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Local contamination, and not feeding preferences, explains elevated PCB concentrations in Labrador ringed seals (*Pusa hispida*)

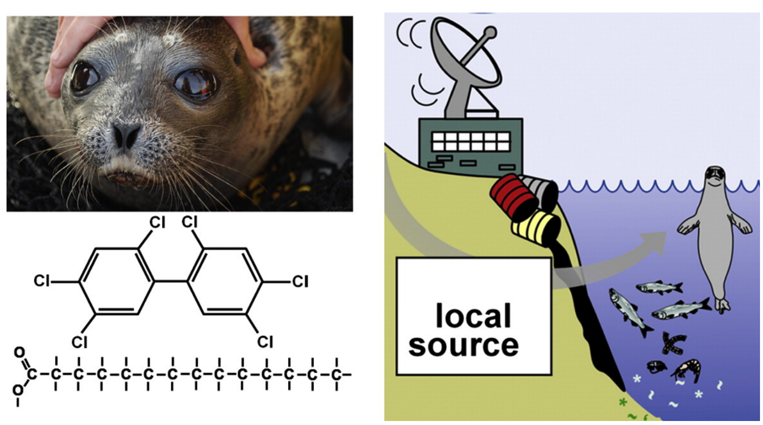
Tanya M. Brown [a,b,c,*], Sara J. Iverson [d], Aaron T. Fisk [e], Robie W. Macdonald [f], Caren C. Helbing [a], Ken J. Reimer [c]

[a] *Department of Biochemistry and Microbiology, P.O. Box 1700, Stn CSC, University of Victoria, Victoria, British Columbia V8W 2Y2, Canada*
[b] *Raincoast Conservation Foundation, P.O. Box 2429, Sidney, British Columbia V8L 3Y3, Canada*
[c] *Environmental Sciences Group, Royal Military College of Canada, P.O. Box 17000 Stn Forces, Kingston, Ontario K7K 7B4, Canada*
[d] *Department of Biology, 1355 Oxford Street, Dalhousie University, Halifax, Nova Scotia, B3H 4R2, Canada*
[e] *Great Lakes Institute of Environmental Research, University of Windsor, 401 Sunset Avenue, Windsor, Ontario N9B 3P4, Canada*
[f] *Institute of Ocean Sciences, Fisheries and Oceans Canada, 9860 West Saanich Road, P.O. Box 6000, Sidney, British Columbia V8L 4B2, Canada*

HIGHLIGHTS

- Fatty acid and PCB profiles in ringed seals reveal the impact of a local PCB spill
- Higher PCBs in some seals could not be attributed to differences in prey selection
- Higher PCBs in these seals were attributed to locally-contaminated prey
- Some ringed seals in Labrador are at risk for PCB-related effects

GRAPHICAL ABSTRACT



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ABSTRACT

Polychlorinated biphenyls (PCBs) in high trophic level species typically reflect the contributions of myriad sources, such that source apportionment is rarely possible. The release of PCBs by a military radar station into Saglek Bay, Labrador contaminated the local marine food web. For instance, while heavier (higher chlorinated) PCB profiles in some ringed seals (*Pusa hispida*) were previously attributed to this local source, differences in feeding preferences among seals could not be ruled out as a contributing factor. Herein, similar fatty acid profiles between those seals with 'local' PCB profiles and those with 'long-range' or background profiles indicate little support for the possibility that differential feeding ecologies underlay the divergent PCB profiles. Ringed seals appeared to feed predominantly on zooplankton (*Mysis oculata* and *Themisto libellula*), followed by the dusky snailfish (*Liparis gibbus*), arctic cod (*Boreogadus saida*), and shorthorn sculpin (*Myoxocephalus scorpius*). Principal components analysis (PCA) and PCB homolog profiles illustrated the extent of contamination of the Saglek food web, which had very different (and much heavier) PCB profiles than those food web members contaminated by 'long-range' sources. Locally contaminated prey had PCB levels that were higher (2- to 544-fold) than prey contaminated by 'long-range' sources and exceeded wildlife consumption guidelines for PCBs. The application of multivariate analyses to two distinct datasets, including PCB congeners ($n = 50$) and fatty acids ($n = 65$),

* Corresponding author at: Department of Biochemistry and Microbiology, University of Victoria, P.O. Box 1700, Stn CSC, Victoria, British Columbia V8W 2Y2, Canada.
E-mail address: tanya@raincoast.org (T.M. Brown).

