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PCB and DDE Contamination in Harbor Seals (*Phoca vitulina*) from North-Central California and Bristol Bay, Alaska

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Abstract

In recent years, concerns have increased regarding accumulation of persistent, lipophilic contaminants by marine mammals. We quantified blood levels of the two most prevalent organochlorine (OC) contaminants of the marine ecosystem in a model species, the harbor seal (*Phoca vitulina*) from three north-central California populations and a population in Bristol Bay, Alaska. Intensive sampling (*n* = 190) produced robust quantification of blood concentrations of selected PCBs and DDE, and allowed us to investigate factors affecting levels of these contaminants in seal populations with distinct environments and exposure histories. In the Alaskan samples, PCB and DDE levels were most strongly related to sex and age; OCs increased with age in males and decreased with age in females, likely due to cumulative exposure in males and load-dumping during lactation in females. Among females, an inverse relationship was observed between condition and PCB blood levels. In contrast, in the California seals, in which loads were generally much greater, pups had greater levels of PCBs and DDE than subadults and adults, suggesting stable to decreasing environmental contaminant levels. Spatial heterogeneity and seasonal differences also contributed substantially to variation among harbor seals in contaminant loads. These findings underscore the importance of accounting for demographic, geographic, seasonal, and physiological effects in toxicological studies of marine mammals.

Key Words: harbor seal, *Phoca vitulina*, PCB, DDE, organochlorine, marine contamination

Introduction

Organochlorine (OC) pollutants in the marine environment pose a potential threat to marine organisms, and especially to marine mammals (Tanabe et al., 1994). Organochlorines are typically persistent and lipophilic, thus accumulating in the adipose tissues of animals (Brooks, 1974; Hutzinger et al., 1974; Tanabe et al., 1988). The greatest accumulations are found in long-lived species that maintain large fat stores and occupy high trophic levels, including many marine mammals (Tanabe et al., 1984; Loganathan & Kannan, 1994).

Polychlorinated biphenyls (PCBs) and p,p'-DDE, the major metabolite of the insecticide DDT (dichloro-diphenyl-trichloroethane), are environmentally ubiquitous and the most prevalent OCs identified in marine mammals (Calambokidis & Francis, 1994; Jarman et al., 1996; Nakata et al., 1998; Skaare et al., 2000; Kajiwara et al., 2001). Although use of DDT is banned in most countries, it is still used in some areas of the world (e.g., to combat malaria). Production of PCBs was banned in the United States and has decreased worldwide, but entry of substantial amounts into the marine environment likely continues.

In recent years, the relatively high levels of OCs found in tissues of marine mammals have raised concerns that exposure to these marine contaminants may compromise individual health (e.g., via immune suppression) and even threaten persistence of certain populations (e.g., via effects on reproduction and/or survival) (Reijnders, 1994; De Guise et al., 1995; Ross et al., 1996; Beckmen et al., 2003). Organochlorines, such as PCBs and

DDT, have been associated with various adverse conditions in marine mammals. These include impaired immunological function and reproductive success in the harbor seal (*Phoca vitulina*) (Reijnders, 1980, 1986; Ross et al., 1996), decreased circulating vitamin A and thyroid hormone levels in juvenile harbor and grey (*Halichoerus grypus*) seals (Brouwer et al., 1989; Jenssen et al., 2003), depressed humoral immune responses in northern fur seal (*Callorhinus ursinus*) pups (Beckmen et al., 2003), and decreased lymphocyte responses in bottlenose dolphin (*Tursiops truncatus*) (Lahvis et al., 1995).

The harbor seal is a useful model species for studies of contamination of the marine food web and potential environmental health effects. Harbor seals tend to feed on species of fish that are coastal and, in contrast to many other phocids, they tend to migrate locally, staying close to their coastal feeding and haul-out areas, and using bays and estuaries for resting, foraging, and reproduction (Brown & Mate, 1983; Kopec & Harvey, 1995; Grigg et al., 2002). Thus, residues of OCs in tissues of harbor seals tend to reflect contamination of the local environment. In addition, because such contaminants are concentrated in harbor seals relative to abiotic matrices, as well as other resident biota occupying lower trophic levels, harbor seals can serve as sentinels of environmental contamination of bay and estuarine ecosystems (Tanabe, 1988; Young et al., 1998).

Factors influencing bioaccumulation of persistent environmental contaminants in marine mammals have been largely understood based on theoretical models and/or very limited samples from stranded dead animals. In the present study, we determined concentrations of selected PCB congeners and p,p'-DDE in whole blood of harbor seals and explored the importance of demographic and other factors on PCB and DDE loads. We sampled harbor seals from north-central California (CA) and Bristol Bay, Alaska (AK). Sampling was much more intensive than in previous toxicological surveys to provide robust estimates of current contaminant levels for these harbor seal populations and to afford sufficient statistical power to examine relationships between contaminant loads and sex, age class, capture site, body condition, and season. Due to the relative lack of industrial, municipal, and agricultural development surrounding Bristol Bay, the AK harbor seals served as a comparison population for which we expected OC contaminant levels to be relatively low. For both populations, we expected to find higher concentrations of PCBs and DDE in adult males compared to adult females due to the offloading of lipophilic contaminants in milk from mother to pup (Addison & Brodie, 1987) and predicted higher concentrations of these contaminants in seals in poorer condition—reflecting use of fat reserves and the related mobilization of fat-sequestered PCBs and DDE into the bloodstream—generally corresponding to the summer season.

