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Satellite Telemetry Informs PCB Source Apportionment in a Mobile, High Trophic Level Marine Mammal: The Ringed Seal (*Pusa hispida*)

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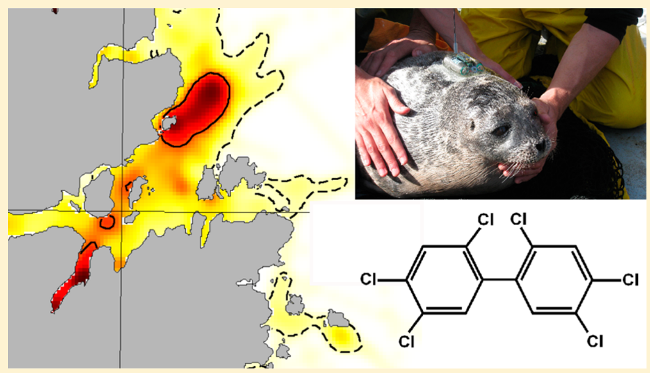
Supporting Information

ABSTRACT: Marine mammals are typically poor indicators of point sources of environmental contaminants as a consequence of their often complex feeding ecologies and extensive movements, all of which mask the contributions of specific inputs. The release of polychlorinated biphenyls (PCBs) by a military radar station into Saglek Bay, Labrador (Canada) has contaminated marine sediments, bottom-feeding fish, seabirds, and some ringed seals, but attributing the PCBs in the latter highly mobile animals to this source is exceedingly difficult. In addition to the application of such tools as stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and univariate and multivariate statistical exploration of contaminant patterns and ratios, we used satellite telemetry to track the movements of 13 seals in their transient use of different feeding areas. Reduced size of home range and core area (i.e., areas of concentrated use), as well as increased time in coastal inlets, were important determinants of increased PCB concentrations in seals reflecting the contribution of Saglek Bay. Seals were classified into the same feeding groups using both space use and their contaminant burdens 85% of the time, highlighting the link between feeding ecology and exposure to PCBs. While the PCB source at Saglek provided a strong local signal in a remote environment, this first use of satellite telemetry demonstrates the utility of evaluating space-use strategies to better understand contaminant exposure, and more specifically the contribution of contaminant hotspots to mobile predators.



INTRODUCTION

Source apportionment for persistent contaminants in high trophic level marine species is notoriously difficult to assess, yet these animals are often the most contaminated and at risk to these bioaccumulative and toxic chemicals.[1] These species generally have a large home range, such that exposure to biomagnifying persistent organic pollutants (POPs) can be attributed to the consumption of prey contaminated by a combination of distant, regional and/or local point sources. Researchers have used a variety of statistical techniques and study designs (e.g., PCB profiles and POP ratios[2−6]) to infer the contributions of regional and/or local POP sources. However, without an understanding of movement and foraging behavior, it remains challenging to adequately explain the contributions of point sources to the body burden of a given species.

Satellite telemetry can be used to assess behavior and movement patterns for marine species by providing detailed records of geographical location. Such information, combined with contaminant residues in prey and/or predator, can provide important insight into the contributions of different source regions. For example, Elliott et al.[7] used satellite tracking of osprey to demonstrate that the northern breeding grounds of this fish-eating bird represented the principal source of contaminant exposure. Satellite telemetry has also been used to investigate geographical differences in contaminant concentrations and patterns in loggerhead sea turtles (*Caretta caretta*) and polar bears (*Ursus maritimus*),[8,9] but this method has never been used to assess the implications of point sources related to spills or discharge of contaminants in marine ecosystems.

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Polychlorinated biphenyls (PCBs) are a POP of concern in arctic food webs, where biomagnification of these persistent and lipophilic contaminants leads to relatively high concentrations in upper trophic level species.[10] PCB contamination in Arctic marine ecosystems is largely attributed to atmospheric deposition following long-range transport from southern industrial source regions, but local sources have been documented in the Arctic (e.g., military radar stations).[11,12] Saglek Bay, Labrador, Canada has been the site of a military radar station since the late 1950s; however, it was not until 1996 that PCB contamination was discovered at the site, along with evidence that PCBs had entered the marine environment.[13] Approximately 260 kg of PCBs had been released into the marine environment,[14] contaminating adjacent marine sediments, benthic invertebrates, bottom-feeding fish, diving seabird, and some ringed seals.[13] While the elevated PCBs in the benthic associated food-web could be causally attributed to the Saglek PCB source, ascertaining the contribution of this local source to the body burden of the highly mobile ringed seals was fraught with uncertainty.[13] "Heavier" PCB profiles and higher PCB:organochlorine pesticide (OCP) ratios provided a recent basis to identify up to 60% of ringed seals sampled in the central and northern Labrador coast as being exposed to the local PCB source at Saglek.[15] This chemocentric approach enabled a classification of seals as either "local" or "long-range", reflecting their respective exposure to these two types of sources.