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afforded the opportunity to clearly distinguish the contribution of locally-released PCBs to a ringed seal food web from those delivered via long-ranged transport. Results from the present study strongly suggest that habitat use rather than differences in prey selection is the primary mechanism explaining the divergent PCB patterns in Labrador ringed seals.

1. Introduction

The often complex feeding habits and extensive movements of marine mammals can present a considerable challenge when attempting to attribute the contribution of specific contaminant inputs to the body burden of a given species. Researchers have used a variety of statistical techniques and study designs (e.g. polychlorinated biphenyl (PCB) profiles and persistent organic pollutant (POP) ratios) to infer the contributions of regional and/or local POP sources (Brown et al., 2014a; Calambokidis and Barlow, 1991; Jarman et al., 1996; Krahn et al., 2007; McKinney et al., 2011; Ross et al., 2004). However, without an understanding of the diet of a given species, it remains difficult to account for the contribution of a point source contaminant to the body burden.

Fatty acids, which represent a large group of molecules that comprise the majority of lipids found in all organisms, have emerged as a powerful tool for the assessment of predator diets (Budge et al., 2006; Iverson et al., 2004). The comparison of fatty acids found in predator fat stores with those found in their prey has allowed both qualitative and quantitative assessments of the spatial and temporal scales of foraging of a given species (Falk-Petersen et al., 2004; Iverson et al., 2004) and has been used to characterize trophic links within and among species (Budge et al., 2002; Iverson et al., 1997; Richoux et al., 2005; Stevens et al., 2004a,b). This inference of diet in predators is possible because many fatty acids transfer from prey to predator adipose tissue with little modification (Budge et al., 2006). Fatty acid analysis has been used to describe dietary processes for contaminants in beluga whales (*Delphinapterus leucas*) (Loseto et al., 2008) and dietary differences which resulted in regional contaminant level differences and temporal contaminant burden declines in polar bears (*Ursus maritimus*) (McKinney et al., 2011, 2013), but has never been used to assess the impacts of point sources in marine ecosystems.

Polychlorinated biphenyls (PCBs) are mixtures of chlorinated hydrocarbons that were banned in the late 1970s in most industrial countries due to their persistent, bioaccumulative, and toxic properties. PCBs are contaminants of concern in arctic food webs, where they can biomagnify to high levels in top predators (Muir et al., 2000). PCB contamination in Arctic marine ecosystems is largely attributed to atmospheric deposition following long-range transport from southern industrial regions (Macdonald et al., 2000; Muir et al., 1999), but local sources within the Arctic (e.g. military sites) have also contaminated local marine food webs (Brown et al., 2009; Kuzyk et al., 2005a). 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 this site (ESG, 1999), along with evidence that PCBs had contaminated the adjacent marine environment (Kuzyk et al., 2005b). Average PCB concentrations in the nearshore marine sediments exceeded the Canadian sediment quality guideline (21.5 ng/g dry weight) by 41-fold and PCB concentrations in benthic invertebrates, bottom-feeding fish, a diving seabird, and some ringed seals were exceptionally high (Kuzyk et al., 2005b). While the elevated PCBs in the benthic-associated food-web closely reflected the concentrations of PCBs in the sediments, determining the contribution of the local source to the body burden of the highly mobile ringed seals proved challenging. 'Heavier' PCB profiles and higher PCB:organochlorine pesticide (OCP) ratios recently provided a 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 (Brown et al., 2014a). This chemocentric approach enabled a classification of seals as either 'local' or 'long-range', reflecting their respective exposure to these two types of sources.

