version 7.1.515.0). Differences were considered significant at $\alpha = 0.05$.

The relationships between OC wet weight concentrations and blubber lipid content were examined via linear regression. Because lipid content varies with the nutritive condition of the animal in addition to the methods used for lipid extraction, and therefore may influence pollutant levels in individuals (Debier et al., 2006), all analyte values for samples were expressed both on a wet weight basis (e.g., tPCB per unit mass of blubber) and on a lipid weight basis (e.g., tPCB per unit of extractable blubber lipids). However, because there were no differences in levels of either of these contaminants when expressed on either a wet weight basis, lipid weight basis, or for samples containing less than 20% lipid, only results standardized on a lipid weight basis are reported to allow our results to be more comparable with similar studies.

Differences among species, age/gender classes, and years were assessed by General linear model (GLM) followed by Tukey's comparison tests *a posteriori* with age, class, and gender as categorical variables and lipid content as a continuous variable. Interspecies differences were evaluated for all individuals (CA sea lions, $n = 92$; Pacific harbor seals, $n = 11$; northern elephant seals, $n = 42$) and for pups (CA sea lions, $n = 14$; Pacific harbor seals,

$n = 9$; northern elephant seals, $n = 34$). To examine contaminant differences between age and gender classes, only the CA sea lion samples were used, due to inadequate sample sizes across age and gender from the other two species. Temporal differences in contaminant levels were assessed for CA sea lions ($n = 92$) and for non-adult northern elephant seals ($n = 42$). Relatively few Pacific harbor seal samples were available for this study for a variety of reasons: the southern California harbor seal stock (∼ 32,000 seals) is relatively low compared to the other pinnipeds (∼ 300,000 CA sea lions, and ∼ 100,000 northern elephant seals). In addition, because harbor seals are non-migratory and show strong site fidelity for their haul-out sites, which are not located near the two stranding centers where samples were collected for this study (Fig. 1), fewer strandings of this species occur in the SCB (Joe Cordaro, personal communication.).

3. Results

3.1. Organochlorine concentrations

In the three species of pinnipeds inhabiting the SCB, total DDT (tDDT), total PCB (tPCB), and chlordane-related compounds (tCHL) were the major persistent OCs found relative to other contaminant groups such as hexachlorocyclohexane (HCH), aldrin, and dieldrin. Endrin, endosulfan I, and methoxychlor were below detection limits due to analytical interferences, while endosulfan II, endosulfan sulfate, endrin ketone, and mirex concentrations were not biologically detectable in any of the blubber samples.

tDDT and tPCB concentrations together contributed up to 99.1% of the total OCs measured (88.2% and 10.9%, respectively). tDDT was the predominant contaminant in all three pinniped species, with concentrations ranging from non-detectable to 13,271 μg/g on a lipid weight (lw) basis (mean ± 1 SD = 496.67 ± 1570.45; Table 2). This was followed by tPCB, which ranged from 0.06 to 2208 μg/g, lw (mea $n = 68.32 \pm 230.03$; Table 2), and tCHL, which ranged from non-detectable to 5770 μg/g, lw (44.41 ± 477.87; Table 2). tCHL contributed up to 0.42% of the total OC composition overall; it was highest in the blubber of northern elephant seals (1%), followed by CA sea lions (0.5%), and Pacific harbor seals (0.1%; Table 2). HCH made up 0.27% of the total OC composition overall; it was highest in the blubber of northern elephant seals (1.25%) followed by CA sea lions (0.25%), and harbor seals (0.05%; Table 2). The remaining chlorinated pesticides contributed up to 0.19% of the total OC composition overall.

Lipid content in the blubber was 47.88 ± 33.55 g/g, wet weight and varied widely from 0.01% to 99% (Table 2), due to the health condition of the animal. There was a significant negative relationship between both tDDT and tPCB concentrations and lipid content in the three pinniped species (tPCB: $r^2 = 0.17$, $p < 0.001$; tDDT: $r^2 = 0.10$, $p < 0.001$; Fig. 2), even when samples containing <20% lipid were omitted from the analysis ($p < 0.001$).