Given the varying degrees to which different ringed seals move (and therefore feed), the determination of PCBs in biopsy-sampled individuals will yield results reflecting their lifelong exposure via prey in different areas. Some ringed seals have a home range that involves movement over large distances,[16−19] whereas other ringed seals stay within much smaller areas and exhibit strong site fidelity.[19−23]

A number of variables can be used to describe space-use and foraging behavior in ringed seals. For example, "home range" captures the area that each ringed seal utilizes, or covers during its normal activities, including feeding, traveling, resting, and breeding.[24] Very little is known about the home range of ringed seals along the Labrador coast; studies indicate that they remain in Labrador waters throughout the year and undergo relatively short migrations northward during the summer and fall foraging months.[25,26] Other important variables that describe the space-use of ringed seals include the extent of latitudinal and longitudinal movements, time spent in coastal inlets or local areas such as Saglek Fjord, and areas of concentrated use, commonly termed core areas.[24] Core areas represent sites of greater ecological significance to the animal and therefore are a better representation of preferred foraging sites. Measures of time spent feeding benthically and stable isotope ratios (carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$)) are important variables that describe foraging behavior in relation to prey choice. For example, enrichment of $\delta^{15}N$ ratios increases with trophic position in marine food chains providing a continuous variable with which to assess both trophic level and food web transfer.[27] Ratios of $\delta^{13}C$ can elucidate trophic interactions by establishing the relative contribution of inshore/benthic versus offshore/pelagic feeding preferences.[28]

Herein, we use satellite telemetry to evaluate whether contaminant profiles can indeed be used to assign seals to either "local" or "long-range" categories. We previously documented divergent PCB signatures in ringed seals in the region, and concluded that "heavy" signatures reflected localized exposure to the Saglek Bay PCB source, whereas "light" PCB signatures reflected background.[15] We hypothesize here that satellite telemetry and $\delta^{13}C$ and $\delta^{15}N$ can be used as measures of habitat use (hence, feeding ecology) that will validate our PCB signature assignment in ringed seals from the Labrador coast. The mobility of marine mammals, coupled with the dearth of major sources of contaminants in the Arctic, highlight the unique opportunity here to evaluate the utility of satellite telemetry in support of exposure attribution to a high trophic level predator in the marine environment.

■ MATERIALS AND METHODS

Seal Capture and Handling. During August and September (2008−2011), 13 ringed seals (2008: $n = 3$; 2009: $n = 3$; 2010: $n = 5$; 2011: $n = 2$) were captured from various locations in Saglek Fjord, Labrador, Canada. Following the methods of Smith et al.,[29] we captured ringed seals using green monofilament floating nets, with 279 mm (11") stretched mesh, 12 and 16 meshes deep and 50 m in length. The nets were set in shallow water (up to 8 m) and were anchored to shore on one end and to the bottom on the other.

Captured seals were manually restrained, and a satellite-linked Platform Transmitter Terminal (PTT; Wildlife Computers Splash; dimensions: 5 × 6 cm; weight: 65 g in air) was glued to the fur mid-dorsally between the scapulae using fast-setting two-component epoxy glue, after drying and cleaning the fur with acetone. Sex, weight, girth, and length (nose to tail, with belly down) were recorded. Age class was estimated by counting annuli on the claws of the forelimbs, observing each animal closely, and taking morphometric measurements (i.e., length and weight) in the field. The determined age class for each seal was further validated by comparing their length and weight measurements to measurements taken previously for aged Labrador ringed seals (Supporting Information (SI) Table S1). A blubber biopsy (6 mm diameter), consisting of the entire blubber column down to muscle was sampled from each seal prior to release and kept frozen at −80 °C until contaminant analyses. Fur samples were obtained from 10 of the 13 seals for stable isotope analysis.

The Nunatsiavut Government, Nunatsiavut Health and Environment Review Committee and The Animal Use Protocol (AUP) administered by Fisheries and Ocean Canada approved all animal-handling and sampling procedures.

Transmitters. PTTs were programmed to send up to 250 transmissions per day. The transmission repetition rate was 45 s in the water and 90 s when the seal was hauled out. Location data was collected using the ARGOS system (System Argos, Toulouse, France). Seal locations were estimated following uplinks when the PTT communicated with ARGOS satellites while the seal was at the surface. PTTs are also equipped with pressure transducers that record depth every 10 s whenever the instrument was wet. Dives were defined as any excursion from the surface to depths exceeding 2 m. Water depth at each location was obtained from 1°-resolution General Bathymetric Chart of the Oceans (GEBCO) data.[30]

Home Range and Core Areas. We fit the two-state "switching" state space model described by Jonsen et al.[31] to the time series of each seal's ARGOS locations to account for observation error and infer behavioral changes along each track. The model was fit using a 12 h time step to estimate locations at regular intervals, given the autocorrelation and turning angles between consecutive locations.

The modeled regular time series was used to study spatial distribution by calculating utilization distribution (UD) maps.

UDs were estimated using a Brownian bridge kernel method,[32] implemented in R-package adehabitatHR,[33] which requires specification of two smoothing parameters: one for uncertainty in displacement distance between successive locations per unit time, and another for uncertainty in location estimates. We determined the former (30 m) using a maximum likelihood method,[32] and the latter (12 km) from published estimates.[34] This procedure, as opposed to the classical kernel estimator, has the advantage of accounting for nonlinear movement between successive locations. Before estimating the UDs, locations were projected to an Albers Equal Area coordinate system with local central meridians and standard parallels chosen using the "one-sixth rule".[35]

The projected data were used to build a spatial raster composed of 1 × 1 km cells, where the value in each cell indicates the percentage level of home-range that it belongs to. We estimated home range (95%) and core area (70%) contour intervals, removing areas over land as determined by the Global Self-consistent Hierarchical High-resolution Shorelines database (http://www.ngdc.noaa.gov/mgg/shorelines/gshhs.html). UDs were estimated using this procedure on locations for each individual and for each group of seals (i.e., "local" versus "long-range").

The average ocean depth of the core areas for each seal was defined as the mean depth from the GEBCO grid within each core area.[30] Although there are data gaps using this method, it provides the most extensive coverage for the area.

