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POLYCHLORINATED BIPHENYL PROFILES IN RINGED SEALS (*PUSA HISPIDA*) REVEAL HISTORICAL CONTAMINATION BY A MILITARY RADAR STATION IN LABRADOR, CANADA

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Abstract: Significant amounts of soil contaminated with polychlorinated biphenyls (PCBs) were discovered at a military radar station in Saglek Bay, Labrador, Canada, in 1996. Subsequent work showed elevated PCB concentrations in local marine sediments, in the benthic-associated food web, and in some ringed seals (*Pusa hispida*). The benthic-associated food web clearly reflected local PCB contamination, but the high PCB concentrations found in some ringed seals remained unexplained. In the present study, the authors assess the extent to which this local PCB source at Saglek Bay is contributing to the contamination of ringed seals in northern Labrador. Among 63 ringed seals sampled along the northern Labrador coast, 5 (8%) had PCB levels that were higher than recorded anywhere else in the Canadian Arctic. In addition, compared with seals exhibiting a long-range signal, 45% and 60% of subadults and adult males, respectively, exhibited heavier PCB congener profiles as characterized by principal components analysis, >1.6-fold higher PCB/organochlorine pesticides ratios, and higher PCB concentration-weighted average log octanol–water partition coefficient values, consistent with a local source. Despite the spatially confined nature of contaminated sediments in Saglek Bay, the influence of this PCB source is not inconsequential; PCB concentrations in locally contaminated adult males are 2-fold higher than concentrations in those exposed only to long-range PCB sources and exceed an established threshold of 1.3 mg/kg for adverse health effects in seals. *Environ Toxicol Chem* 2014;33:592–601. © 2013 SETAC

Keywords: Ringed seal   Contaminated site   Polychlorinated biphenyls   Saglek Labrador   Marine mammal   Point source

INTRODUCTION

Persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) bioaccumulate to very high concentrations in high-trophic-level Arctic marine mammals [1,2]. Although long-range atmospheric transport is the primary pathway by which POPs reach the Arctic [3,4], local sources within the Arctic (e.g., military sites) also exist and can contaminate local food webs [5,6]. A PCB source (commercial Aroclor 1260 mixture) associated with a military radar station in Saglek Bay, Labrador, Canada, has contaminated the local benthic food web [6–8]. Saglek Bay has been the site of a military radar station since the late 1950s; however, it was not until 1996 that the PCB contamination, in excess of 50 parts per million (ppm), the maximum allowable amount specified in the Canadian Environmental Protection Act PCB material storage regulations, was discovered in 3 areas (Site Summit, Antenna Hill, and beach area) at the site, along with evidence that PCBs had entered the marine ecosystem of Saglek Bay [6].

From 1997 to 1999, concurrent with the terrestrial remediation of PCBs, a study was undertaken to assess PCB contamination in the marine sediments and associated uptake and accumulation in species representing various trophic levels of the local marine food web [6]. An effects-based ecological risk assessment (ERA) was completed to identify risks to the local biota [9]. The PCB-contaminated marine sediments were widely distributed between the mainland, where the radar station is situated, and Big Island, located approximately 6 km to the north [6]. Average PCB concentrations in the nearshore marine sediments exceeded the Canadian sediment quality guideline (21.5 ng/g dry wt [10]) by 41-fold, and PCB concentrations in benthic invertebrates, a bottom-feeding fish (shorthorn sculpin, *Myoxocephalus scorpius*), and a diving seabird (black guillemots, *Cepphus grylle*) were exceptionally high [6]. Results of the ERA indicated that the survival and reproduction of shorthorn sculpin and black guillemot nestlings were at risk [9]. Relatively high PCB concentrations were also measured in some ringed seals (*Pusa hispida*) and great black-backed gulls (*Larus marinus*), and an alarmingly high PCB concentration (9400 ng/g wet wt) was measured in the blubber of a 10-yr-old male ringed seal from Saglek Bay [6]. Comparable concentrations had never been previously found in ringed seals in the Canadian Arctic [6]. Concentrations of PCBs in surface sediments and the marine benthic-associated food web clearly reflected the input of the local source, but no conclusions could be drawn for the high PCB concentrations measured in the ringed seals [6]. These marine mammals have a home range (2138 km, average migration distance for ringed seals in the Beaufort and Chukchi Seas [11]) that is far greater than the spatial extent (~10 km^2) of the contamination at Saglek Bay [6,12,13] and thus were not expected to be strongly influenced by the local PCB contamination.

Ringed seals are especially vulnerable to elevated exposure of PCBs as a result of their high trophic level, low detoxification capacity, large lipid reserves, and long life span [14,15]. Elevated PCB concentrations in ringed seals from the northern Labrador coast could represent a serious threat to the health of

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ringed seals and the local wildlife that prey on them (e.g., polar bears). Furthermore, because ringed seals are an important part of the local Inuit diet [16], PCB exposure of local communities harvesting along the northern Labrador coast may be of significant concern. By understanding these PCB sources in ringed seals and their concentrations, we can better understand and manage the potential health risks and effects.

Marine mammals are exposed to complex mixtures of POPs, so researchers have used a variety of tools to pinpoint regional (i.e., local) POP sources. Persistent organic pollutant ratios are one approach that has been used to assess regional sources of POPs transferred to predators from their prey [17–19]. As a consequence of the heavy use of DDT in California before the ban in the 1970s, the $\sum$DDTs/$\sum$PCBs ratio is generally higher in Californian marine species than in species from other North Pacific locations, thereby providing a California signature [17,20]. Polychlorinated biphenyls congener pattern analysis is another approach used to determine a regional signature (i.e., a result of heavier congeners from regional sources adhering to organic particles and remaining closer to source) versus long-range signature (i.e., a result of the volatilization of PCBs away from distant sources) [21]. For example, as a result of long-term discharges of PCBs into Puget Sound, harbor seals from this area were found to have a regional or heavy (i.e., increased degree of chlorination) PCB signature relative to harbor seals from more remote waters, which had a global or long-range signature, with a greater contribution of lighter PCB congeners [21].

The objective of the present study was to assess whether the local source at Saglek Bay is contributing to the elevated PCB levels in ringed seals from the northern Labrador coast. To this end, we investigated not only PCBs in ringed seals but also 5 OCPs that usually exhibit patterns of transport and fate similar to those of PCBs. Ringed seals were collected from 4 marine inlets in northern Labrador (Figure 1): Nachvak Fjord, a pristine fjord surrounded by Torngat Mountains National Park; Saglek Fjord, a fjord with a PCB point source of contamination; Okak Bay, a fjard in central Labrador frequently used for harvesting and travel by Inuit from Nain, the nearest community approximately 100 km to the south; and Anaktalak Bay, another fjard in central Labrador that is the shipping route to a mine and concentrator at the head of the bay (Figure 1). A fjard is a type of inlet that is similar to a fjord; however, it has shallow, irregularly shaped, and glacially formed inlets with more gently sloping sidewalls and large intertidal zones [22].