3.2. Species related effects

Mean concentrations of both tDDT and tPCB in blubber among the three pinniped species differed significantly (tDDT: $F = 11.61$; $df = 2, 100$; $p < 0.001$; tPCB: $F = 9.31$; $df = 2, 100$; $p < 0.001$; Fig. 3). CA sea lion and Pacific harbor seal blubber samples had higher concentrations of both tDDT and tPCB than northern elephant seals (Tukey's; $p < 0.05$ for tDDT and tPCB), but did not differ from each other (Tukey's; $p = 0.99$ for tDDT and tPCB; Fig. 3). It should be noted that Pacific harbor seal and elephant seal blubber samples were obtained mainly from pups, whereas CA sea lion blubber samples were obtained from all age classes. However, comparison of just the pup age class for each of the three species yielded the same relationship ($p < 0.05$).

3.3. Age and gender trends: CA sea lions

In mature CA sea lions, significant differences in tDDT and tPCB concentrations were found between genders; mean concentrations of tDDT and tPCB in females were 94.31% and 93.42% lower, respectively, than males (Tukey's; $p = 0.003$ for tDDT and tPCB; Fig. 4). Both tDDT and tPCB concentrations in mature females were significantly different from pups and yearlings, with mean concentrations in mature females 81.0% and 81.3% lower, respectively,

Table 2
Mean (±SD) tDDT, tPCB, tCHL, tHCH (μg/g, lipid weight) in blubber of California sea lions, Pacific harbor seals, and northern elephant seals collected in the Southern California Bight

Species	*n*	% Lipid mean ± S.D.	tDDT mean ± S.D.	tPCB mean ± S.D.	tCHL mean ± S.D.	tHCH mean ± S.D.
Overall	145	47.88 ± 33.55	469.67 ± 1570.45	68.32 ± 230.03	44.41 ± 477.87	2.02 ± 5.34
CA sea lion	92	54.95 ± 31.92	594.38 ± 1667.83	86.55 ± 263.18	68.51 ± 601.86	1.68 ± 3.90
Range (min–max)		(0.01–99.00)	(2.64–13,271.32)	(0.29–2208.20)	(ND–5770.78)	(ND–24.45)
Pup (F)	9	52.46 ± 39.97	1801.73 ± 4309.53	293.53 ± 719.64	648.28 ± 1920.98	1.02 ± 1.52
Pup (M)	5	25.47 ± 19.49	702.85 ± 956.27	87.33 ± 108.86	61.50 ± 132.90	1.88 ± 1.71
Yearling (F)	17	59.75 ± 30.60	636.72 ± 1160.17	87.29 ± 143.99	3.79 ± 6.51	3.16 ± 6.77
Yearling (M)	9	49.15 ± 38.83	481.23 ± 715.57	89.14 ± 127.05	4.77 ± 10.46	3.10 ± 5.41
Sub-adult (F)	13	53.23 ± 35.05	284.98 ± 346.27	35.16 ± 39.09	1.24 ± 1.61	1.39 ± 1.96
Sub-adult (M)	10	64.16 ± 23.83	165.62 ± 217.48	22.58 ± 29.44	0.72 ± 0.74	0.48 ± 0.71
Adult (F)	24	61.34 ± 26.83	129.27 ± 399.40	19.15 ± 55.05	0.24 ± 0.71	0.34 ± 0.91
Adult (M)	5	38.34 ± 40.62	2266.91 ± 2921.64	291.04 ± 440.69	4.90 ± 7.67	4.67 ± 6.23
Pacific harbor seal	11	48.64 ± 30.15	1041.17 ± 2989.44	123.19 ± 334.34	0.15 ± 0.18	0.07 ± 0.14
Range (min–max)		(7.33–89.79)	(9.98–10,031.82)	(1.19–1125.22)	(ND–0.47)	(ND–0.43)
Pup (F)	3	57.53 ± 28.85	85.52 ± 42.37	9.59 ± 8.67	0.03 ± 0.05	0.04 ± 0.07
Pup (M)	6	48.28 ± 32.24	1850.13 ± 4018.01	217.81 ± 447.07	0.23 ± 0.20	0.10 ± 0.18
Yearling (F)	1	65.44	85.53	18.31	0.19	ND
Adult (M)	1	7.32	9.98	1.19	ND	ND
Northern elephant seal	42	32.21 ± 33.29	46.82 ± 53.42	14.02 ± 16.87	3.21 ± 7.12	3.26 ± 8.01
Range (min–max)		(0.01–95.57)	(ND–226.34)	(0.06–75.96)	(ND–28.79)	(ND–28.90)
Pup (F)	15	36.12 ± 31.92	36.77 ± 38.05	11.55 ± 17.37	2.47 ± 5.92	2.04 ± 7.44
Pup (M)	18	32.21 ± 36.71	40.31 ± 44.32	12.96 ± 13.29	3.04 ± 6.89	3.40 ± 8.15
Pup (sex unknown)	1	0.01	226.34	75.96	ND	ND
Yearling (M)	3	58.18 ± 20.93	101.26 ± 94.74	13.21 ± 8.92	1.21 ± 1.93	2.39 ± 3.70
Age class unknown (M)	4	13.93 ± 24.39	30.13 ± 16.37	12.13 ± 9.99	9.86 ± 13.58	9.51 ± 12.82
Age/sex unknown	1	0.01	38.48	18.44	ND	ND