Ringed seals typically feed on a variety of fish, amphipods, euphosiids, mysids, shrimp, bivalves, and cephalopods (Holst et al., 2001; Lowry et al., 1980; McLaren, 1958; Smith, 1987). Spatial and temporal differences have been detected in the diet of ringed seals (Yurkowski et al., in press) and some studies have shown diet variability due to age, sex, and season (Holst et al., 2001; Lowry et al., 1980; Thiemann et al., 2007). Contaminant levels and patterns among prey species can differ due to differences in trophic positions, foraging strategies, and metabolic transformations along with other biological factors (Bang et al., 2001; Hoekstra et al., 2003). Thus, two mechanisms could explain the divergent PCB pattern and increased PCB concentrations in locally versus long-range contaminated ringed seals (Brown et al., 2014a): differences in prey selection between the two groups or feeding on similar prey items that are more contaminated in Saglek Bay.

Herein, we use fatty acid and PCB signature analysis in ringed seals and their prey to determine which mechanism may be involved. In doing so, we evaluate whether contaminant profiles can indeed be used to assign seals to either 'local' or 'long-range' categories. For this, we first determine if there are dietary differences between the previously assigned 'local' and 'long-range' seals. Next, we determine which prey items are most important in the ringed seal diet. Lastly, we compare the PCB patterns in prey species to the patterns observed previously in ringed seals to further investigate the pattern similarities among different species and trophic levels.

2. Materials and methods

2.1. Sample collection

Ringed seal adult male ($n = 20$) and subadult ($n = 22$; male: $n = 6$; female: $n = 16$) blubber samples were obtained from Inuit hunters in four marine inlets (Nachvak Fjord, Saglek Fjord, Okak Bay, and Anaktalak Bay; Fig. 1) along the northern Labrador coast during the fall (September and October) of 2008. Adult female seals were not used in the present study, because females could not be classified into the 'local' and 'long-range' groupings in Brown et al. (2014a). This was due to females transferring individual POPs and congeners at differential rates to their offspring (Borga et al., 2004; Desforges et al., 2012). Sex, length, girth, and blubber thickness (at the sternum) were recorded for each ringed seal. Ages were determined at Matson's Laboratory, Milltown, MT, USA by longitudinal thin sectioning a lower canine tooth and counting annual growth layers in the cementum using a compound microscope and transmitted light. Prey species 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 (Table 1). Most of the fish (Arctic cod (*Boreogadus saida*); dusky snailfish (*Liparis gibbus*); slender eelblenny (*Lumpenus fabricii*); fish doctor (*Gymnelus viridis*) and pelagic invertebrates (Striped pink shrimp (*Pandalus montagui*); greenland shrimp (*Eualus macilentus*); hyperiid amphipod (*Themisto libellula*); *Mysis oculata*) were collected from the 4 inlets from the CCGS *Amundsen* using integrated vertical tows taken with a double Tucker Trawl (200 μm mesh and 500 μm mesh) and from depth-stratified samples taken with a Hydrobios multinet (200 μm mesh). Nearshore fish (shorthorn sculpin (*Myoxocephalus scorpius*); rock cod (*Gadus ogac*); sand lance (*Ammodytes* spp.); daubed shanny (*Leptoclinus maculatus*); capelin (*Mallotus villosus*)) and the benthic invertebrates (Arctic argid (*Argis dentata*); Iceland cockle (*Clinocardium ciliatum*); northern astarte (*Astarte borealis*); Chalky macoma (*Macoma calcarea*)) were collected from the 4 inlets from the long-liner vessel