No known local sources of OCPs exist along the northern Labrador coast; therefore, seals that have been influenced by Saglek's local source would likely show elevated levels of PCBs relative to OCPs. We also evaluated the patterns of PCB congeners in ringed seals to determine whether there is a difference in congener composition, signifying more than 1 PCB source. Condition and diet indices were evaluated to eliminate the ecological and biological factors that confound the interpretation of contaminant data in marine mammals.

MATERIALS AND METHODS

Sample collection

Tissue samples (muscle and blubber) from ringed seals ($n = 63$; Table 1) were obtained from Inuit hunters at several locations in the 4 marine inlets during the fall (September and October) of 2008. Prey species, shorthorn sculpin (*Myoxocephalus* scorpius), sand lance (*Ammodytes* spp.), daubed shanny (*Leptoclinus maculatus*), rock cod (*Gadus ogac*), northern astarte (*Astarte borealis*), and chalky macoma (*Macoma calcarea*), were collected from 2008 to 2011 from the zone of contamination in Saglek Bay (i.e., Saglek Anchorage) and from several locations in the 3 reference inlets. All samples were placed in aluminum foil and Whirl Pak bags and frozen at −20 °C until analysis. Sex, weight, length, girth, and blubber thickness (at the sternum) were determined in the field for all ringed seals. Ages were determined by Matson's Laboratory, USA, by longitudinally thin sectioning a lower canine tooth and counting the annual growth layers in the cementum using a compound microscope and transmitted light [23]. All samples were stored at −20 °C until analyzed for stable isotopes (muscle) and organochlorines (blubber) within 1 yr of sample collection. For all samples collected, appropriate permits and community approval were obtained from the Nunatsiavut Government, Nunatsiavut Health, and Environment Review Committee and Department of Fisheries and Ocean Canada.

Chemical analysis

Concentrations of 53 PCB congeners (19, 18/17, 16/32, 26, 28/31, 33/20, 22, 45, 46, 52, 49, 47/48, 44, 64/41/71, 74, 70/76, 66/95, 60/56, 92/84/101, 99, 87, 85, 110, 151/82, 144/135, 149, 118, 146, 153, 105/132, 141, 137, 130/176, 138/163, 158, 178, 187/182, 183, 128/167, 185, 174, 177, 156/171/202, 157/201, 172, 180, 170/190, 199, 203/196, 195/208, 207, 194, 206) and OCPs (α-, β-, γ-hexachlorocyclohexane; α- and γ-chlordane; *cis*-nonachlor; *trans*-nonachlor; oxychlordane; heptachlor epoxide; dichlorodiphenyldichloroethane [*p,p′*-DDD]; dichlorodiphenyldichloroethylene [*p,p′*-DDE]; *p,p′*-DDT; dieldrin; hexachlorobenzene [HCB]) and percentage lipid were measured in ringed seal blubber samples and prey by the Great Lakes Institute for Environmental Research's accredited organic analytical laboratory, Windsor, Ontario, Canada (Canadian Association for Laboratory Accreditation and ISO17025 certified). Organochlorine pesticide (Quebec Ministry of Environment Congener Mix) and PCB standard mixtures were supplied by Sigma-Aldrich and AccuStandard, respectively. The 1,3,5-Tribromobenzene (TBB) was used as the OCP/PCB recovery efficiency standard.

Homogenized wet tissue (0.5–1 g), anhydrous sodium sulfate, and surrogate standard were ground with motor and pestle and then extracted following the microextraction technique described previously [24,25]. Sample lipid contents were determined gravimetrically using 1 mL of sample extract and a microbalance [26]. The remaining extract was concentrated to 2 mL under vacuum with sample cleanup performed by Florisil (F100-500; Fisher Scientific) chromatography as described by Lazar et al. [27], followed by collection of the first (50 mL hexane; ACP) and second (50 mL; hexane/dichloromethane 85/15 v/v; Fisher Scientific) fractions. After Florisil chromatography, extracts were concentrated to 1 mL under vacuum and transferred to 1.8-mL gas chromatography vials. Samples were analyzed for individual PCB congeners and OCPs by gas chromatography electron capture detection (GC-ECD).

For each batch of 6 samples, an in-house reference homogenate tissue (carp muscle), method blank, and external TBB recovery standard were analyzed. All PCB congeners and OCPs were detected with sufficient frequency to be included in the data analysis. Recoveries of individual PCB congeners in the homogenate reference tissue with each batch of samples were within 2 standard deviations of the mean laboratory database value derived from laboratory control charts. Recovery efficiencies for the TBB standard were $89 \pm 0.9\%$ (mean ± standard error). Procedural blanks ($n = 18$) were below detection