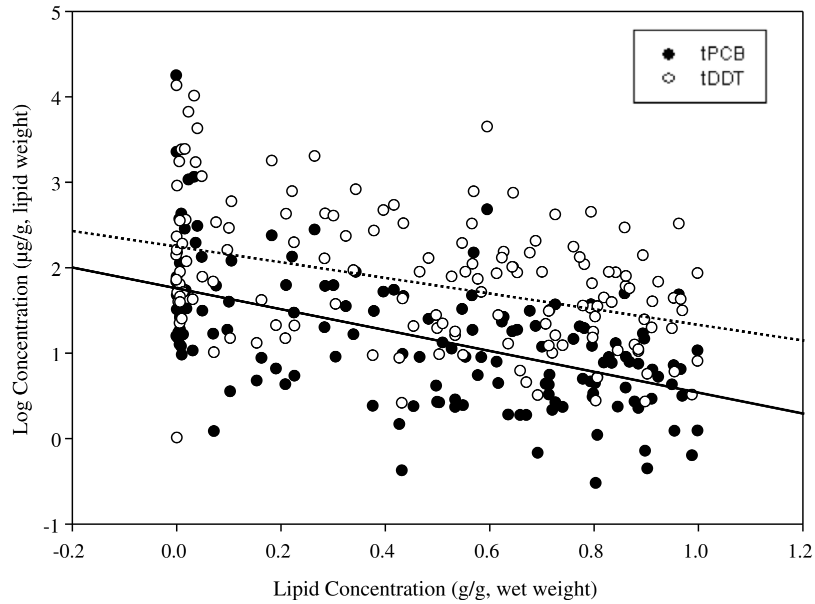

Fig. 2. Relationship between tDDT or tPCB concentrations (μg/g, lipid weight) and lipid concentration (g/g, wet weight) in blubber samples in all three species (California sea lion, Pacific harbor seal, and northern elephant seal) pooled together. The dashed line represents tDDT: $r^2 = 0.10$, $p < 0.001$; the solid line represents tPCB: $r^2 = 0.17$, $p < 0.001$.