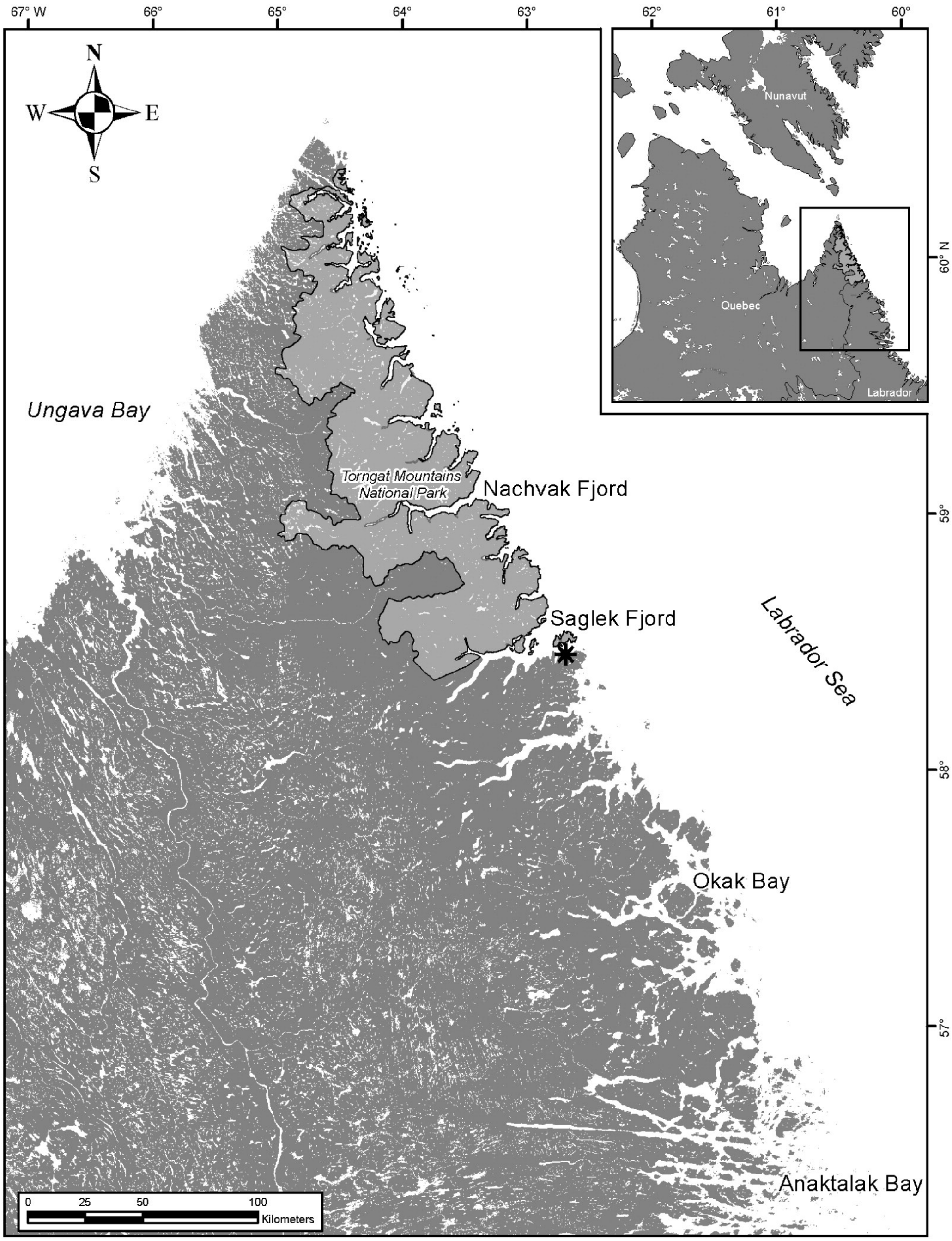

Fig. 1. Map of northern Labrador, Canada, showing the location of the four marine inlets where ringed seals and prey collections were taken. The asterisk shows the location of the former polychlorinated biphenyl (PCB) source and PCB-contaminated sediments at Saglek Bay.