Index of Benthic Diving. PTTs reported 6 h histograms of dives performed within 0−4 m, 4−10 m, at 10 m intervals from 10 to 100 m, at 40 m intervals from 100 to 220 m, 220−300 m, and deeper than 300 m.

Mean dive depth (x) was calculated as for the i^{th} 6-h period as

$$x_i = \frac{\sum_{j=1}^{n} x_j f_{ij}}{\sum_{j=1}^{n} f_{ij}}$$

Where x_j is the midrange value of the jth histogram bin, f_{ij} is the number of dives in the jth histogram bin and n is the number of bins. The difference between mean dive depth and average ocean depth throughout each individual's track was calculated. The proportion of dives where the difference was smaller than 50 m was used as an index of benthic activity; i.e. if the difference was smaller than 50 m, the seal was considered to be in the benthic layer.

Tissue Analysis. The three 2008 blubber biopsies were analyzed by the Great Lakes Institute for Environmental Research's (GLIER) accredited organic analytical laboratory, Windsor, ON, Canada (Canadian Association for Environmental Analytical Laboratories Accreditation and ISO17025 certified). The other 10 blubber biopsies collected from 2009 to 2011 were analyzed by the Laboratory for Expertise of Aquatic Chemical Analysis (LEACA) at the Institute of Ocean Sciences, Sidney, BC, Canada.

PCBs and organochlorine pesticides (OCPs): α-, β-, γ-hexachlorocyclohexane, α- and γ-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, heptachlor epoxide, *p,p′*-DDD, *p,p′*-DDE, *p,p′*-DDT, dieldrin, hexachlorobenzene (HCB) were measured by GLIER and LEACA using gas chromatography electron capture detection (GC-ECD) and high-resolution gas chromatography/high resolution mass spectrometry (HR-GCMS), respectively. The detailed methodology for extraction, cleanup, and quantification of target analytes has been reported elsewhere [GLIER,[15,36,37] LEACA[38]]. Percent lipid was determined using gravimetric lipid determination by weight of extract method with dichloromethane.

The sample batch submitted for analysis at GLIER consisted of three samples, an in-house reference homogenate tissue, a procedural blank, an in-house reference sample and the external recovery standard (2-ethylhexyl-2,3,4,5-tetrabromobenzoate; TBB). 62 PCB congeners and 14 OCPs were detected in 90% of the samples and were included in the data analysis, in samples where an individual congener was not detected it was replaced with a random number between the detection limit (0.011 to 0.150 ng/g) and zero. Recoveries of individual PCB congeners in the homogenate reference tissue with each batch of samples were within 2 standard deviations from the mean laboratory database value derived from laboratory control charts. Recovery efficiencies for the TBB standard were 95 ± 5.1% (mean ± SE). Procedural blanks (n = 18) were below detection for all PCB congeners and OCPs. The in-house reference sample (n = 1) was within the normal tolerance limits as specified in the GLIER quality control manual.

Sample batches submitted for analysis at LEACA consisted of 10 samples, a procedural blank, certified reference material, and a random duplicate sample. Recovery efficiencies based on the internal standards were 79.8 ± 1.1% (mean ± SE). Procedural blanks (n = 5) had concentrations slightly above detection (<20 pg/g) for 80 ± 11 (mean ± SE) of the 182 PCB congeners measured. Analytical duplicates were within 10% (n = 3).

All samples were recovery-corrected for concentrations of PCBs and OCPs. The smaller congener suite (n = 62) detected by GLIER was used as the primary reference so as to synchronize the two data sets (versus n = 182 for LEACA). An interlaboratory comparison between GLIER and LEACA was completed for 10 ringed seal blubber samples (data not shown), with 47 individual congeners having <30% ($r^2 = 0.96 \pm 0.03$; $p < 0.05$) difference and ∑PCBs having <15% difference between the laboratories. These congeners were therefore chosen for inclusion in the final data set.

Stable Isotope Analysis. Prior to analysis, fur from each individual was washed in a diluted standard detergent, rinsed in Milli-Q water baths, and dried for 24 h at room temperature. Fur samples were homogenized, weighted into tin capsules, and stable carbon and nitrogen isotope ratios were analyzed by continuous flow ion ratio mass spectrometer (CFIR-MS) (Finnigan MAT Delta^{plus}, Thermo Finnigan, San Jose, CA). Stable isotope abundances are expressed in delta (δ) values as the deviation from standards in parts per thousand (‰) using the following equation:

$$\delta_{\text{sample}}‰ = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 1000 \quad (1)$$

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 two standards (bovine muscle (NIST 8414) and an internal lab standard (tilapia fish muscle); n = 65 for each) were <0.16 and <0.08‰ 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. Data pretreatment, methods and rationale for dividing the ringed seals into two groups: "local" and "long-range", are described elsewhere.[15] Briefly, principal components analysis (PCA) was used to elucidate differences in PCB patterns in the 13 tagged ringed seals. PCA ordinations have been widely used to capture differences in PCB patterns for both physical (i.e., sediment) and biological environments.[2,15,39−42] In particular, PCB congener pattern analysis has been used in harbor seal and ringed seal blubber to determine a regional "local" signature versus long-range signature.[2,15] This established approach was applied to the present study to determine which ringed seals had been influenced by the local PCB source compared to seals which had only come into contact with long-range "distant" sources. Samples were standardized to the concentration total before PCA to remove artifacts related to concentration differences between samples.