Materials and Methods

Sampling

Sampling of harbor seals in CA was conducted year-round from July 2000 through August 2002 in the vicinity of primary haulouts in Monterey Bay (MB), San Francisco Bay-Estuary (SFB), and Point Reyes Headland (PRH). Capture included land-grab, beach seine, and tangle-net techniques aimed to sample as randomly as possible (with respect to sex and age) (Harvey, 1987; Yochem et al., 1987; Jeffries et al., 1993). Seals that appeared to be clinically unhealthy at time of capture were excluded from the sample. Seals were physically restrained, and sex, mass, and standard length (SL) were recorded. Blood was drawn from the extradural vein into sterile evacuated blood collection tubes containing EDTA (Becton Dickinson Vacutainer Systems, Franklin Lakes, NJ, USA). Samples were transferred to glass vials with TFE-lined screw caps and stored frozen. The sampling of harbor seals in Bristol Bay, AK, was conducted during September 2000 and 2001 at the haulouts of Kvichak, Egegik, and Ugashik subbays. Samples were collected as above and transported frozen to the laboratory at the University of California–Davis for chemical analyses. The spatial scale of sampling in both CA and AK was roughly equivalent (approx. 200 km straight-line distance encompassed all sites for each population) so as not to confound any differences in trends between the two populations (heterogeneity generally increases with distance).

Peripheral blood from clinically healthy, free-ranging seals was sampled. Although circulating blood is the matrix most relevant to cellular toxicity of OCs, most studies of contaminants in marine mammals are based on blubber samples (and, to a lesser extent, liver and other organs), often collected from dead and decaying carcasses with little regard for the disease state of the animal. Although the use of blood as a substrate for contaminant analyses has been criticized for species that fast for a substantial period of time (Lydersen et al., 2002), blood may be an excellent substrate in species that do not fast, such as the harbor seal, or in fasting species if this is explicitly accounted for in the sampling design and data interpretation. Further, whereas blubber concentrations of OCs vary greatly depending on where in the blubber layer (inner vs outer) the sample is taken (Severinsen et al., 2000), blood is relatively homogeneous. Blood also is the tissue that is usually easiest to obtain, and many other types of analyses in veterinary medicine are based on blood samples.

Laboratory Analysis

Specific PCB congeners were selected for analysis based on toxicity information and reported abundances in the environment and in biota (Van den Berg et al., 1998, 2006; Valoppi et al., 2000). Targeted analytes included CBs (IUPAC#) 128, 153, 156, 167, 169, 170, 180, 189, 195, 206, and 209, and p,p′-DDE. PCB and DDE standards were purchased from AccuStandard, Inc. (New Haven, CT, USA). All solvents (highest analytical grade) were purchased from Fisher Scientific (Pittsburgh, PA, USA). Methods for analytical chemistry and quality assurance were presented in detail elsewhere (Neale et al., 2005b). Briefly, extraction (acetonitrile/hexane) and clean-up (Florisil) of organohalogen residues from 5-ml whole blood was followed by quantitative analysis via GC-ECD (injection 100° C, 1.5 min; 15° C/min to 165° C; 20° C/min to 285° C; run time, 114 min). Chlorinated biphenyls #14, 65, and 166 were used as method surrogates and CBs 30 and 204 as internal standards. Multi-level internal standard calibrations, using a minimum of five standard concentrations, were used as the basis for quantification. Due to co-elution with the pesticide Mirex, CB 169 was excluded from quantitative analysis. Blood lipids were measured using the colormetric technique of Frings et al. (1972) as described in Neale et al. (2005b).

Data Analysis

Age of seals was estimated based on SL as follows: class 1 (pup: M and F < 100 cm); class 2 (yearling/subadult: M 101-134 cm, F 101-129 cm); and class 3 (adult: M > 135 cm, F > 130 cm) (Neale et al., 2005b). A condition index, introduced by Neale et al. (2005b) and based on the relationship between body mass and the cube of SL, was calculated for each seal as follows: a regression of the $\sqrt[3]{mass}$ on SL was calculated using all seals (there was no difference between sexes or among populations in this relationship) and using the overall regression line (expected $\sqrt[3]{mass}$ = 0.0248*SL + 0.547) as the expected value (i.e., of a seal in average body condition). The condition index was defined as the observed $\sqrt[3]{mass}$ minus expected $\sqrt[3]{mass}$, which controlled for age-related allometry between mass and length, such that positive values represented above-average condition and negative values represented below-average condition.