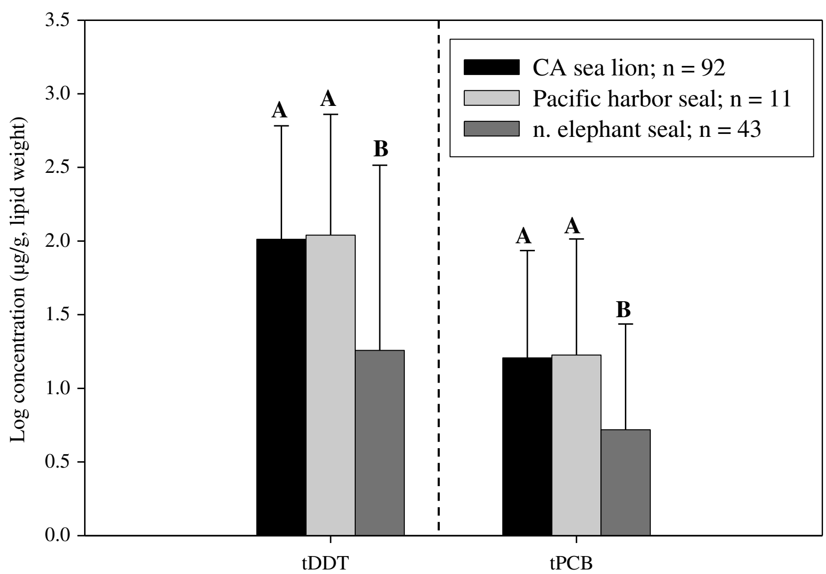

Fig. 3. Mean and standard deviation for tDDT and tPCB concentrations (μg/g, lipid weight) in the blubber of California sea lions, Pacific harbor seals, and northern elephant seals (all age classes and sexes combined). Species that have different letters above their bars are significantly different from each other.

than that of all non-adult individuals for tDDT and tPCB (Tukey's; $p < 0.05$; Fig. 4), with the greatest differences occurring between adult females and pups for both contaminants. Conversely, concentrations of these compounds in mature males were not significantly different from pups, yearlings or sub-adults ($p > 0.05$).

3.4. Temporal trends

tDDT and tPCB in CA sea lions examined from 1994 to 2006 significantly decreased over time (tDDT: $F = 12.01$; $df = 1, 100$; $p = 0.001$; tPCB: $F = 11.10$; $df = 1, 100$; $p = 0.001$; Fig. 5). However, this trend was not seen in northern elephant seals between 1997 and 2005 (tDDT: $F = 0.05$; $df = 1, 100$; $p = 0.82$; tPCB: $F = 1.38$; $df = 1, 100$; $p = 0.25$; Fig. 5). Temporal trends were not examined for Pacific harbor seals due to a low sample size among years for this species.

4. Discussion

4.1. Organochlorine concentrations

Mean concentrations of OCs in CA sea lions, Pacific harbor seals, and northern elephant seals studied exclusively in the SCB were found to be much higher than previously reported in CA sea lions examined by Le Boeuf et al. (2002) and Kannan et al. (2004) from southern California. Specifically, the mean concentrations of tDDT and tPCB for CA sea lions in this study ($n = 92$) were 9.8-fold (tDDT) and 4.8-fold (tPCB) higher than that found by Kannan et al. (2004) ($n = 7$) in CA sea lions in the southern range of their study. Although previous studies on CA sea lions showed higher overall levels of tDDT than tPCB, in our study levels of tDDT were almost 7 times higher than levels of tPCB in CA sea lions. Because the previous studies examined pinnipeds occurring along the entire coast of