Linear regression was used to assess the relationship between home range size and the number of relocations. Linear regression was used to assess the relationships between PCA projections and $\sum$PCB/OCP ratios, log K_{ow} values (log of the octanol−water partition coefficient, a proxy for particle affinity), δ^{15}N, and δ^{13}C. Log K_{ow} values for the PCB congeners were taken from literature values.[43] The log K_{ow} for each PCB congener was used to calculate concentration-weighted average log K_{ow} values[44] for each ringed seal according to the following equation:

$$\text{Concentration-weighted average log } K_{ow} \text{ value} = \frac{\sum_{\text{allcongeners}} \text{concentration of individual congener x(log } Kow\text{) value for that congener}}{[\text{total concentration of all congeners}]}$$

To further validate the division of ringed seals into two groups ("local" versus "long-range"), a one-way ANOVA was used to compare the mean of contaminant concentrations, $\sum$PCB/OCP ratios and concentration-weighted average log K_{ow} values for ringed seals in the two groups.[15]

Univariate statistical analyses were performed using the IBM SPSS 20.0 for Windows. Data were log transformed when necessary to meet the normality assumptions for parametric analyses. Contaminant concentrations are expressed on a lipid-weight (lw) basis.

A one-way ANOVA was used to examine the effects of space use and foraging behavior variables and biological variables (length, weight, age class, gender, year) on "local" and "long-range" seals.

■ RESULTS AND DISCUSSION

PCB Patterns and POP Ratios Determine "Local" And "Long-Range" Groupings. A principal components analysis of ringed seal PCB congener profiles identified seals with a heavier (more chlorinated) "local" PCB signature from seals with a lighter (less-chlorinated) "long-range" PCB signature (Figure 1). The first principal component (PC1:45.8%) differentiates ringed seals with a greater proportion of the lighter congeners from seals with a greater proportion of the more heavily chlorinated congeners (Figure 1). The light PCB signature found in seals to the right of the t1-axis is characteristic of a long-range atmospheric transport signal, whereas the heavy PCB signature found in seals to the left of the t1-axis is characteristic of a local source signal. The divergent PCB profiles shown in SI Figure S1 further corroborate these findings and are consistent with the PCB profiles observed in "local" and "long-range" ringed seals from northern Labrador.[15]

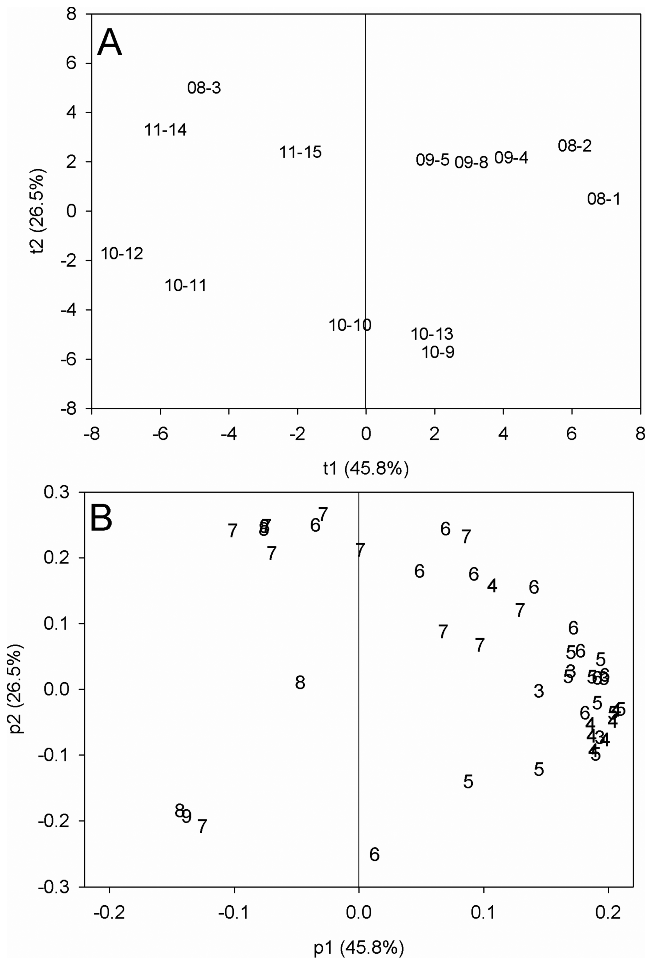

Figure 1. A principal components analysis (PCA) of PCB patterns (47 congeners) in ringed seals reveals that individual seals (numbers depict two digit year of sampling, followed by seal ID) to the left of the scores plot (A) are dominated by heavier congeners (B), consistent with exposure of a local PCB source. Numbers in (B) identify the degree of chlorination of each PCB congener (i.e., number of chlorines per congener).

The $\sum$PCBs/OCPs ratios were correlated ($p < 0.05$) with t1 (the sample scores of the first principal component) for ringed seals (SI Table S3) indicating that ringed seals to the left of the t1-axis with a heavier PCB signature (i.e., local signature) have a higher PCB concentration relative to the five different OCPs. These relationships support previous observations for ringed seals in Labrador,[15] with the local source POP (i.e., PCBs) being elevated relative to OCPs.

The log of total PCBs was correlated with t1 for ringed seals ($r^2 = 0.52$; $p = 0.005$). No relationship was found between the five OCPs ($\sum$DDTs, $\sum$HCHs, $\sum$chlordanes, dieldrin, HCB) and t1 ($p > 0.05$). 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. The logK_{ow} for the PCBs was correlated with p1 (the variable loadings of the first principal component) for ringed seals ($r^2 = 0.58$; $p < 0.001$), suggesting that seals to the right of the t1-axis are exposed to lighter PCB mixtures, consistent with a long-range source, whereas seals to the left of the t1-axis are exposed to more heavily chlorinated congeners (e.g., hepta-, octa-. and nona-PCBs) consistent with a local source. No relationship ($p > 0.05$) was found between $\delta^{15}N$ or $\delta^{13}C$ and t1.