Concentrations (ppb) of analytes determined in whole blood were expressed on wet-weight (ww; ng/g whole blood) and lipid-weight (lw; ng/g blood lipid) bases. Spearman Rank Correlation was used to assess the similarity of PCB congener profiles between the two populations. For each sample, the measured concentrations of the 10 specific PCB congeners were totaled to obtain ∑PCB values. Before statistical procedures, contaminant concentrations were log-transformed [log(x + 1)] to better approximate normality. An analysis of covariance utilizing the General Linear Model (GLM) procedure in *SYSTAT*, Version 9 (SPSS, Chicago, IL, USA) was used for statistical testing of relationships. Analyses of contaminant concentrations for each population included capture site, sex, age class, and the interaction of sex and age class (sex*age) as factors, and the condition index as covariate, on $\sum PCB_{ww}$, $\sum PCB_{lw}$, DDE_{ww}, and DDE_{lw}. Capture season (winter = December-February; spring = March-May; summer = June-August; fall = September-November) also was included as a variable in the CA analyses (AK samples were all collected during September). All statistics reported are from the full, conservative models (i.e., no variables were dropped; direction and strength of relationships were determined while controlling for all other variables). Due to the small sample from PRH (*n* = 5), these data were not included in the statistical analyses.

Results

One hundred-seventy eight harbor seals were analyzed for contaminant levels in whole blood, including 99 individuals (49 M, 50 F) from three sites in CA and 79 individuals (29 M, 50 F) from three sites in AK. In CA, four individuals were recaptured (one was recaptured twice) for a total of 104 samples.

As expected, levels of PCBs and especially DDE were greater in CA samples than in AK samples (Tables 1 & 2). Specifically, the mean $\sum PCB_{ww}$ for CA harbor seals was 10.84 +/- 1.64 (SE) ppb vs 0.74 +/- 0.07 ppb in AK samples. Mean DDE_{ww} was 33.28 +/- 5.27 ppb in CA and 0.67 +/- 0.09 ppb in AK. Lipid-adjusted values indicated similar relationships: average $\sum PCB_{lw}$ for CA harbor seals was 3,222.16 +/- 462.96 ppb vs 222.33 +/- 18.68 ppb in AK samples, and DDE_{lw} averaged 9,477.18 +/- 1,401.09 ppb in CA and 195.78 +/- 24.74 ppb for AK samples.

Lipid-normalized values for ∑PCB and DDE (ranging in the low ppm) were orders of magnitude greater than ww values (Tables 1 & 2). In general, however, lipid normalization of contaminant concentrations did not affect results qualitatively, and a systematic relationship was observed between ww and lipid-adjusted concentrations for ∑PCB and DDE. Because analyses based on lipid-adjusted concentrations produced qualitatively similar

Table 1. Concentrations (mean + SE) of ΣPCB and p,p'-DDE determined in 104 harbor seal samples from San Francisco Bay (SFB), Monterey Bay (MB), and Point Reyes Headlands (PRH), California; sample sizes in parentheses; age class 1 = pup, 2 = subadult, 3 = adult.

	ppb wet weight			ppm blood lipid		
	SFB	MB	PRH	SFB	MB	PRH
ΣPCB						
Male 1	25.5 + 11.5 (3)	5.6 + 1.3 (11)	6.2 (1)	7.5 + 3.5 (3)	1.4 + 0.3 (11)	2.3 (1)
Male 2	13.3 + 4.6 (7)	7.3 + 4.0 (20)	8.7 + 3.9 (2)	4.0 + 1.5 (7)	2.2 + 1.1 (19)	1.9 (1)
Male 3	27.1 + 7.3 (6)	5.3 + 1.2 (4)	--	7.1 + 2.1 (6)	1.6 + 0.4 (4)	--
Female 1	10.8 + 1.9 (3)	21.5 + 16.0 (6)	4.8 (1)	3.4 + 0.7 (3)	5.8 + 4.6 (5)	1.5 (1)
Female 2	16.6 + 5.5 (17)	2.7 + 1.0 (15)	4.7 (1)	5.0 + 1.7 (17)	1.2 + 0.4 (11)	1.3 (1)
Female 3	8.9 + 2.5 (5)	3.4 + 1.3 (2)	--	1.9 + 0.4 (5)	1.0 + 0.0 (2)	--
DDE						
Male 1	44.2 + 18.1 (3)	71.1 + 29.0 (11)	18.9 ($n = 1$)	12.1 + 5.0 (3)	19.5 + 8.7 (11)	7.1 (1)
Male 2	19.2 + 5.5 (7)	18.0 + 3.4 (20)	57.6 + 39.7 (2)	5.6 + 1.8 (7)	5.7 + 0.9 (19)	9.2 (1)
Male 3	24.7 + 4.6 (6)	39.7 + 1.9 (4)	--	6.5 + 1.6 (6)	12.0 + 0.7 (4)	--
Female 1	20.8 + 6.7 (3)	114.5 + 64.3 (6)	17.3 (1)	6.6 + 2.2 (3)	27.9 + 18.5 (5)	5.2 (1)
Female 2	21.7 + 6.6 (17)	21.5 + 8.4 (15)	6.7 (1)	6.3 + 1.8 (17)	10.1 + 3.3 (11)	1.9 (1)
Female 3	14.0 + 4.6 (5)	38.9 + 37.7 (2)	--	3.0 + 0.6 (5)	9.7 + 6.8 (2)	--