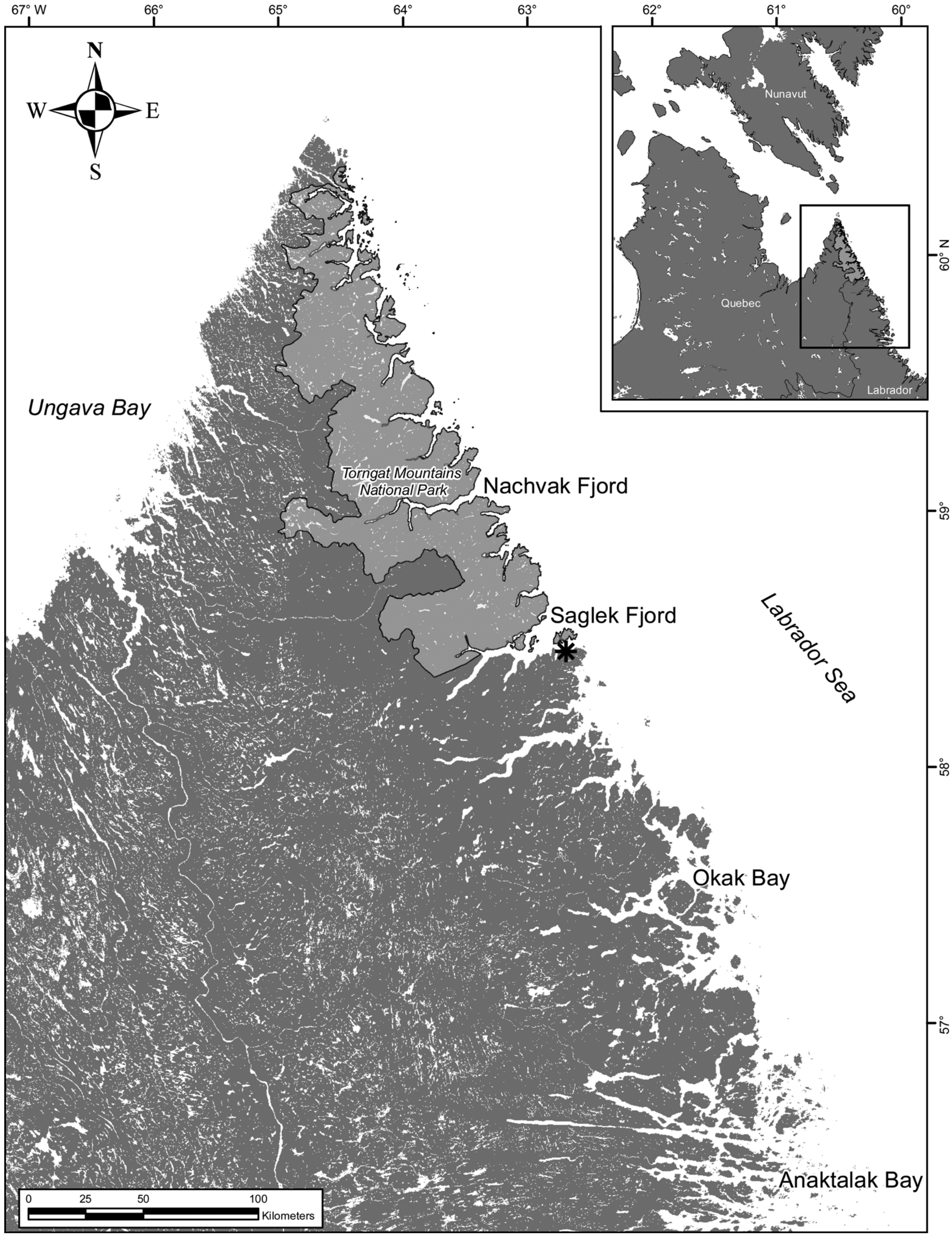

Figure 1. Map of northern Labrador, Canada, showing the location of the 4 fjords where ringed seal collections were taken. Asterisk shows the location of the former polychlorinated biphenyl (PCB) source and PCB-contaminated sediments at Saglek Bay.

for all PCB congeners and OCPs. Sample PCB congener and OCP concentrations were recovery corrected.

Hereinafter, $\sum$PCBs refers to the sum of the 53 PCB congeners; $\sum$HCH refers to the sum of α-, β-, and γ-hexachlorocyclohexane; $\sum$chlordanes refers to the sum of α- and γ-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, and heptachlor epoxide; and $\sum$DDT refers to the sum of *p,p′*-DDD, *p,p′*-DDE, and *p,p′*-DDT.

Stable isotope analysis

Muscle tissue was freeze-dried and homogenized. Lipid was extracted from all samples by agitating the dried powered muscle tissue in a 2:1 chloroform–methanol solution for 24 h. The tissue and solvent were then filtered, and the resulting residue-filter paper dried at 60 °C for 48 h to evaporate the remaining solvent. Next, 500 μg of lipid-extracted tissue was weighted into tin capsules, and stable carbon and nitrogen isotope ratios were

Table 1. Morphometric, percentage lipid, and stable isotope data for subadult (<6 yr; male and female combined), adult male (≥6 yr), and adult female (≥6 yr) ringed seals collected from northern Labrador, Canada[a]

	Subadults	Adult males	Adult females
N	22	20	21
Age in years (range)	0.45 (0 – 4)	16.1 (6 – 28)	13.6 (6 – 25)
Sex (male:female)	6:16	NA	NA
Length (cm)	110 ± 2.0	129 ± 1.9	141 ± 3.0
Girth (cm)	81 ± 1.7	111 ± 2.2	107 ± 2.4
Blubber thickness (cm)	3.8 ± 0.2	5.3 ± 0.2	5.6 ± 0.2
Percentage lipid (blubber)	93.3 ± 0.7	93.7 ± 0.5	91.5 ± 1.6
$\delta^{13}C$ (‰)	−18.6 ± 0.1	−18.0 ± 0.1	−17.3 ± 0.9
$\delta^{15}N$ (‰)	13.7 ± 0.1	14.6 ± 0.2*	14.2 ± 0.2*

[a]Values represent mean ± standard error. Stable isotope data were obtained from muscle tissue.
NA = not available.
*$p < 0.05$ compared with subadults.

analyzed by continuous flow ion ratio mass spectrometer (Finnigan MAT Delta^{plus}; Thermo Finnigan). Stable isotope abundances are expressed in delta (δ) values as the deviation from standards in parts per thousand (‰) using the equation

$$\delta_{\mathrm{sample}}\text{‰} = [(\mathrm{R}_{\mathrm{sample}}/\mathrm{R}_{\mathrm{standard}}) - 1] \times 1000 \qquad (1)$$

where R is the ratio of heavy to light isotope ($^{15}N/^{14}N$ or $^{13}C/^{12}C$) in the sample and standard. The nitrogen stable isotope standard was atmospheric nitrogen; Pee Dee Belemnite limestone formation was the standard for the carbon stable isotope. Precision based on 2 standards (bovine muscle; NIST 8414] and an internal laboratory standard [tilapia fish muscle], $n = 144$ for each) were <0.17‰ and <0.09‰ for $\delta^{15}N$ and $\delta^{13}C$, respectively. Accuracy of isotope analysis, based on NIST standards (sucrose [NIST 8542] and ammonia sulfate [NIST 8547], n = 3 for each) analyzed during the study were within <0.1‰ of certified $\delta^{15}N$ and $\delta^{13}C$ values.