The second principal component (PC2:26.5%) was positively correlated ($r^2 = 0.58$; $p < 0.001$) with $\delta^{15}N$, with the 2010 seals (local and long-range subadult males and females, Figure 1), located below the t2-axis, feeding at a much lower trophic level ($\delta^{15}N = 12.5 \pm 0.5$) than the 2009 and 2011 subadult and adult male seals, located above the t2-axis ($\delta^{15}N = 15.0 \pm 0.5$). The stable isotope profiles in fur reflect the isotopic elements deposited during the annual moult.[45] The low $\delta^{15}N$ levels in the 2010 seals could be due to changes in their foraging ecology in response to unfavorable ice conditions (i.e., below normal extent of coverage and earlier spring breakup) reported for that year.[46] These factors may have influenced the abundance and/or availability of key prey species that year such that seals may have been forced to feed lower on the food chain. For example, the 2010 ice conditions and timing of the spring bloom were factors linked to low capelin abundance that year;[47] other key prey species may have been affected as well. Collectively our PCA results reveal the strong influence of a local PCB source on some of the seals sampled (i.e., seals to the left along the PC1 axis), with trophic level (i.e., $\delta^{15}N$) dietary choices resigned to a secondary role influencing the PCB patterns in both the local and long-range seals. No relationship was found between $\delta^{13}C$ and t2 ($p > 0.05$).

Based on these divergent PCB profiles and PCB/OCP ratios, we hereafter refer to ringed seals to the left of the p1-axis as "local" and seals to the right of the p1-axis as "long-range". Our data indicate that 46% of the tagged ringed seals have been influenced by the local PCB source at Saglek Bay (Figure 1; SI Table S2).

A second PCA, which included PCB concentrations for 84 congeners for LEACA ringed seals ($n = 10$), confirmed the PCB pattern (Figure 1) and group classification (i.e., "local" versus "long-range") for each of the 2009 to 2011 seals (SI Figure S2). Lastly, a PCA, which included the GLIER 2008 harvested seals from Brown et al.[15] and the three GLIER 2008 tagged seals ($n = 3$) from the present study confirmed the group classification for the three 2008 tagged GLIER seals.

Age class, sex, length, weight and year were similar ($p > 0.05$; SI Table S2) between the two groups and did not influence contaminant concentrations and patterns (data not shown).

The geometric mean $\sum$PCB concentrations (1,930 ± 1,640 ng/g lw) in "local" ringed seals were 4-fold higher ($p = 0.02$, SI Table S4) than "long-range" seals (538 ± 89 ng/g lw), suggesting that 75% of the PCB burden of ringed seals sampled originated from the local PCB source at Saglek Bay. OCP concentrations did not differ between "local" and "long-range" seals (SI Table S4). The five $\sum$PCB/OCP contaminant ratios were higher ($p < 0.05$) in "local" ringed seals than in "long-range" seals (SI Table S5). These results are consistent with the observations presented above, with the "local" seals having higher PCB concentrations relative to the OCP concentrations (i.e., higher $\sum$PCB/OCP ratios) than the "long-range" seals. The concentration-weighted average log K_{ow} value for PCBs in "local" ringed seals (7.2 ± 0.04) was higher than "long-range" seals (6.9 ± 0.05, $p = 0.005$). Overall these results are consistent with Brown et al.[15] and further support the grouping of the seals with regard to PCB source. These results also demonstrate that regional contaminant hot spots can have a larger influence on contaminant concentrations than factors (e.g., age and sex) that have been established as drivers of contaminant accumulation.[10,48−50]

Comparison to PCB Concentrations in Ringed Seals in the Canadian Arctic. The PCB and OCP concentrations in the "long-range" seals were generally within the range reported previously for ringed seals in Labrador[15] and the Canadian Arctic.[10,51−54] However, four of the six (67%) "local" ringed seals exceeded the highest PCB values reported for ringed seals in the Canadian Arctic over the past 20 years.[10,52,55] The subadult female (ID: 08−3, 10 700 ng/g lw; SI Table S6) that had the highest $\sum$PCB concentration in the present study, also exceeded the highest values reported in ringed seals in Labrador (9400 ng/g lw, 10-year old male seal[13]) and in a 7-year old female seal (4500 ng/g lw) that was collected sometime between 1989 and 1991, from Inukjuak, Eastern Hudson Bay. The PCB concentration in the "local" subadult female was also 2-fold greater than the "local" adult male (ID: 11−14, 5760 ng/g lw; SI Table S6). This result confirms that this female is prereproductive and has been feeding more in the contaminated area or on locally contaminated prey than the "local" adult male ringed seal. This subadult female spent more time (2-fold) in Saglek Fjord than the "local" adult male, as indicated below, which suggests that this female has a stronger affinity to the fjord, and demonstrates the value of using satellite telemetry to assess contaminant exposure.

While none of our long-range seals exceeded established effects thresholds for marine mammals,[56−58] some of the "local" seals exceeded endocrine and immune thresholds (67%) and reproductive thresholds (17%). Despite the improvements observed in local marine sediments and lower food web-levels,[59] these observations highlight the risks in long-lived ringed seals.