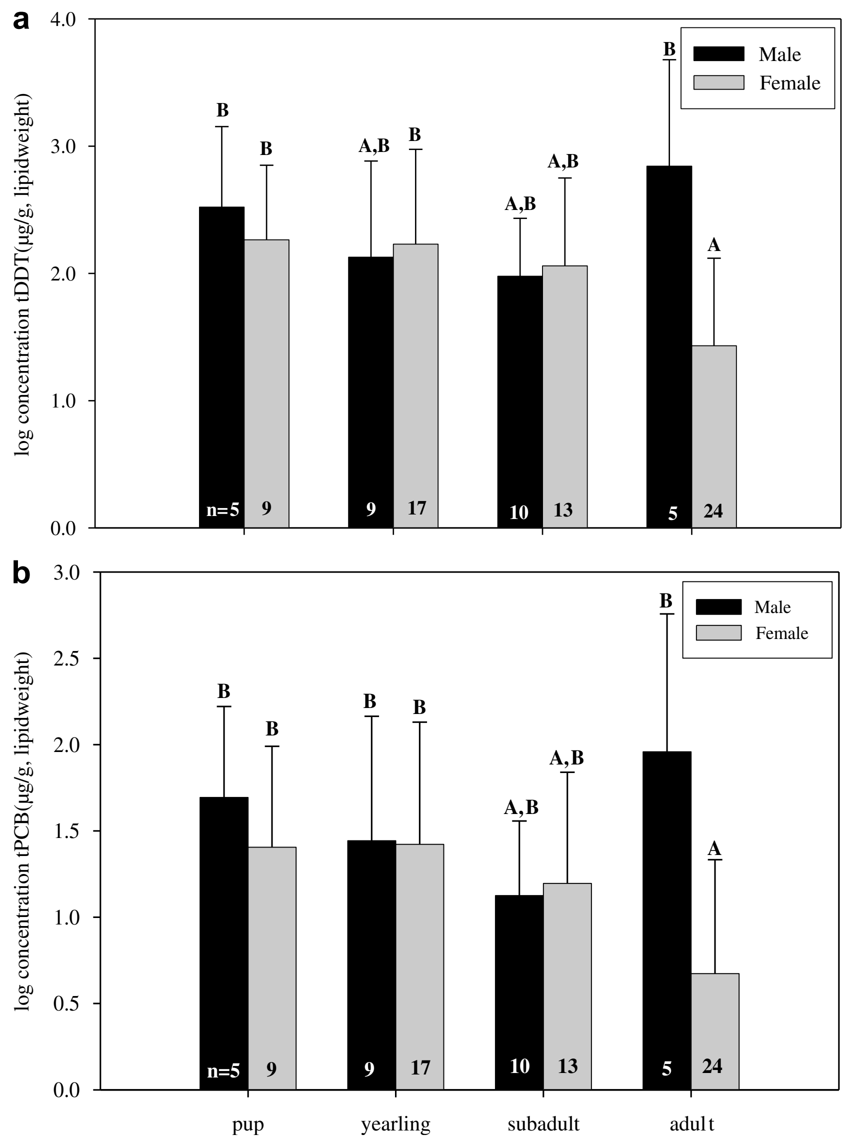

Fig. 4. Mean and standard deviation for (a) tDDT and (b) tPCB concentrations (µg/g, lipid weight) in California sea lion blubber by age and gender. Age and gender classes that have different letters above their bars are significantly different from each other. Numbers within bars are the sample size for a particular age/gender class.

California, the high levels of tDDT found in the present study reflect the extremely high levels that still remain in top-level carnivores in the SCB.

Although blubber contaminants are not directly comparable to liver contaminants, Kajiwara et al. (2001) reported higher concentrations of tPCB than tDDT in the livers of the harbor seals while we found higher levels of tDDT than tPCB in the blubber of all three species of pinnipeds sampled in the SCB. The greater concentrations of tPCB than tDDT found in harbor seals from northern California suggest that their exposure sources are different from those in harbor seals from the SCB.

Very few studies have reported the occurrence of CHL in marine mammals from the coastal waters of California (Kajiwara et al., 2001; Kannan et al., 2004). Concentrations of CHL in blubber of pinnipeds from the SCB region in this study were within the range of values found by Kajiwara et al. (2001) in the blubber of CA sea lions and northern elephant seals, and in the livers of Pacific harbor seals. Although we found one CA sea lion that had an extremely high CHL concentration (5770 µg/g, lw), this was probably due to its low lipid content (<1%). Overall, northern elephant seals had the highest composition of CHL compared to CA sea lions and Pacific harbor seals, which has been shown for organisms that feed away from the contaminant hotspot (Braune et al., 2005).

Concentrations of HCH in blubber of CA sea lions and seals from the SCB region in this study were also within the range of values reported in pinnipeds from California coastal waters (Kajiwara et al., 2001). The higher HCH composition found in the northern elephant seal blubber compared to the CA sea lion and Pacific harbor seal blubber is again a characteristic pattern in marine mammals exposed to contaminant sources located far from contaminant hotspots (Muir and Norstrom, 2000).