Data analysis

Total PCB and OCP concentrations and PCB congener composition in the blubber of phocids are influenced by age, sex, and reproductive status [28–30]. To control for these confounding factors, we separated the data into 3 groups for statistical exploration: subadults (<6 yr, male and females combined), adult males (≥6 yr), and adult females (≥6 yr). Univariate statistical analyses were performed in SPSS 20.0 for Windows. The level of statistical significance used was $p \leq 0.05$. Data were log transformed when necessary to meet the normality assumptions for parametric analyses. Any PCB congeners less than the detection limit were replaced with a random number between the detection limit and zero. Geometric means and ranges were calculated and tissue concentrations were converted to a lipid-weight basis by dividing wet-weight values by the proportion of lipid in the samples. The ∑PCB and OCP (∑DDT, ∑chlordanes, ∑HCH, dieldrin, HCB) concentrations are expressed on a lipid-weight basis. Linear regression analysis was used to determine relationships between contaminant concentrations and condition indices (length, girth, blubber thickness, percentage lipid) and stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and between stable isotopes and condition indices. One-way analysis of variance (ANOVA) was used to compare the mean of contaminant concentrations and stable isotopes among subadult and adult ringed seals by gender and differences between locations. A Tukey's posthoc test was performed to determine differences among the locations when significant differences were found by ANOVA.

Principal components analysis (PCA) was used to elucidate differences in PCB patterns in subadults and adult male ringed seals. Females seals were not used to study the differences in PCB patterns or ratios, because females can transfer individual POPs and congeners at differential rates to their offspring [2,31]. Samples were standardized to the total concentration total before PCA to remove artifacts related to concentration differences between samples. The centered log ratio transformation (division by the geometric mean of the concentration-normalized sample followed by log transformation) was then applied to these data sets to produce a data set that was unaffected by negative bias or closure [21]. Data were autoscaled before PCA. One outlier was removed from the adult male PCA model.

Linear regression was used to assess the relationships between PCA projections and ∑PCB/OCP ratios, log octanol–water partition coefficient (K_{OW}) values (a proxy for particle affinity), and location. Log K_{OW} values for the PCB congeners were taken from Hawker and Connell [32].

The log K_{OW} for each PCB congener was used to calculate concentration-weighted average log K_{OW} values [33] for each subadult and adult male ringed seal according to the following equation.

$$\text{Concentration-weighted average } \log K_{OW} \text{ value} = \frac{\sum \textit{concentration of individual congener} \times (\textit{log}\ \mathrm{Kow})\ \textit{value for that congener}}{[\textit{total concentration of all congeners}]} \qquad (2)$$

A one-way ANOVA was used to compare the mean of contaminant concentrations, ∑PCB/OCP ratios, and concentration-weighted average log K_{OW} values among subadults and adult ringed seals for differences between local and long-range seals.

RESULTS AND DISCUSSION

PCBs and organochlorine pesticide concentrations

The average concentrations for condition indices (length, girth, blubber thickness, percentage lipid; Table 1) and the average concentrations of PCBs and OCPs found in ringed seals in the present study (Table 2) generally fell within the range reported previously for ringed seals in the Canadian Arctic [1,34–36]. However, 2 adult females (1090 ng/g lipid wt, 6-yr-old from Okak Bay; and 1050 lipid wt, 9-yr-old from Nachvak Fjord) and 3 adult males (3160 ng/g lipid wt, 22-yr-old from Saglek Bay; 3380 ng/g lipid wt, 28-yr-old from Saglek Bay; and 8770 ng/g lipid wt, 20-yr-old from Nachvak Fjord) exceeded the highest values for PCBs reported for adult female (821 ng/g lipid wt) and adult male (3060 ng/g lipid wt) ringed seals at other Canadian Arctic locations (Holman, Resolute, Eureka, Arctic Bay, Grise Fjord, Pangnirtung) over the past 20 yr [1,35,37];

Table 2. Geometric means and ranges (ng/g lipid wt [range, 95% upper confidence limit]) of ∑polychlorinated biphenyls (PCBs) and organochlorine pesticide concentrations in blubber tissue of subadult (<6 yr; male and females combined), adult male (≥6 yr), and adult female (≥6 yr) ringed seals collected from northern Labrador, Canada

	Subadults	Adult males	Adult females
∑PCBs	356 (180 – 886, 494)*	818 (139 – 8770, 2380)	337 (69 – 1090, 543)*
∑DDTs	142 (51 – 522, 202)*	309 (71 – 1010, 571)	133 (30 – 851, 297)*
∑Chlordanes	79 (40 – 162, 100)*	163 (30 – 616, 303)	73 (16 – 307, 139)*
∑Hexachlorocyclohexanes	49 (29 – 83, 57)	49.7 (18.5 – 118, 70)	45 (21 – 85, 58)
Dieldrin	25 (11 – 95, 40)	32 (10 – 78, 45)	25 (10 – 112, 40)
Hexachlorobenzene	4.4 (2.4 – 6.5, 4.9)*	5.1 (2.8 – 8.0, 5.9)	4.7 (3.1 – 22, 7.2)

*$p < 0.05$ compared with adult males.

Hoekstra PF, et al., National Water Research Institute, Burlington, Ontario, Canada, unpublished data. The OCP concentrations in 4 (3 adult males and the 6-yr-old female) of the 5 seals were not elevated relative to the other ringed seals in the present study and were similar to OCP concentrations reported for ringed seals from other Canadian Arctic locations [1,34,35]; Hoekstra PF, et al., National Water Research Institute, Burlington, Ontario, Canada, unpublished data. As a result, concentrations of $\sum$PCBs relative to the 5 OCPs measured (i.e., $\sum$PCB/OCP ratios) in these 4 seals were higher than the mean $\sum$PCB/OCP ratios for adult males and females collected from the area (Supplemental Data, Figure S1). The elevated levels of PCBs relative to OCPs in these seals are most likely explained by the influence of different PCB sources (e.g. Saglek's local PCB contamination to the marine environment). The 9-yr-old adult female from Nachvak, however, had relatively high concentrations of all 5 OCPs, and as a result the $\sum$PCB/OCP ratio was lower than the mean $\sum$PCB/OCP ratio for adult females. A possible explanation for this is that this female seal, which was not pregnant at the time of sampling, is infertile and as a result maintains a higher PCB and OCP burden.

Only 2 of the 5 ringed seals with elevated $\sum$PCB concentrations were from Saglek Fjord. Although these 2 adult males may display some degree of site fidelity to this particular fjord, the other 3 seals from reference fjords suggest that a portion of the population may roam more widely. From our entire data set, average PCB concentrations in adult male ringed seals from Saglek Fjord (1751 ng/g lipid wt; $n=8$) were higher ($p=0.01$) than concentrations detected in those sampled in Nachvak Fjord (448 ng/g lipid wt; $n=6$; Supplemental Data, Table S1). No differences ($p\geq 0.05$) were found among the 4 fjords for average concentrations of OCPs for adult males (Supplemental Data, Table S1). No differences ($p \geq 0.05$) were found among the 4 fjords for average concentrations of $\sum$PCBs and OCPs for adult female and subadult ringed seals (Supplemental Data, Tables S2, S3). The results indicate that location influences PCB exposure more in adult males than in adult females or subadults.