Space-Use and Foraging Behavior Determine the Exposure of Ringed Seals to a Pollution Hotspot. No relationship ($p = 0.34$) was found between home range size and the number of relocations indicating that the computed home range for each seal was not influenced by the amount of time it was tracked. Mean home range was smaller ($p = 0.03$; Table 1; Figure 2) for "local" seals (2281 ± 616 km^2) than for "long-range" seals (11 854 ± 3,642 km^2). "Local" ringed seals maintained a relatively consistent pattern of home range dispersion which was generally within 100 km of Saglek Bay (Figure 2A), indicating that "local" seals were able to secure sufficient prey resources within a small geographic area located within and directly surrounding Saglek Fjord. Home range dispersion of "long-range" seals was much larger (6-fold) (Figure 2B), varied geographically, and displayed little, if any, site fidelity to one area. For example, one of the "long-range" subadult males (ID: 09-5; home range of 16 493 km^2; SI Figure S3) traveled north of Saglek and west into Ungava Bay, where he remained for the duration of his tagging period. A "long-range" subadult female (ID: 10-13; home range = 27 141 km^2) traveled offshore north of Saglek, across Hudson Strait, and remained along the southwestern coastline of Baffin Island (SI Figure S3). While, the other "long-range" subadult female (ID: 10-9; home-range = 16 675 km^2; SI Figure S3) and one of the "long-range" subadult males (ID: 09-8; home-range = 6722 km^2; SI Figure S3) traveled south of Saglek, toward the southern tip of Labrador. One of the "long-range" subadult males (ID: 08-1; home-range = 15 491 km^2; SI Figure S3), remained in Saglek Bay for 1% of his time, then traveled south to Okak Bay and then moved offshore (~150 km) and remained there until the end of his tagging period. The two (28%) remaining "long-range" seals

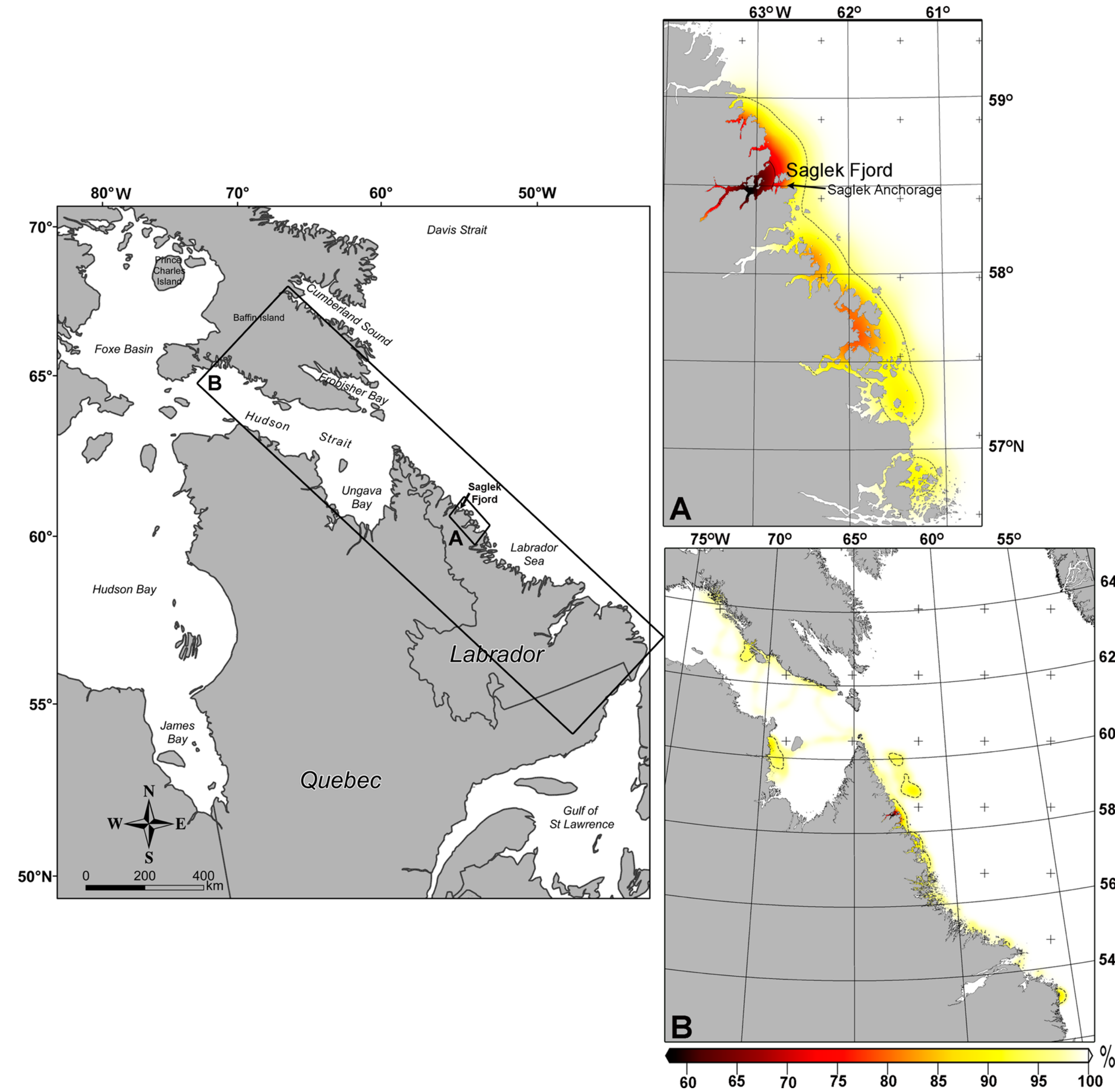

Figure 2. Divergent space-use indices are evident in Labrador ringed seals, as determined by home range and core area. Home range (95%) is defined as the contour intervals for (A) six local and (B) seven long-range ringed seals tagged with satellite transmitters and deployed in Saglek Fjord between 2008 and 2011. Core area (70%) is defined as the contour intervals delineated by the red outline. The outlined boxes in the map of eastern Canada show the area included in the color utilization distribution maps to the right.