M/V *Whats Happening*. Prey species selection was based on results from stomach content data from the seals in the present study (B. Sjare pers. comm.) and from other ringed seal diet studies (Gjertz and Lydersen, 1986; Labansen et al., 2007; McLaren, 1958; Siegstad et al., 1998; Smith, 1987). 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 Oceans Canada.

2.2. Fatty acid analysis

Lipid was extracted from the inner blubber layer of ringed seals and from whole homogenized fish and invertebrate samples according to Iverson et al. (2001). Whole homogenized fish and invertebrate samples were used to provide the best representation of the prey fatty acid signatures in ringed seal diets (Budge et al., 2002). Fatty acid methyl esters (FAME) were prepared from the extracted lipid using an acidic catalyst (H_2SO_4 in methanol (Thiemann et al., 2004)). Duplicate analyses and identification of FAME were performed using temperature-programmed gas–liquid chromatography according to (Budge et al., 2002, 2006; Iverson et al., 1997, 2001). Samples were analyzed on a VarianCP-3800 gas chromatograph with a flame ionization detector fitted with a flexible fused silica capillary column (30 m × 0.25 mm inner diameter) coated with 50% cyanopropyl polysiloxane (0.25-μm film thickness) (DB-23; Agilent Technologies, Palo Alto, California, USA). Sixty-five fatty acid methyl esters were identified using known standard mixtures, silver nitrate chromatography and mass spectroscopy, and all chromatograms and identifications were individually

Table 1
Latin and common names including abbreviations for prey items evaluated for fatty acid and PCB congener analyses in the present study. The sample size (n), length ($\pm$SD), and sample location sites in Labrador (N = Nachvak; S = Saglek; O = Okak; A = Anaktalak) are indicated. n.a., not applicable.

Latin name	Common name	Abbrev.	n	Length (cm)	Sample location
Fish					
Ammodytes spp.	Sand lance	SDL	3	7.6 ± 1.1	N, S
Boreogadus saida	Arctic cod	ACD	5	5.5 ± 2.9	N, O
Gadus ogac	Rock cod	GDO	9	33.4 ± 5.2	N, S
Gymnelus viridis	Fish doctor	GMV	2	7.6 ± 2.5	S
Leptoclinus maculatus	Daubed shanny	DBD	21	6.5 ± 2.4	N, S, O, A
Liparis gibbus	Dusky snailfish	LPG	3	8.8 ± 0.6	N
Lumpenus fabricii	Slender eelblenny	LPF	8	5.1 ± 0.8	A, N
Mallotus villosus	Capelin	CAP	9	10.9 ± 4.1	N, S
Myoxocephalus scorpius	Shorthorn sculpin	SSC	12	23.5 ± 3.7	N, S, O
Pelagic invertebrates					
Eualus macilentus	Greenland shrimp	ELM	12	n.a.	S, O, A
Mysis oculata	None	MYS	23	n.a.	N, S, O, A
Pandalus montagui	Aesop shrimp	PDM	7	n.a.	N, S, O, A
Themisto libellula	None	TL	28	n.a.	N, S, O, A
Benthic invertebrates					
Argis dentata	Arctic argid	AGD	21	n.a.	N, S, O, A
Astarte borealis	Northern astarte	AB	15	n.a.	N, S, O, A
Clinocardium ciliatum	Iceland cockle	CCC	12	n.a.	N, S, O
Macoma calcarea[a]	Chalky macoma	MC	4	n.a.	S, O

[a] Only analyzed for PCB congeners.

examined for accuracy in identification and integration of peak areas and corrected and reintegrated as necessary. Fatty acid data are expressed as the mass percentage of total fatty acids. Individual fatty acids are referred to by the shorthand nomenclature of carbon-chain length: number of double bonds, and position of the first double bond relative to the terminal methyl group. Mean weight percent ± standard deviations of fatty acids for prey items and adult male and subadult ringed seals are reported in Tables S1 and S2, respectively.