Because the lipid content in the blubber of pinnipeds varied widely, concentrations of OCs were normalized to lipid content to facilitate comparison among individuals. A negative correlation was observed between OC concentration and lipid content, where individuals with the highest concentrations of OCs had the lowest lipid content, which has also been reported in earlier studies (Fig. 5; Kajiwara et al., 2001; Kannan et al., 2004). Stranded animals with low lipid content have been known to concentrate contaminants in the remaining blubber, resulting in abnormally high levels of contaminants (Debier et al., 2006), therefore one should be cautious in making conclusions based on elevated normalized lipid

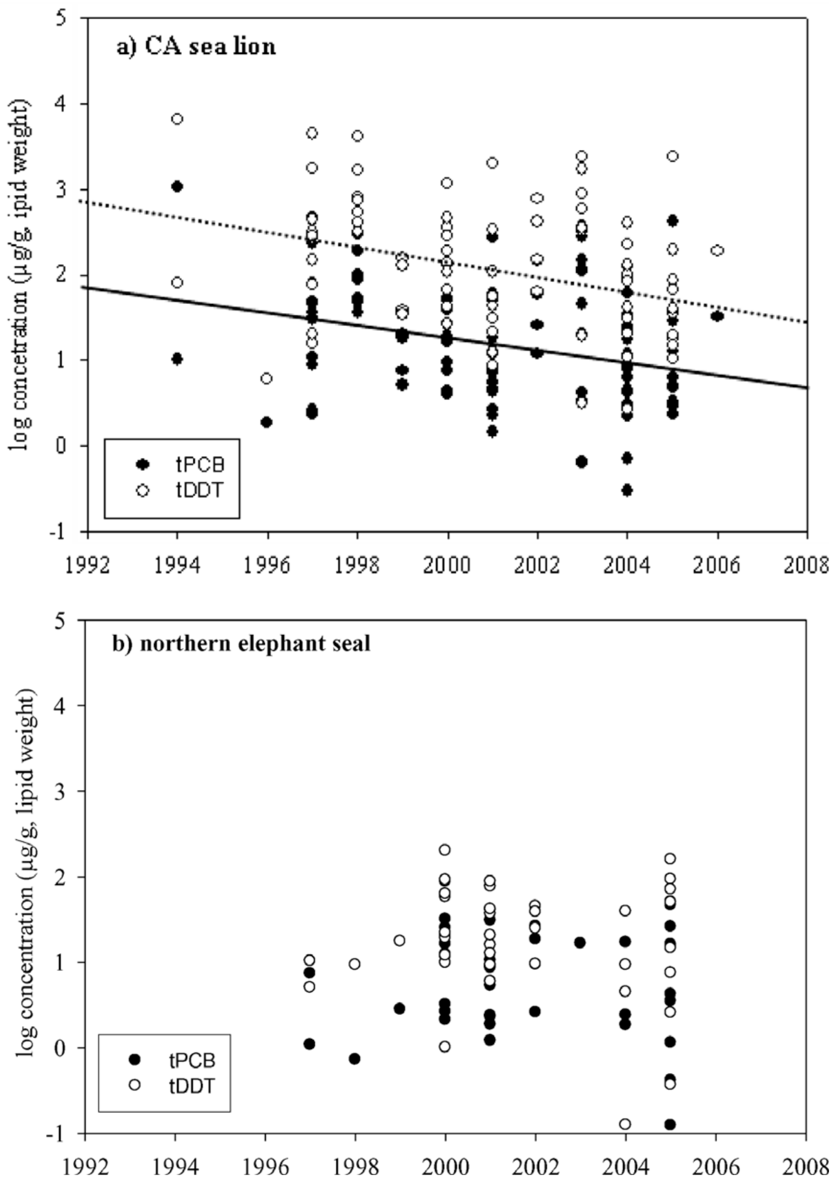

Fig. 5. Temporal changes in the mean tDDT and tPCB concentrations (µg/g, lipid weight) in the combined age and gender classes of (a) California sea lion, and (b) northern elephant seal blubber. Dashed line represents tDDT; solid line represents tPCB.

values and should consider wet weight values in addition to lipid weight values.