The 5 seals with exceptionally high PCB concentrations from the present study support the previous observation of an anomalous 10-yr-old male seal with unusually high PCB levels in the Saglek Bay area [6]. To our knowledge, comparable PCB concentrations have not been found previously in ringed seals from other areas in the Canadian Arctic. Muir et al. [38] did, however, find a surprisingly high concentration (4500 ng/g) in a 7-yr-old female seal from Inukjuak (eastern Hudson Bay), which was collected sometime between 1989 and 1991, but the concentration was still not as high as the concentrations reported in the 20-yr-old male from Nachvak from the present study and the 10-yr-old male ringed seal from Saglek collected during the 1997 to 1999 site investigations [6]. The PCB congener pattern was examined for the 7-yr-old female seal from Inukjuak, but the authors concluded that there was no evidence to suggest the influence of different PCB sources or altered metabolism [38].

PCB congener patterns in subadult and adult male ringed seals

The first principal component (p1: subadults = 47.4% and adult males = 53.1%) clearly differentiates ringed seals with a greater proportion of the lighter (less-chlorinated) congeners from ringed seals with a greater proportion of the more heavily chlorinated congeners (Figure 2). The light PCB signature is typical of a long-range atmospheric transport signal [39], which is a result of more volatile (e.g., lighter) congeners traveling great distances, whereas the heavy signature is more characteristic of a local source signal, in which more chlorinated congeners (e.g., heavier) dominate the composition of PCBs [21]. These divergent PCB signatures in ringed seals are consistent with observations in harbor seals in which proximity to regional sources explained their heavier PCB profiles compared with those in more remote locations [21].

The $\sum$PCBs/OCP ratios correlated ($p<0.05$) with t1 (the sample scores of the first principal component) for subadult and adult male ringed seals (Figure 3 and Supplemental Data, Table S4), indicating that ringed seals with a heavier PCB signature had a higher PCB concentration relative to the 5 different OCPs. These results are consistent with our observations above for 4 of the 5 ringed seals with elevated PCB concentrations. To validate our results further, we assessed whether t1 correlated with $\sum$PCBs/*p,p'*-DDE and $\sum$PCBs/*trans*-nonachlor ratios, all of which have been shown to biomagnify in seals. The $\sum$PCB/*p,p'*-DDE and $\sum$PCB/*trans*-nonachlor ratios correlated ($p<0.05$) with t1 (Supplemental Data, Table S4). These relationships support previous reports of regional sources of POPs being transferred to top predators from their prey, whereby the concentration of a regional-source POP would be elevated relative to POPs with no known regional or local sources [17–19].

The log of total PCBs was correlated with t1 for subadult and adult male ringed seals ($p<0.001$; Figure 3). No relationship was found among the 5 OCPs ($\sum$DDTs, $\sum$HCHs, $\sum$chlordanes, dieldrin, HCB) and t1 ($p\geq 0.05$). These results are consistent with the observations presented above, with the more heavily chlorinated seals having higher PCB concentrations relative to the OCP concentrations (i.e., higher $\sum$PCB/OCP ratios) than the lighter, less chlorinated, long-range seals. A correlation was found between log K_{OW} for the PCBs and p1 (the variable loadings of the first principal component of the individual PCB congeners) for subadult and adult male ringed seals ($p<0.001$; Figure 4). Our results are consistent with those of Ross et al. [21] showing a PCB profile that strongly correlated with both total PCB concentrations and log H (Henry's law constant, which describes the partitioning between surface waters and the atmosphere). Log K_{OW} values and log H constants can be used to characterize the partitioning of PCB congeners across different environmental interfaces (e.g., air–water or lipid–water) [32]. As such, they play a key role in affecting the environmental fate and bioaccumulation of POPs in top predators. Polychlorinated biphenyls concentrations from ringed seal prey species were added to a second PCA to provide a more complete view of the regional food web and to illustrate further the influence of the Saglek PCB source on adult male seals sampled (Supplemental Data, Figure S2). This PCA clearly shows that Saglek prey species to the left of the score plot are dominated by heavier congeners (i.e., local signature), whereas reference fjord species to the right of the score plot are dominated by lighter congeners (i.e., long-range signature).

The t1 was correlated with location (4 marine inlets) for adult males ($p=0.015$). No correlation was found between t1 and location for subadults ($p\geq 0.05$). Among the subadult and adult male ringed seals with a heavy local PCB signature, 30% and 58% respectively, were from Saglek Fjord (Figure 2). These results suggest that these seals might have shown some site fidelity to this fjord and therefore had a greater chance of exposure to the local PCB contamination. The higher percentage observed for adult male ringed seals is consistent with other studies [40–42] in which adult ringed seals were more likely to demonstrate signs of site fidelity than younger subadults that