2.3. PCB analysis

Concentrations of 50 PCB congeners were measured in ringed seal prey species (*C. ciliatum*, *E. macilentus*; *P. montagui*; *M. oculata*) by gas chromatography electron capture detection (GC-ECD) by the Great Lakes Institute for Environmental Research's organic analytical laboratory (Canadian Association for Environmental Analytical Laboratories Accreditation and ISO17025 certified; Windsor, ON, Canada). PCB congener data for ringed seal blubber, whole homogenized fish (*L. maculatus*; *G. ogac*; *Ammodytes* spp.; *M. scorpius*) and invertebrate (*A. borealis*; *M. calcarea*) species from Brown et al. (2014a) were used to supplement the food web analysis in the present study. The detailed methodology has been reported elsewhere (Brown et al., 2014a; Drouillard et al., 2004; Lazar et al., 1992). PCB congener concentrations for prey items and adult male and subadult ringed seals are reported in Tables S3 and S4, respectively. For each batch of six samples, an in-house reference homogenate tissue and method blank were analyzed for 50 PCB congeners. For all samples, including homogenate tissue and method blank, an external 1,3,5-tribromobenzene (TBB) recovery standard was added prior to the extraction procedure to quantify extraction efficiency. All PCB congeners were detected in 90% of the samples and were included in the data analysis, in samples where an individual congener was not detected (maximum of three undetectable values per sample) it was replaced with a random number between the detection limit (0.011 to 0.150 ng/g) and zero. Replacing an undetectable value with a random number influences the PCA algorithm less than an arbitrary substitution such as the detection limit or one half of the detection limit (Ross et al., 2004). Recoveries of individual PCB congeners in the homogenate reference tissue with each sample batch run were within 2 standard deviations from the mean laboratory database value derived from laboratory control charts. Recovery efficiencies for the TBB standard were 89 ± 0.9% (mean ± standard error). Procedural method blanks ($n = 18$) were below detection for all PCB congeners. All study samples were recovery corrected for PCB congener concentrations. Hereinafter, $\sum$PCBs refers to the sum of the 50 PCB congeners.

Concentrations of 91 PCB congeners were measured in individual *M. calcarea* ($n = 4$) and *G. ogac* ($n = 7$) by AXYS Analytical Services Ltd, Sidney, BC, Canada to calculate toxic equivalent concentrations (TEQs) to 2,3,7,8-tetrachlorodibenzo-p-dioxin for PCBs for a sub-set of prey items. These species were pre-selected and analyzed for another study in Saglek Bay (unpublished data) and are used in the present study to compare TEQs for PCBs in 'local' and 'long-range' fish and invertebrates species. Samples were analyzed by gas chromatography/mass spectrometry (GC/MS) using AXYS in-house methods (see Brown et al., 2009 for detailed methods). Detection limits for PCB congeners were sample specific and ranged from 0.01 to 1.0 ng/g. Recoveries of 29 PCB congeners from 2 samples of spiked reference materials averaged 96 ± 3.3%. Procedural blanks ($n = 2$) were below detection for all congeners. Analytical duplicates were within 10% ($n = 2$). TEQs to 2,3,7,8-tetrachlorodibenzo-p-dioxin were calculated for PCBs using World Health Organization International toxic equivalent factors for humans and wildlife (Van den Berg et al., 1998).