Table 2. Concentrations (mean + SE) of ΣPCB and p,p'-DDE determined in 79 harbor seals from three sub-bays of Bristol Bay, Alaska; sample sizes in parentheses; age class 1 = pup, 2 = subadult, 3 = adult.

	ppb wet weight			ppm blood lipid		
	Egegik	Kvichak	Ugashik	Egegik	Kvichak	Ugashik
ΣPCB						
Male 1	0.5 + 0.3 (2)	--	0.7 + 0.2 (5)	0.2 (1)	--	0.2 + 0.0 (5)
Male 2	0.6 + 0.1 (5)	--	0.7 + 0.1 (3)	0.2 + 0.0 (5)	--	0.2 + 0.0 (3)
Male 3	1.0 + 0.3 (6)	--	1.6 + 0.3 (8)	0.3 + 0.1 (6)	--	0.5 + 0.1 (8)
Female 1	1.6 + 0.4 (2)	0.3 (1)	0.7 + 0.1 (6)	0.4 + 0.1 (2)	0.1 (1)	0.2 + 0.0 (6)
Female 2	0.8 + 0.1 (11)	0.6 + 0.2 (3)	0.8 (1)	0.3 + 0.0 (11)	0.2 + 0.0 (3)	0.2 (1)
Female 3	0.4 + 0.1 (18)	--	0.4 + 0.1 (8)	0.1 + 0.0 (18)	--	0.2 + 0.1 (8)
DDE						
Male 1	0.5 + 0.4 (2)	--	0.2 + 0.1 (5)	0.2 (1)	--	0.1 + 0.0 (5)
Male 2	0.6 + 0.27 (5)	--	1.5 + 0.9 (3)	0.2 + 0.1 5)	--	0.4 + 0.2 (3)
Male 3	1.2 + 0.3 (6)	--	1.3 + 0.5 (8)	0.3 + 0.1 (6)	--	0.4 + 0.1 (8)
Female 1	2.5 + 0.7 (2)	0.5 (1)	0.7 + 0.3 (6)	0.7 + 0.1 (2)	0.2 (1)	0.2 + 0.1 (6)
Female 2	0.8 + 0.2 (11)	1.0 + 0.5 (3)	0.0 (1)	0.3 + 0.0 (11)	0.3 + 0.2 (3)	0.0 (1)
Female 3	0.2 + 0.1 (18)	--	0.1 + 0.0 (8)	0.1 + 0.0 (18)	--	0.0 + 0.0 (8)

results as those based on ww concentrations, only the latter were used in GLM analyses.

Profiles of the 10 PCB congeners were similar for the two harbor seal populations (Spearman rank correlation = 0.72_{ww}, 0.54_{lw}). For both populations, CB 153 and CB 180 contributed most to ∑PCB. In CA samples, CB 153 and CB 180 together constituted an average 79% of $\sum PCB_{ww}$, with CB 153 contributing 54% of total PCBs. In AK, these two congeners together were 96% of $\sum PCB_{ww}$, with CB 153 alone contributing 90%.

Correlates of Contaminant Levels—Alaskan Harbor Seals

In AK, contaminant concentrations in harbor seals tended to increase with age in males but decreased with age in females; statistically, this resulted in highly significant interaction (sex*age) terms in both contaminant models ($\sum PCB_{ww}$ $F_{2,69} = 13.94$, $p < 0.001$; DDE_{ww} $F_{2,69} = 12.32$; $p < 0.001$). Therefore, these models were decomposed and performed for males and females separately, with condition index, site, and age class as factors on $\sum PBC_{ww}$ and DDE_{ww}.

For males ($n = 29$), $\sum PCB_{ww}$ increased with age ($F_{2,24} = 4.75$, $p = 0.018$), with levels in adult males about twice that of pups and subadults (Figure 1) The same general trend was observed for DDE_{ww}, but this effect was marginally nonsignificant ($F_{2,24} = 2.91$, $p = 0.074$). Condition index and capture site (males were captured only at Ugashik and Egegik) were not significant factors of contaminant concentrations ($p > 0.21$).

For females ($n = 49$), contaminant concentrations decreased with age (e.g., $\sum PCB_{ww}$; see Figure 2) in sharp contrast to the pattern for males, and this effect was highly significant in both models ($\sum PCB_{ww}$ $F_{2,43} = 8.66$, $p = 0.001$; DDE_{ww} $F_{2,43} = 14.95$, $p < 0.001$). Condition index was also a significant factor of $\sum PCB_{ww}$ for females ($F_{1,43} = 9.38$, $p = 0.004$), and it decreased with increasing loads ($R^2 = 0.14$; see Figure 3). Likewise, an inverse (but nonsignificant) relationship was observed between condition index and DDE_{ww} ($R^2 = 0.03$). Lastly, capture site was a significant factor of DDE_{ww} ($F_{2,43} = 3.25$, $p = 0.048$), with levels greatest in females from Egegik and least in those from Ugashik.