(subadult and adult male) displayed similar home ranges (ID: 09-4; home range = 1035 km^2 and ID: 08-2; home range = 420 km^2; Figure S3) and behaviors to that of the locally contaminated seals, whereby they remained closer to shore (<52 km E-W) and to Saglek Fjord (<40 km N–S) (SI Table S2). The similar space-use measures for these two "long-range" seals may reflect the fact that these seals had not yet come into contact with the contaminated prey at Saglek. The adult male (ID: 08-2) displayed extreme site fidelity to the southwestern part of Saglek Fjord, but did not approach or swim through the zone of PCB contaminated sediments in the Saglek Anchorage area. This observation is consistent with large adult ringed seals in Svalbard which displayed extreme site fidelity to areas in front of tidewater glaciers.[23] The minimum distance traveled by these two seals may be the reason for the borderline differences between the two groups for mean longitude (p = 0.047) and mean latitude (p = 0.055) (Table 1).

Mean core areas were smaller (p = 0.03) for "local" seals (487 ± 128 km^2) than "long-range" seals (1912 ± 722; Table 1, Figure 2). All core areas for the "local" seals were located within Saglek Bay and other nearby surrounding inlets (SI Figure S4), consistent with exposure to contaminated prey. In contrast, the core areas for the "long-range" seals were generally located further offshore and at great distances from Saglek Bay (e.g., Ungava Bay, Baffin Island, and central and southern Labrador coast; SI Figure S3).

Time spent in coastal inlets was greater (p = 0.02) for "local" seals (31 ± 6%) than "long-range" seals (9 ± 0.4;

Table 1. Summary of Space Use and Foraging Ecology Information for 13 Ringed Seals Tagged at Saglek Fjord, Labrador, Canada[a]

	local	long-range
home range (km^2)	2,281 ± 616 (1,145–4,341)[b]	11,854 ± 3,642 (420–27,141)
longitude (east–west) (UTM)	102 ± 20 (62–173)[b]	306 ± 88 (34–614)
latitude (north–south) (UTM)	129 ± 25 (71–233)	344 ± 99 (24–710)
average size of core use areas (km^2)	487 ± 128 (245–1,101)[b]	1,912 ± 722 (143–5,102)
average depth of core use areas (m)[c]	51 ± 17 (1–102)	95 ± 33 (15–196)
time spent in coastal inlets (%)	31 ± 6 (14–56)[b]	9 ± 0.4 (1–44)
time spent in Saglek Fjord (%)	17 ± 1 (1–47)	10 ± 8 (0–52)
time spent feeding benthically (%)	49 ± 0.7 (45–50)	47 ± 1.4 (41–50)
$\delta^{15}N$	14.1 ± 0.9 (12.5–17.1)	15.4 ± 0.7 (12.8–17.4)
$\delta^{13}C$	−16.8 ± 0.4 (−18.0-(−)15.7)	−16.6 ± 0.2 (−17.1-(−)15.8)

[a]Values represent mean (standard error). [b]$P \leq 0.05$ compared to long-range seals. [c]Values for two of the seals (long-range seal 0802, local seal 1010) could not be calculated due to the current GEBCO bathymetric data coverage.

Table 1, Figure 2). This observation is consistent with the locations of the "local" seal core areas which were generally located within coastal inlets compared with "long-range" seals which were further offshore (SI Figure S3 and S4).

Although there was no difference ($p > 0.05$) in the average water depth of the core areas between the two groups, the data suggests (Table 1) that the water depth for "local" seals tended to be shallower (51 m; range 1−102 m; $n = 5$) than that of the "long-range" seals (95 m; range 15−196; $n = 6$). The lack of significance between the two groups could be due to the data being underpowered due to a small sample size (values for two of the seals could not be calculated due to the current GEBCO bathymetric data coverage). The observation of "local" seals feeding at shallower depths than the "long-range" seals is consistent with the locations of their core areas (Figure 2); such that "local" seal core areas were located in shallow bay areas and at the heads of the inlets rather than toward the mouths of the inlets and/or further offshore.

Although time spent within Saglek Fjord did not differ ($p > 0.05$) between the two groups, the data suggests (Table 1) that the "local" seals tended to spend more time in Saglek Fjord (17%; range 1−47%; $n = 6$) than that of the "long-range" seals (10%; range 0−52%; $n = 7$). The lack of difference between the two groups is likely due in part to the "long-range" adult male seal (ID: 08−2) that spent (52%) of his time in the inner part of Saglek Fjord. Four (67%) of the "local" tagged ringed seals, including the highly contaminated adult female, surfaced at least once within the zone of PCB contaminated sediments in the Saglek Anchorage area (Figure 2A). The tracks of the other two "local" seals (ID: 10-12 and 11-15; subadult females) suggest that they swam through the contaminated zone, with locations recorded just west and east of Saglek Anchorage. Only two (29%) of the "long-range" seals (ID: 09-5 and 10-13) were recorded to have surfaced within the Saglek Anchorage area. The remaining five (79%) "long-range" seals left Saglek Fjord via the north entrance (Figure 2A).