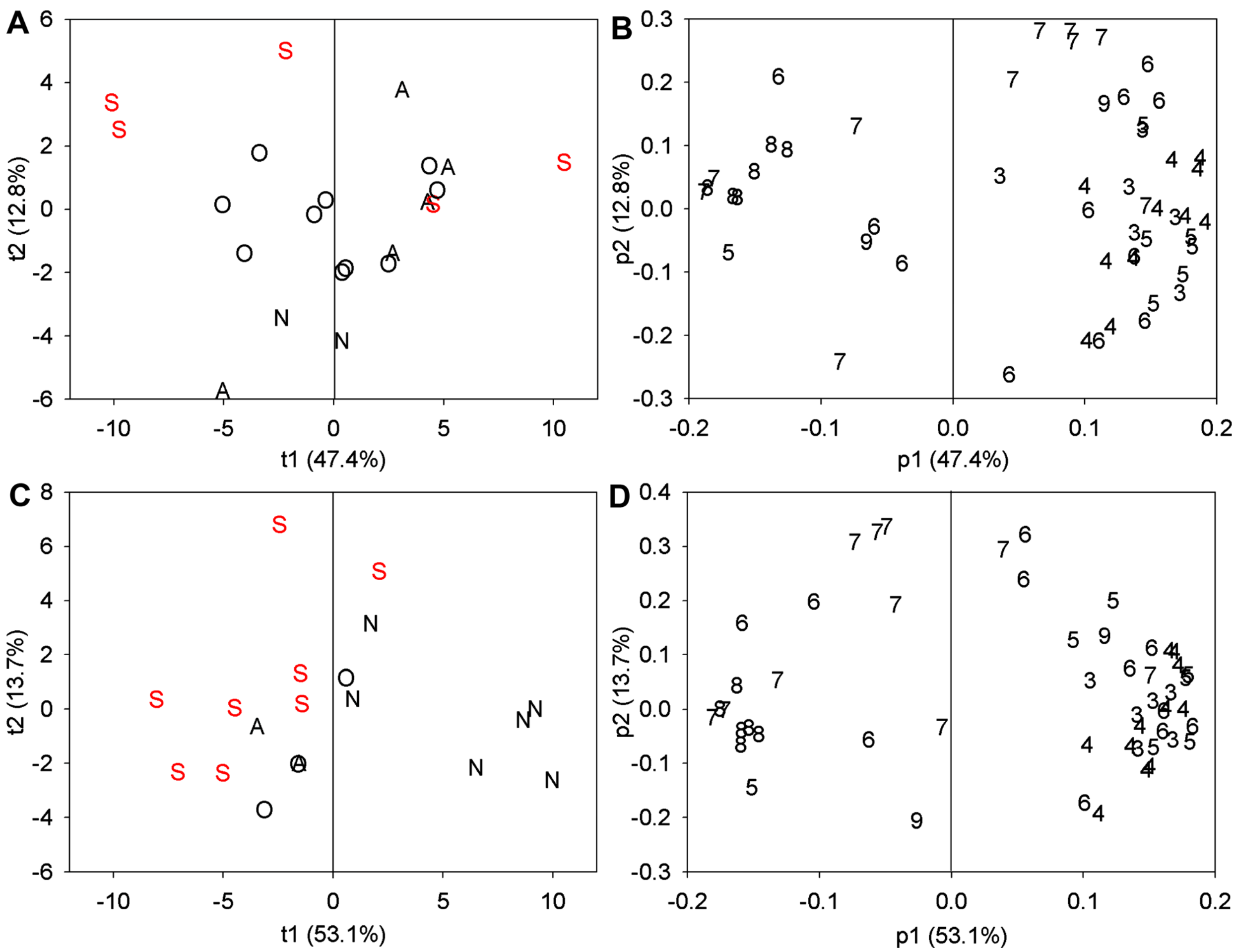

Figure 2. Principal components analysis (PCA) of polychlorinated biphenyl (PCB) patterns (53 congeners) in subadult (**A**,**B**) and adult male (**C**,**D**) ringed seals reveals that seals to the left of each score plot (**A**,**C**) are dominated by heavier congeners (B,D), consistent with exposure of a local PCB source. (**A**) and (**C**) are PCA scores for individual seals; (**B**) and (**D**) are PCA loadings for the individual PCB congeners. Symbols represent seals from 4 fjords in Labrador (N = Nachvak; S = Saglek; O = Okak; A = Anaktalak). (**B**,**D**): Numbers identify the degree of chlorination of each PCB congener (i.e., number of chlorines per congener). One adult male outlier was removed, a 20-yr-old ringed seal from Nachvak Fjord with elevated PCB concentration (8770 ng/g lipid wt). This seal was dominated by heavier congeners (PC1 = − 16) and fell to the far left of the PCA.

roam more widely. These observations with PCB patterns complement the finding of higher total PCB concentrations in adult males sampled in Saglek. The remaining 70% and 42% of subadult and adult male ringed seals, respectively, with a heavy local PCB signature were from reference fjords (Figure 2; Nachvak, Okak, Anaktalak), which were sampled within 250 km of the source (Figure 1). The 20-yr-old adult male ringed seal from Nachvak, which had the highest PCB concentration (8,770 ng/g lipid wt) in the study, had the heaviest PCB profile (PC1 = − 16). A possible explanation for these reference seals with a local PCB signature is that they have fed previously in the contaminated area of Saglek Bay. The distance to which these

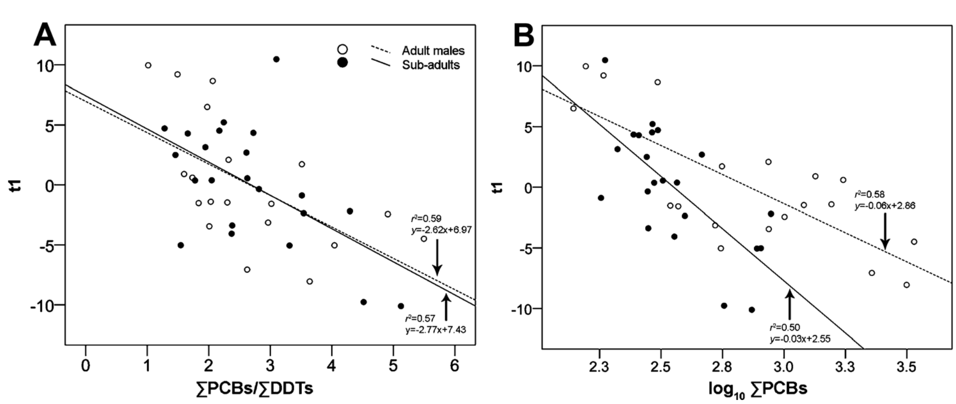

Figure 3. The first principal component (t1) is correlated with ∑polychlorinated biphenyls (PCBs)/organochlorine pesticides (OCPs) ratios (**A**) ∑PCBs/∑DDTs shown here; see Supplemental Data, Table S4, for other ∑PCBs/OCP ratio results) and $\log_{10}$ ∑PCBs ng/g lipid weight for adult male and subadult ringed seals (**B**).

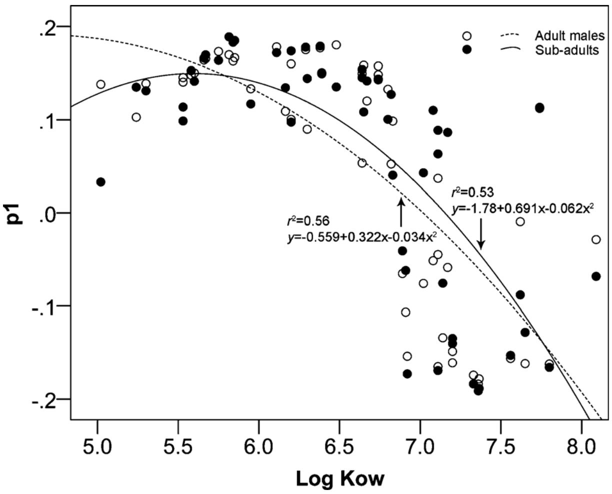

Figure 4. The first principal component (p1) for adult males (solid line) and subadults (dashed line) correlated with the octanol–water partition coefficient (K_{OW}) for polychlorinated biphenyls (PCBs). The data pattern for adult males and subadults suggests that there are 2 groups of ringed seals along the Labrador coast, seals that have been exposed to a heavier PCB mixture from a local source (e.g., Aroclor 1260, local Saglek signature) and seals that have been exposed to a lighter PCB mixture from long-range atmospheric sources.

seals might have travelled away from Saglek is not uncommon; ringed seals are known to carry out extensive migrations [43], in some instances traveling thousands of kilometers [44]. Further research is needed to characterize the role of movement and foraging patterns in influencing POP concentrations in ringed seals for the area.