Correlates of Contaminant Levels—California Harbor Seals

In contrast to the AK reference population, contaminant concentrations of seals in CA did not relate significantly to condition index ($p > 0.27$), sex ($p > 0.28$), or the interaction of sex and age ($p = 0.469$ for DDE_{ww}, $p = 0.093$ for $\sum PCB_{ww}$). Rather, site, age class, and season were the primary factors of PCB and DDE levels in CA harbor seals.

Capture site was a significant determinant for $\sum PCB_{ww}$ ($F_{1,88} = 12.98$, $p = 0.001$) and DDE_{ww} ($F_{1,88} = 4.06$, $p = 0.047$). Levels of PCBs were greater in seals from SFB ($n = 41$) than from MB ($n = 58$); however, DDE had the reverse trend (Figure 4). Harbor seals from PRH, an outer coastal location directly northwest of SFB, had lesser overall PCB levels than seals caught in SFB, with mean $\sum PCB_{ww}$ (6.61 +/- 1.39 SE ppb, $n = 5$) approximately the same as that for MB. DDE_{ww} levels for PRH (x = 31.61 +/- 15.63) were intermediate between SFB and MB.

Contaminant loads in CA harbor seals also varied by age class ($\sum PCB_{ww}$ $F_{2,88} = 3.39$, $p = 0.038$; DDE_{ww} $F_{2,88} = 8.86$, $p < 0.001$). The greatest concentrations of both PCBs and DDE were found in pups (age class 1 > 2 ~ 3; e.g., DDE; see Figure 5).

Lastly, season was an important factor of contaminant levels in CA harbor seals. To assess seasonal differences, separate analyses of covariance for SFB and MB were performed since SFB seals were not sampled during all four seasons. In MB, season was a significant factor for $\sum PCB_{ww}$ ($F_{3,48} = 5.37$, $p = 0.003$); levels were greatest during summer and least during winter and fall (Figure 6). Although not significant, a similar seasonal trend was indicated for DDE_{ww}. In SFB, captures occurred during summer and winter only. Similar to the MB result, OC levels were greater in summer than winter ($\sum PCB_{ww}$ $F_{1,33} = 4.61$, $p = 0.039$; DDE_{ww} $F_{1,33} = 4.16$, $p = 0.050$; see Figure 7).

Recaptures of individual harbor seals provided an opportunity to quantify seasonal changes in contaminant concentrations within individuals.

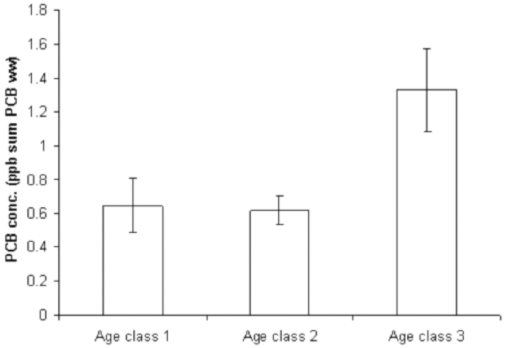

Figure 1. ΣPCB (mean + SE) in AK male harbor seals by age class

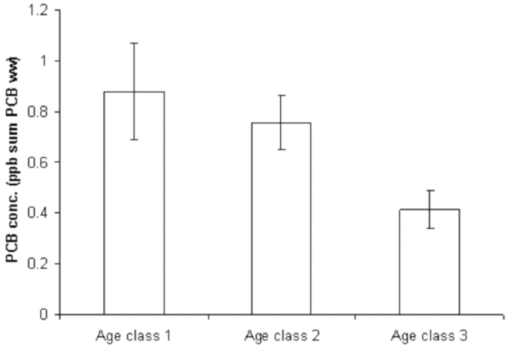

Figure 2. ΣPCB (mean + SE) in AK female harbor seals by age class

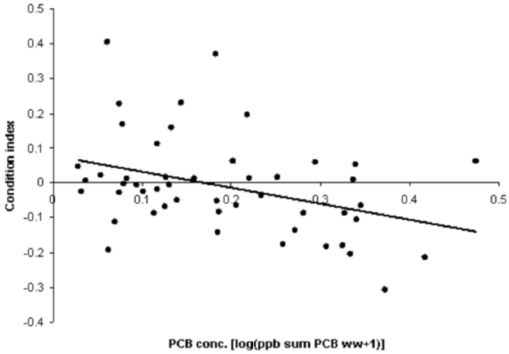

Figure 3. Relationship between condition index and ΣPCB in AK female harbor seals ($n = 49$)