4.2. Species related effects

Considerable between-species variation occurred in the concentrations of tDDT and tPCB examined in CA sea lions, Pacific harbor seals, and northern elephant seals from the SCB. California sea lions and Pacific harbor seals exhibited significantly higher levels of tDDT and tPCB than northern elephant seals, but did not differ from each other (Fig. 3). Stable isotope ratio studies on prey conducted on CA sea lions, Pacific harbor seals, and northern elephant seals indicate that they feed at the same trophic level (Pauly et al., 1998; Burton and Koch, 1999); however, they do exploit different food resources, which could help explain differences in OC levels between the three species. While CA sea lions and Pacific harbor seals are both generalist feeders, taking advantage of locally available species, their preferred prey differs: CA sea lions mainly feed on northern anchovy (*Engraulis mordax*), Pacific sardine (*Sardinops sagax*), Pacific whiting (*Merluccius productus*), and market squid (*Loligo opalescens)* (Lowry et al., 1990), while Pacific harbor seals mainly feed on plainfin midshipman (*Porichthys notatus*), octopus, market squid, a variety of rockfish and flatfish species, and Pacific whiting (Antonelis and Fiscus, 1980). Northern elephant seals have the least varied diet, specializing mainly on cephalopods and a few fish species (Condit and Le Boeuf, 1984; Le Boeuf et al., 2000).

Since contaminants in local prey species will affect the degree of bioaccumulation of OCs in these pinnipeds, the significantly higher OC levels in the CA sea lion and Pacific harbor seal compared to the northern elephant seal may be related to their dietary differences. For example, in an assessment of tDDT and tPCB in pelagic forage fishes caught in the SCB, 99% of the northern anchovy, 83% of the Pacific sardine, and 33% of the Pacific chub mackerel exceeded wildlife risk screening values for tDDT (14 µg/kg), but none exceeded the screening value for PCB_{TEQ} (0.79 µg/kg; Jarvis et al., 2007). With pelagic forage fish exceeding wildlife screening values for tDDT, there is great potential for these fish from the SCB to serve as sources of substantial contaminant loads in CA sea lions and Pacific harbor seals.

In addition to dietary influences, differences in concentrations of tDDT and tPCB are also influenced by the habitat range of the pinnipeds. For example, differences in concentrations of tDDT and tPCB between Pacific harbor seals and northern elephant seals may be explained by the harbor seal's continual residency in the SCB compared to the elephant seal's dual habitats (the SCB for short periods of breeding and molting, and offshore and Alaska during all other times of the year; Condit and Le Boeuf, 1984; Reeves et al., 2002).

In the present study, tDDT concentrations in the blubber of non-adult northern elephant seals were higher than those reported previously for this species (Beckman et al., 1997; Kajiwara et al., 2001; Debier et al., 2005; Debier et al., 2006). Because the levels of tDDT are higher than tPCB, this supports the theory that pups and yearlings of this species are feeding more often in the SCB than previously thought (Kajiwara et al., 2001). However, comparisons of contaminant levels between adult and non-adult northern elephant seals are necessary to better understand this occurrence. Overall, it should be stressed that OC concentrations in southern California pinnipeds are in the highest range of those detected in all other marine mammals found worldwide (Table 3).

4.3. Age and gender trends: CA sea lion

In CA sea lions, the pattern of contaminant concentrations increasing with age in males and decreasing in females is consistent with patterns observed in other marine mammals (Ross et al., 2000). CA sea lion pups had the highest OC concentrations of all age classes with pups having higher tDDT and tPCB concentrations than the adult female age/gender class (Fig. 4). The high concentrations of OCs of pups reflect the absorption of these lipophilic contaminants via maternal lipid stores during gestation and lactation (Wolkers et al., 2004; Shaw et al., 2005).

OC loads initially decreased with age during the juvenile stages for both sexes (Fig. 4), which may be due to dilution of OCs by rapid growth during the first few years, excretion by metabolic degradation exceeding ingestion of OCs, and the diet switch from a lipid-rich and highly contaminated milk diet to a relatively less contaminated fish diet (Shaw et al., 2005). In adult males, OC loads did not change with age (Fig. 4). This implies that the amount of contaminants ingested is equal to that excreted or degraded, resulting in a steady-state concentration over the life-span of the animal. Conversely, the adult female's ability to transfer contaminants to their offspring during gestation and lactation (Greig et al., 2007) may result in a significant decline in OC concentrations below the 'steady-state' achieved by adult males (Fig. 4). Although this pattern of higher OC levels in adult males vs. adult females has been found in other pinniped species (Lee et al., 1996), and hypothesized to occur in the CA sea lion (Le Boeuf et al., 2002; Kannan et al., 2004), this is the first study where it has been definitively shown to occur in CA sea lions.