Collectively, our results strongly suggest that some ringed seals from the north Labrador coast have been influenced by the PCB exposure at Saglek Bay. Because no known local PCB sources other than Saglek exist along the Labrador coast, these results reflect the exposure of 2 distinct PCB sources (i.e., long-range atmospheric transport via the effect of a volatilization of PCBs away from southern sources and a local signal, a result of heavier congeners from Saglek Bay adhering to organic particles and remaining closer to source). It is possible that another local source of PCBs exists on the Labrador coast but is likely to be much smaller [45]. In addition, ringed seals very rarely frequent this area (W. Piercy, AngajukKâk, Hopedale Inuit Community Government, Hopedale, Labrador, Canada, personal communication, 2013).

Although depuration and metabolic processes can deplete ringed seals and their prey of, for example, lighter PCBs [46], a PCA using only recalcitrant congeners revealed no differences from the results presented in Figure 2 (data not shown), suggesting that metabolism was not overtly shaping the pattern differences observed in subadult and adult male ringed seals.

Local contribution to ringed seal PCB body burden

Based on the divergent PCB congener profiles (Figure 2 and Supplemental Data, Figure S3) and PCB/OCP ratios (Figure 3 and Supplemental Data, Table S4), the subadults and adult males were divided into 2 groups. Ringed seals to the left of the t1 axis hereafter will be referred to as local, and ringed seals to the right of the t1 axis hereafter will be referred to as long-range. Our data indicate that 45% and 60% of the subadult and adult male ringed seals, respectively, have been influenced by the local PCB source at Saglek Bay. $\sum$PCB concentrations in local subadults (527 ± 82 ng/g lipid wt) and adult males (1386 ± 329 ng/g lipid wt) were 2-fold higher ($p < 0.05$; Figure 5) than concentrations in long-range seals (subadults 293 ± 21 ng/g lipid wt, adult males 662 ± 214 ng/g lipid wt). No significant differences in OCPs were found between local and long-range subadults or adult males (Figure 5).

The 5 $\sum$PCB/OCP contaminant ratios were significantly higher in local subadult and adult male ringed seals than in long-range seals (Table 3). These results are consistent with the observations presented above, with the more heavily chlorinated local seals having higher PCB concentrations relative to the OCP concentrations (i.e., $\sum$PCB/OCP ratios) than the lighter, less chlorinated long-range seals. Furthermore, the lack of difference found between the local and long-range seals for all OCP concentrations (Table 2) further supports the use of these contaminants as a reference to identify the sources of PCBs to northern Labrador ringed seals. The concentration-weighted average log K_{OW} values for PCBs in local ringed seals (subadults $= 6.8 \pm 0.02$, adult males $= 6.9 \pm 0.01$) was higher than long-range seals (subadults $= 6.7 \pm 0.01$, adult males $= 6.7 \pm 0.03$; $p < 0.001$ in both cases).

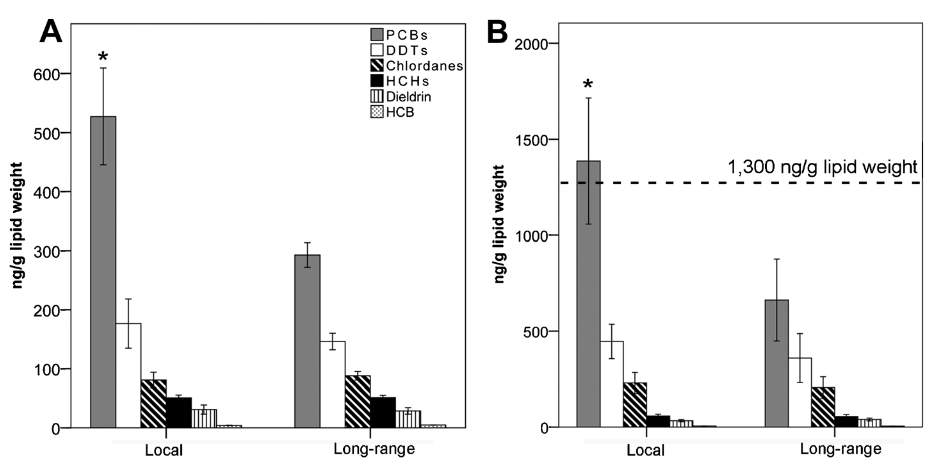

Figure 5. Local and long-range polychlorinated biphenyl (PCB) and organochlorine pesticides (OCP) concentrations measured in the blubber of subadults (**A**) and adult male (**B**) ringed seals. Mean (± standard error) PCB concentrations in subadult (**A**) and adult male (**B**) ringed seals are higher in local ringed seals than in long-range seals, whereas OCP ($\sum$DDTs, $\sum$chlordanes, $\sum$hexachlorocyclohexanes, dieldrin, hexachlorobenzene) concentrations did not differ between groups. Average PCB concentrations in adult male ringed seals exceed the effects threshold (1300 ng/g [ppb] [58] indicated by a dashed line) in a similar pinniped species, the harbor seal. An asterisk above a column indicates significant difference between the local and long-range seals for that compound.

Table 3. Arithmetic means ± standard errors of the ratios of ∑polychlorinated biphenyls (PCBs)/legacy organochlorine pesticides (∑DDTs, ∑chlordanes, ∑hexachlorocyclohexanes [HCHs], dieldrin, hexachlorobenzene [HCB]) in blubber tissue of local and long-range subadult and adult male ringed seals

	Subadults		Adult male	
	Local	Long-range	Local	Long-range
∑PCBs/∑DDTs	3.3 ± 0.3	$2.1 \pm 0.2^{*}$	3.2 ± 0.4	$1.9 \pm 0.3^{*}$
∑PCBs/∑chlordanes	6.7 ± 0.7	$3.5 \pm 0.3^{*}$	6.9 ± 0.8	$3.0 \pm 0.3^{*}$
∑PCBs/∑HCHs	10 ± 1.2	$5.8 \pm 0.3^{*}$	23 ± 3.0	$11 \pm 2.8^{*}$
∑PCBs/ dieldrin	23 ± 3.7	$13 \pm 1.5^{*}$	39 ± 3.9	$16 \pm 4.2^{*}$
∑PCBs/ HCB	125 ± 15	$63 \pm 5.6^{*}$	247 ± 50	$128 \pm 41^{*}$

$^{*}p < 0.05$ compared with the local group.