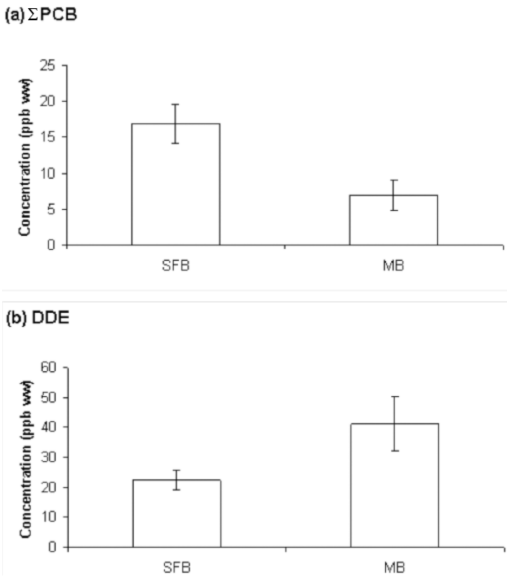

Figure 4. Organochlorine levels (mean + SE) in harbor seals from San Francisco Bay and Monterey Bay, CA, for (a) ΣPCB and (b) DDE

Three individuals from SFB were each captured once during winter and once during summer. For all three SFB seals, loads were greater in summer than in winter. The percentage decrease in DDE_{ww} from summer to winter ranged from 16 to 74%, which represented decreases in concentration of up to 34 ppb (this male, captured as a pup during summer, also had the greatest loads of the three recaptured SFB seals). Changes were more consistent among individuals for $\sum\text{PCB}_{\text{ww}}$, with the summer-to-winter loads decreased by 48 to 56% (absolute ppb changes of 5 to 12). A single individual from MB was captured three times (fall, winter, and spring). This seal had lesser PCB and DDE loads than the SFB recaptures and also had more modest seasonal changes (fall > spring > winter), with 10 to 20% reductions among seasons (changes of ~0.1 ppb).

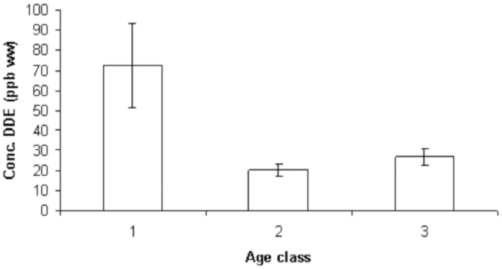

Figure 5. DDE (mean + SE) in CA harbor seals by age class

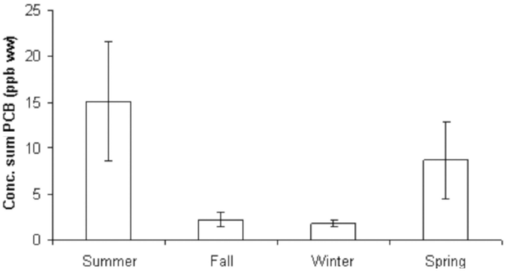

Figure 6. ΣPCB (mean + SE) in harbor seals from Monterey Bay, CA, by season

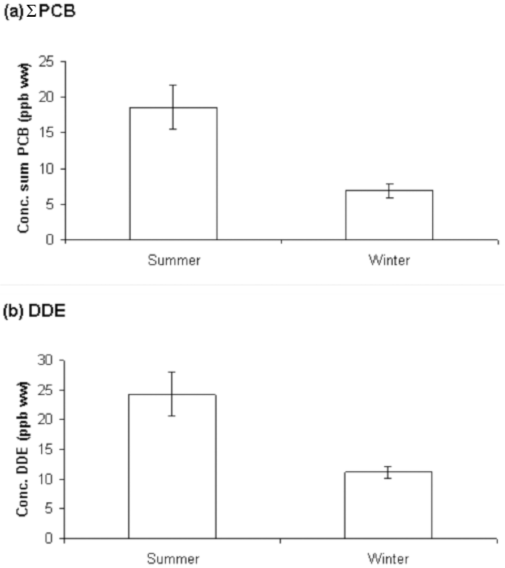

Figure 7. Summer vs winter organochlorine levels (mean + SE) in harbor seals from San Francisco Bay, CA, for (a) ΣPCB and (b) DDE

Discussion

Alaska Seals

Average ∑PCB and DDE levels for Bristol Bay harbor seals were considerably lower than the corresponding ww or lipid-based blood concentrations reported for marine mammals elsewhere (i.e., studies measuring blood concentrations in > five individuals) (Addison & Brodie, 1987; Lahvis et al., 1995; Lydersen et al., 2002; Beckmen et al., 2003; Jenssen et al., 2003). Nevertheless, given that there are no known local sources of PCB and DDE contamination to Bristol Bay, it is interesting to note that detectable levels of most analytes were observed for most samples. This finding is consistent with the hypothesis that OC concentrations in the environment and biota are likely to be found in noteworthy concentrations at high latitudes, remote from tropical and temperate latitudes of contaminant origin, due to the processes of global fractionation and distillation associated with long-range air transport (Oehme, 1991; Wania & Mackay, 1993; Tanabe et al., 1994).