4.4. Temporal trends

There was a significant decline in both tDDT and tPCB over the duration of the sampling period in CA sea lions, but not in northern elephant seals (Fig. 5). The decrease in tDDT and tPCB concentrations is symptomatic of exposure to point sources in highly contaminated areas generated by historical dumping in the SCB and other regions (Addison et al., 1984; Riget et al., 2006). This rapid decrease in tissue contamination seen in the CA sea lions occurs in regions of high initial pollution and can be attributed to the cessation of dumping along with the dispersal and dilution of contaminants from the point source by physical processes. This then leads to a subsequent increase in contaminants to a steady-state condition in distant pristine regions (Aguilar et al., 2002).

Although levels of tDDT and tPCB in the CA sea lions have significantly decreased over time, several individuals in this study contained contaminant levels similar to adult female sea lions examined in the 1970s that gave birth to premature pups (DeLong et al., 1973). In addition, concentrations of tPCB above 17 mg/kg, lipid weight have been linked to a suppressed immune system (Ross et al., 1996) and many animals in this study (48% of the CA sea lions, 36% of the harbor seals, and 23% of the northern elephant

Table 3
Comparison of mean and ranges of tDDT and tPCB blubber concentrations (μg/g, lipid weight) in pinnipeds

Species Sampling location; year	*n*	tDDT	tPCB	References
Steller sea lion Alaska; 1976–1978	29	5 (0.19–77)	11.15 (0.57–41)	Lee et al. (1996)
California sea lion California; 1991–1997	12	485.25 (13–2900)	363.85 (7.2–1300)	Kajiwara et al. (2001)
California sea lion California; 2000	36	143 (4.1–1400)	44.1 (NR-410)	Le Boeuf et al. (2002) Kannan et al. (2004)
California sea lion SCB; 1994–2006	92	594.4 (2.6–13,271)	86.6 (0.29–2208)	Present study
Harbor, grey, harp, hooded seals St. Lawrence Gulf; 1995–2000	37	6.2 NR[a]	13.7 NR	Hobbs et al. (2002)
Caspian seal Caspian Sea; 2000	13	91.7 (6.3–470)	46.5 (2.4–320)	Kajiwara et al. (2002)
Harbor seal Northwestern Atlantic; 2001–02	27	13.1 (1.4–57.5)	34.4 (5.7–151)	Shaw et al. (2005)
Pacific harbor seal SCB; 1995–2003	11	1042.2 (9.9–10,031)	123.2 (1.2–1125)	Present study
Northern elephant seal California; 1992	24	5.4 (1.9–11.8)	2.0 (0.72–4.3)	Beckman et al. (1997)
Northern elephant seal California; 1991–1997	4	35.58 (8.3–110)	19.23 (6.1–58)	Kajiwara et al. (2001)
Northern elephant seal SCB; 1997–2005	42	46.82 (0.0–226.3)	14.02 (0.06–76.0)	Present study

[a] NR = Ranges not reported.

seals) contained tPCB levels above this threshold. The tDDT and tPCB concentrations found in these pinnipeds are of potential concern because several blubber samples had concentrations within the range that have been shown to negatively affect many physiological and metabolic functions in other marine mammals (de Swart et al., 1996). Furthermore, the tPCB concentrations from CA sea lions seen at the levels in this study are consistent with mean tPCB concentrations that have been associated with carcinomas and premature death in this species (Ylitalo et al., 2005).

5. Conclusions

Although the CA sea lion population in southern California is healthy and growing, massive epizootics have occurred in pinniped populations in other regions of the world (Kennedy et al., 2000). Because more than 50% of the CA sea lions sampled had levels of contaminants above the threshold known to cause a depressed immune system, and assuming our sampling represents that of the SCB population, an epizootic episode in CA could have serious impacts on the local population. In addition, high levels of contaminants were also found in harbor seals, and to a lesser extent, northern elephant seals, whose smaller populations cannot suffer the consequences of a large-scale epizootic episode as well as can the CA sea lion population. The highly elevated levels of tDDT and tPCB warrant further examination into the relationship of OC exposure to fitness of both healthy and unhealthy individual pinnipeds.

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