Variations with sex, condition, and diet

No significant differences ($p > 0.05$) were found between subadult males and females for ∑PCBs and the 5 OCPs (∑DDT, ∑chlordanes, ∑HCHs, HCB, and dieldrin). Concentrations of ∑PCBs, ∑DDT, ∑chlordanes, and HCB were significantly higher in adult male ringed seals than in subadult ringed seals ($p \leq 0.05$). No significant differences ($p > 0.05$) were found between subadult and adult female ringed seals for ∑PCBs and the 5 OCPs. Concentrations of ∑PCBs, ∑DDT, and ∑chlordanes were significantly higher in adult male ringed seals than in adult female ringed seals ($p \leq 0.05$). These differences between males and females are typical for pinnipeds, whereby females are able to maintain a lower burden by transferring a proportion of their contaminant load to offspring via placental and lactational transfer [35,47]. The ∑HCHs, HCB, and dieldrin concentrations in adult ringed seals did not differ ($p > 0.05$) between sexes. No relationship between sex for ∑HCHs and HCB has been observed previously in adult ringed seals [35]. The average concentrations for condition indices (length, girth, blubber thickness, percentage lipid) found in ringed seals from the present study (Table 1) fell within the range reported previously for ringed seals in the Canadian Arctic [1,34–36]. Concentrations of ∑PCBs and all 5 OCPs in subadults, adult males, and adult females did not differ with any of the condition indices ($p > 0.05$).

Values of $\delta^{15}N$ and $\delta^{13}C$ did not influence PCB and OCP concentrations in subadult, adult female, or adult male ringed seals ($p > 0.05$; Table 1). We also evaluated whether feeding ecology was being influenced by sex, age, or condition indices. For subadult and adult ringed seals, there were no sex-related differences in $\delta^{15}N$ or $\delta^{13}C$ values ($p > 0.05$; Table 1). Similarly, no sex-related differences were found in the diet of ringed seals from other Canadian Arctic locations [48,49], Svalbard [50], or Alaska [51]. Contrary to other observations for ringed seals [49,52], adult females and males were enriched in $\delta^{15}N$ over subadults ($p = 0.05$ and $p = 0.001$, respectively; Table 1). No differences were found among adult females and males and subadults for $\delta^{13}C$ ($p = 0.17$ and $p = 0.69$, respectively; Table 1). Similarly, no age-related differences were found for $\delta^{13}C$ for ringed seals from the Canadian Arctic [52]. The $\delta^{15}N$ and $\delta^{13}C$ in subadults, adult males, and adult females were not found to differ with any of the condition indices measured ($p > 0.05$; Table 1).

We evaluated whether diet indices or age were influencing the contaminant concentrations and ∑PCB/OCP ratios in local and long-range ringed seals. No significant differences were found between local and long-range subadult and adult male ringed seals for $\delta^{15}N$. No significant differences were found between local and long-range adult male ringed seals for $\delta^{13}C$. However, local subadult seals had slightly higher $\delta^{13}C$ values than long-range seals (local $= -18.4 \pm 0.1$; long-range $= 18.8 \pm 0.1$; $p = 0.03$), suggesting that local seals were consuming more benthic or inshore prey than long-range seals. Benthic or inshore feeding behavior could increase a seal's chance of being influenced by the PCB contamination at Saglek Bay, such that these seals would be more likely to forage directly within the Saglek Anchorage area and/or Saglek Fjord than seals feeding more pelagically or offshore. No significant differences were found between local and long-range adult male ($p = 0.45$) and subadult ($p = 0.31$) ringed seals for age.

Health risks for northern Labrador ringed seals

Numerous field studies have shown associations between PCBs and adverse health effects (e.g., impaired reproduction, endocrine disruption, bone lesions, reduced immune function, and tumour incidence) in ringed seals [15,53–57]. However, with the confounding factors (e.g., complex mixtures) inherent in field studies, evidence of a cause–effect linkage between an adverse health effect and a single contaminant has been difficult to achieve. As a result, effects thresholds or toxicity reference values (TRV) for marine mammal species are virtually nonexistent [58]. To our knowledge, only 3 toxicity thresholds for PCBs exist for marine mammals. These include a TRV for immunotoxicity (17 mg/kg lipid wt; blubber) determined during a captive feeding study of harbor seals (*Phoca vitulina* [59]), a TRV for reproductive effects (10 mg/kg lipid wt; blubber) established in an epidemiological study of free-ranging bottlenose dolphins (*Turisiops truncatus* [60]), and a TRV for immunotoxicity and endocrine disruption (1.3 mg/kg lipid wt; blubber) in juvenile free-ranging harbor seals (*Phoca vitulina* [59]).

Average PCB levels in adult male seals from the local group exceed the toxicity threshold for immunotoxicity and endocrine disruption established in harbor seals (1.3 mg/kg lipid wt [58]) but are lower than the other 2 thresholds established for marine mammals (Figure 5). The average PCB levels in the long-range adult male and local and long-range subadult ringed seals were below all thresholds established for marine mammals. These results collectively suggest that adult male ringed seals that have been exposed to local sources of PCBs may be at risk for toxic effects, including the alteration of vitamin A levels and its receptor (retinoic acid receptor [RARα]), altered thyroid hormone physiology and receptor function, and impaired immune function [61].

CONCLUSIONS

The present study illustrates how a contaminated site can become a source of PCBs to a wide-ranging, upper-trophic-level species. We found elevated PCB concentrations in a proportion of individual ringed seals from Labrador relative to those observed in ringed seals from across the Canadian Arctic. Divergent PCB patterns and PCB/OCP ratios provided further evidence that seals from both within Saglek Fjord as well as outside this area had been exposed to PCBs from the former radar station. We estimate that approximately 50% of the PCB burden of ringed seals in the region originates from the local source. Despite their ecology (i.e., wide-ranging foraging behavior), our research suggests that ringed seals could be used as an indicator of local marine contamination and to model contaminant accumulation in Arctic food webs. Furthermore, our study shows that locally contaminated adult male ringed seals sampled in 2008 exceed an adverse health effects threshold, despite documented reduction in sediment, bottom-feeding fish (shorthorn sculpin), and seabird (black guillemot) contamination during the period of 1999 to 2006 [6,13].

SUPPLEMENTAL DATA

Tables S1–S4.
Figures S1–S3. (486 KB DOCX).

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