Trends in PCB and DDE loads for harbor seals in AK indicated that males accumulate contaminants with age, whereas adult females unload part of their contaminant burden, presumably to their offspring during pregnancy and especially during lactation. These results were consistent with previous findings. Past studies have documented increases with age in organochlorine concentrations in the blubber of male pinnipeds (e.g., Born et al., 1981; Donkin et al., 1981; Helle et al., 1983). Female pinnipeds (and other marine mammals) tend to have increasing organochlorine concentrations in blubber with age until they mature and reproduce; as OCs are lost through lactation, concentrations decrease or reach a plateau (Tanabe et al., 1982; Ronald et al., 1984; Addison & Brodie, 1987; Bacon et al., 1992).

In AK, the condition index of harbor seal females was inversely related to PCB load. All of these seals were caught at the end of summer—an energetic low point for harbor seals, especially females that have undergone pupping and lactation, followed by breeding and the annual molt (Boulva & Mclaren, 1979; Renouf et al., 1988; Fedoseev, 2000). Increased fat metabolism of females in decreasing condition would cause PCBs (and other lipophilic chemicals) stored in fat to be released into the bloodstream. Thus, greater blood levels of PCBs might be expected in seals with lower condition indexes, having depleted more of their fat reserves (Lydersen et al., 2002). This inverse relationship between organochlorine concentration and lipid reserves in mammals has been documented previously (Britt & Howard, 1983) as has seasonal variation in blubber thickness, reflecting the lipid storage pool (Pitcher, 1986).

Harbor seals sampled in Bristol Bay had far greater contaminant loads than seven spotted seals (*Phoca largha*) (two M, five F) captured in the same area during September (Neale et al., 2007). This difference is most likely explained by species differences in foraging area and trophic level of prey. Spotted seals tend to prey on small, schooling fishes, often taking juveniles, whereas the harbor seal diet includes larger fishes. Thus, harbor seals tend to feed on a slightly higher trophic level than spotted seals (Dehn et al., 2005; K. Frost, pers. comm, 2005[1]). Spotted seals are strongly associated with ice, and they make an annual migration in the fall and winter to the edge of the pack ice where the seals haul out on floes. After the breeding season, Alaskan spotted seals remain on the ice to molt and then move back to the shoreline to spend the summer (Fedoseev, 2000; Lowry et al., 2000; Simpkins et al., 2003). Thus, whereas harbor seals tend to feed in more coastal waters, spotted seals acquire much of their contaminant burdens in distant, more pelagic—therefore, generally less contaminated—environments.

California Harbor Seals

Compared to the corresponding ww or lipid-based blood concentrations reported for marine mammals elsewhere, the average $\sum\text{PCB}_{\text{lw}}$ levels in CA harbor seals (3,222 ppb, sum of 10 congeners) were much greater than recent measurements from bottlenose dolphins off the west coast of Florida (402 ppb, total of tri- through decachlorinated unspecified CBs) (Lahvis et al., 1995), or from adult female harp seals (*Phoca groenlandica*) in the Greenland Sea (201 to 1,447 ppb, sum of 15 congeners) (Lydersen et al., 2002), but lower than levels in adult female grey seals and their pups sampled 20 y ago in Nova Scotia (6,080 and 6,240 ppb, respectively, unspecified number of summed congeners) (Addison & Brodie, 1987). Average $\sum\text{PBC}_{\text{ww}}$ for CA pups (13 ppb) was similar to that of pups in a declining population of Northern fur seals from the Pribilof Islands (16.2 to 22.8 ppb, sum of 14 congeners) (Beckmen et al., 2003) and somewhat greater than those of grey seal pups from Norway (7.7 ppb, sum of 21 congeners) (Jenssen et al., 2003). These comparisons are conservative given the generally greater number of specific CBs summed for ∑PCB in the other studies.

$\text{P,p'-DDE}_{\text{lw}}$ levels in CA harbor seals (9,477 ppb) were far greater than in Florida bottlenose dolphins (237 ppb) (Lahvis et al., 1995), adult female harp seals (approx. 50 to 400 ppb) (Lydersen et al., 2002), and adult female grey seals and their pups (760 and 920 ppb, respectively) (Addison & Brodie, 1987). CA harbor seal pups had greater DDE_{ww} levels (72.3 ppb) than Northern fur seal pups (3.0 to 13.5 ppb) in a declining population of the Pribilofs (Beckmen et al., 2003).

The greatest levels of PCBs and DDE in CA seals were found in pups, with lesser levels in subadult and adult seals. Levels of PCBs and DDE in pups might be expected to be greater than levels in adult females given that the contaminant burden of pups comes from the offloading of fat and associated contaminants into mother's milk. That pups also had greater levels than subadult and adult males indicates that metabolism and excretion of OCs by male harbor seals over time may exceed accumulation via current food web exposure. This pattern is consistent with a hypothesis of mother's

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milk as the major source of lipophilic contaminants (reflecting previously high environmental contamination) coupled with a current plateau or decline in environmental PCB and DDT levels in north-central coastal California (Lieberg-Clark et al., 1995; Jarman et al., 1997; SFEI, 2000; Neale et al., 2005a).