No differences ($p > 0.05$) in mean time spent feeding benthically and stable isotope ratios ($\delta^{13}C$ and $\delta^{15}N$) were found between the two groups (Table 1). Even though no significant differences were found, it is however still possible that seals from the two groups have distinct preferences for particular prey species which influence their exposure to the local source. However, prior to drawing conclusions on the importance of particular prey species in delivering local contaminants to ringed seals, more research is needed on feeding ecology, using such methods as fatty acid analysis.[60] Furthermore, stable isotope measurements ($\delta^{15}N$) obtained from fur reflect the isotopic elements deposited during the annual moult (preceding spring), which is prior to when the animal was tagged.

These results establish that space-use, as defined by home range, core area, and time spent in coastal inlets is an important determinant of PCB accumulation in ringed seals. The "local" seals tend to have a smaller home range and displayed a strong preference and/or fidelity to a number of core areas located mostly within inlets both within and directly surrounding Saglek Fjord. In contrast, the "long-range" seals had a larger home range with a more dispersed offshore distribution and displayed little, if any, site fidelity to one area.

Strong site fidelity has been previously observed in ringed seals. Kelly et al.[61] observed fidelity to breeding sites among adult Arctic ringed seals. In two different years, Smith and Hammill[21] observed the same ringed seal resting on the ice of a Baffin Island fjord. Krafft et al.[62] marked and subsequently recaptured adult ringed seals (unidentified number) in a fjord on Svalbard in the following year or after many years at sites only a few 100 m from where they were originally marked. Lydersen et al.[23] observed large adult ringed seals showing strong site fidelity in front of tidewater glaciers, compared with subadults which were found to be highly mobile. In the Baltic Sea, Harkonen et al.[22] monitored ringed seal activity over 10 months and found strong spatial fidelity in animals tagged from different areas. Fidelity to small ranges, in general, have numerous biological implications (e.g., foraging ecology, breeding biology, and population structure) for ringed seals,[61] and with respect to ringed seals along the northern Labrador coast, increased exposure to a local PCB source.

The fidelity of "local" seals to Saglek Fjord and its surrounding marine inlets may be related to preferred prey species and/or density of prey in these areas. Saglek Fjord and the marine inlets surrounding Saglek (e.g., Okak Bay) support a diverse and productive benthic and pelagic feeding ground for ringed seals, with a number of diverse habitats including productive nearshore areas dominated by boulders and kelp.[63,64] Sculpin are one of the most common benthic fish species found in these nearshore environments and are also the most locally contaminated fish species in the Saglek Anchorage area.[13,65] It is possible that seals with a strong affinity to these marine inlets are preferentially feeding on sculpin than less contaminated prey. There may also be genetic differences between the two groups of seals, with "local" and "long-range" seals representing two subpopulations with divergent biological and ecological characteristics. Guertin et al.[66,67] used fecal genotyping in river otters to show that time spent in a contaminated harbor influenced POP concentrations.

The satellite telemetry data from the tagged ringed seals provide a forward-looking representation of their movements subsequent to the attachment of tags at capture. In this manner, we can only assume that the biopsy-based PCB, OCP, and stable isotope values are representative of their prior feeding ecology and habitat use. In addition, stable isotope measurements (δ^{15}N) obtained from fur reflect the isotopic elements deposited during the annual moult,[45] whereas the contaminant measurements obtained from blubber reflect total lifetime organic contaminant exposure and accumulation. Despite the temporal and pharmacokinetic differences between these two measurements, the present study shows that the feeding ecology of the seals (i.e., δ^{15}N) influenced PCB profiles, and that the satellite telemetry data were generally consistent with the notion of a distinct point source PCB contribution in "local" seals. It is important to note that the tagging period varies among seals as a function of moult, polar bear predation, or detachment, such that the longer the seal has been tagged, the more confidence we have regarding its space-use behavior.

This first study applying space-use to inform the source apportionment of pollutants in a marine mammal complements the results of stable isotopes and contaminant profiles and ratios. Using this coupled approach, we were able to explain the mode of exposure as a function of habitat use and reaffirm our previous assertion that divergent PCB patterns, PCB/OCP ratios, and PCB concentration-weighted average log K_{ow} values observed in Labrador ringed seals can be attributed to the Saglek PCB source.[15] We suggest that those seals in Labrador that display reduced home range and core area, as well as an increased time spent in coastal inlets, exhibit an increased likelihood of coming into contact with prey contaminated by the local PCB source at Saglek Bay. The present study demonstrates that satellite telemetry can be used to inform the impact of regional or point source pollution exposure on mobile marine animals. We suggest that space-use studies, such as this one, can be used to inform mitigation and monitoring efforts for contaminated sites, and may well be useful in the exploration of other regional stressors.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Variation in PCB profiles as a percentage of total PCB concentrations in "local" and "long-range" ringed seals. Principal components analysis (PCA) of PCB patterns (84 congeners) in ringed seals. Utilization distributions for individual "local" and "long-range" seals. Morphometric and age data for harvested ringed seals collected from Labrador, Canada. Sex, age-class, PCB profile classification, PCB source confirmed by space use measures and transmission period for the seals. Regression analyses for t1 correlated with the ratios of $\sum$PCBs/organochlorine pesticides. Average $\sum$PCB and organochlorine pesticide concentrations in "local" and "long-range" seals. $\sum$PCB/organochlorine pesticides ratios for "local" and "long-range" seals. $\sum$PCB concentrations, δ^{15}N and δ^{13}C ratios for the 13 seals. This material is available free of charge via the Internet at http://pubs.acs.org/.

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Notes

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