Contaminant loads were greatest during summer for harbor seals in California; this result was further supported by parallel seasonal changes for recaptured individuals. As indicated above, one possible explanation for the seasonal trend in contaminant levels is increased metabolic activity to sustain seals during summer. Alternatively, or in combination with a lipid-metabolism hypothesis, seasonal changes in blood contaminant levels in seals may mirror those of their prey. A recent report on fluctuations in sport fish contamination (SFEI, 2004) documented a summer increase in PCB and DDT levels in sport fish caught in SFB, including several common prey fish of harbor seals such as white croaker (*Genyonemus lineatus*) (Harvey & Torok, 1994).

Greater levels of PCBs in seals of SFB compared with MB seals were expected. The highly industrialized SFB has been heavily contaminated with organochlorines, and PCB in particular, since the 1940s. The SFB is the largest estuary in California, and, relative to MB (and Bristol Bay), is largely interior with a small mouth to the outer coastal waters. This configuration reduces water circulation and, coupled with historically high inputs from the surrounding developed areas, serves to retain high levels of contaminants. Sources of PCBs to SFB have included industrial sites, direct emissions (when emission guidelines were much less stringent than we now have), and landfills with improperly stored PCB-contaminated waste. Although new inputs are probably negligible due to the U.S. ban on PCB production and use restrictions in the 1970s, a current major source of PCBs to surface water (and subsequently to marine food webs) is remobilization/redeposition of residues in soils, sediment, or the atmosphere (SFEI, 1995). Indeed, a recent mass budget model for PCBs using sediment and water data indicated no significant changes during recent decades (Jarman et al., 1997; SFEI, 1998).

DDE concentrations were greater in harbor seals of MB than in harbor seals in SFB, probably because of a more concentrated source of DDTs in the watershed of Elkhorn Slough. Probable sources of DDE and other pesticides to MB are the intensively farmed Salinas Valley to the south (drained by the Salinas River) and the Pajaro Valley to the north (drained by the Pajaro River). Elkhorn Slough, where the majority of MB seals were captured, is centrally located in MB. This slough extends several km inland, and its waters receive inputs from both of these highly cultivated valleys.

The MB sample included eight pups from Pebble Beach, located on the Monterey Peninsula (all others were from Elkhorn Slough). Due to its outer coast exposure, Monterey Peninsula might be expected to differ from central MB with respect to contaminant loads of harbor seals. However, we compared loads in these eight pups to those of pups captured in Elkhorn Slough ($n = 9$) and found no differences (data not shown). Contaminant burdens in pups largely reflect mother's milk and, thus, are primarily representative of their mothers' exposure history. Movement between areas (at least among adult females) was apparently sufficient to homogenize contaminant differences between these two sites.

The lower average $\sum$PCB in harbor seals from PRH as compared with SFB seals was intriguing, especially because all PRH animals were young and had been sampled during the summer (factors which, based on the above findings, would bias the average toward relatively high burdens). This may reflect the lower levels of contaminants found in outer coast environments relative to SFB. On the other hand, DDE concentrations were not lower in PRH seals than in SFB seals. Furthermore, movements of radio- and satellite-tagged harbor seals between SFB and the Pt Reyes Peninsula and Farallones Islands suggest moderate mixing of these groups, which would also confound comparison of contaminant levels by location in the SFB region.

Conclusions

With this relatively large sample, we aimed to assess correlations between contaminant levels in harbor seals and demographic, geographic, seasonal, and physiological variables. We determined distinct factors of PCB and DDE levels in harbor seals from north-central California vs seals from Bristol Bay, AK, confirming some apparent correlations between sex, age, and contaminant loads of earlier studies while highlighting relationships not previously reported. In AK, contaminant levels were substantially less and largely determined by sex and age, whereas for harbor seals in CA, capture site, season, and age were primary factors explaining contaminant levels. Season could not be assessed for AK seals because all captures occurred during September.

The differences in relevant factors affecting contaminant loads in harbor seals in CA and AK may relate to the greater loads in CA seals compared with AK and/or the relatively more complex and heterogeneous exposure environment in CA, with

AK representing the more simple case (sex/age effects), which is obscured in the CA sample by high variation in contaminant exposure. Such variation is likely due to the local point and non-point sources of OCs creating hotspots of environmental contamination in north-central CA locations and especially in the SFB. Additional sampling may illuminate more clearly the differences among sites and sex/age classes. Finally, these findings underscore the importance of controlling for demographic and other variables in studies associating tissue burdens of OCs with parameters of health, and they provide robust OC residue estimates for north-central CA and Bristol Bay, AK, harbor seals which will be useful in management and conservation efforts.

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