2.4. Stable isotope analysis and trophic level calculations

Stable nitrogen isotope ratios were measured by Continuous Flow Ion Ratio Mass Spectrometer (CFIR-MS) (Finnigan MAT Delta^{plus}, Thermo Finnigan, San Jose, CA, USA) in fish (*G. ogac*; *Ammodytes* spp.; *M.* scorpius) and invertebrate (*A. borealis*; *C. ciliatum*; *L. maculatus*; *E. macilentus*; *M. calcarea*; *P. montagui*; *Calanus hyperboreus*) species. Stable nitrogen isotope ratios for ringed seal muscle from Brown et al. (2014a) and Yurkowski et al. (in press) were used to supplement the trophic level calculations in the present study. Stable isotope abundances are expressed in delta (δ) values as the deviation from standards in parts per thousand (‰) using the following equation:

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

where R is the ratio of heavy to light isotope ($^{15}N/^{14}N$) in the sample and standard. The nitrogen stable isotope standard was atmospheric nitrogen. Precision was estimated from replicate analyses for two standards (bovine muscle (NIST 8414) and an internal lab standard (tilapia fish muscle); $n = 149$ for each) were <0.2 for $\delta^{15}N$. Accuracy of isotope analysis, estimated from NIST standards (sucrose (NIST 8542) and ammonia sulfate (NIST 8547); $n = 3$ for each) analyzed during the study, was within <0.1‰ of certified $\delta^{15}N$ values.

Trophic levels relative to the copepod *C. hyperboreus*, which we assumed occupied trophic level 2 (i.e. primary herbivore), were determined using equations modified from Hobson et al. (1995). For each individual sample of pelagic and benthic invertebrates, fish, and ringed seals trophic level was determined using the following relationship:

$$TL_{consumer} = 2 + \left(\delta^{15}N_{consumer} - \delta^{15}N_{C.\ hyperboreus}\right)/3.8 \quad (2)$$

where $TL_{consumer}$ is the trophic level of the organism, $\delta^{15}N_{C.\ hyperboreus}$ is equal to 9.4 ± 0.2 (mean ± SE, $\delta^{15}N$ for *C. hyperboreus*), and 3.8 is the isotopic enrichment factor (Hobson et al., 2002).

Trophic magnification factors (TMFs) were determined from the slope (b) of the linear relationship between log10-transformed PCB 153 concentrations (lipid-normalized) and TL (Fisk et al., 2001) using the following relationship:

$$TMF = 10^{b}. \quad (3)$$

2.5. Data analysis

Fatty acid composition in ringed seal blubber can be influenced by age and sex, given potential differences in feeding patterns among demographic groups (Thiemann et al., 2007). To control for these confounding factors, we separated the data into 2 groups for statistical exploration: subadults (<6 yr, male and females combined) and adult males (≥6 yr). Seals 6 yr and older were considered adults (McLaren, 1958; Smith, 1987; Smith et al., 1973) and those under 6 yr were considered subadults. Although 65 fatty acids were identified, only the fatty acids known to transfer from prey to predator (Thiemann et al., 2004) were quantified in the present study. The percent fatty acid values were subjected to centered log ratio transformation (division by the geometric mean of the sample followed by log transformation) prior to multivariate analyses. Multivariate analyses were carried out using Pirouette 4.0 software and the Primer v6 package (PRIMER-E Ltd, Ivybridge, UK). Univariate statistical analyses were performed in SPSS 20.0 for Windows (IBM Corporation, Armonk, NY).

Ringed seal and prey fatty acid profiles were explored using a principal component analysis (PCA) of the covariance matrix. Multivariate ordination techniques such as PCA have been successfully used for qualitative diet analyses (Bradshaw et al., 2003; Budge et al., 2002; Dahl et al., 2000; Iverson et al., 1997; Loseto et al., 2009). This qualitative analysis shows similarities and differences among ringed seal diet profiles and among prey items, which provide insight into their habitat use and feeding ecology. Qualitative analysis is completed on the fatty acid PCA ringed seal and prey plot by examining the positioning of prey in relation to the ringed seals. For example, prey items positioned closer to the ringed seals have similar fatty acid profiles and likely represent an important prey species. Linear regression was used to assess the relationships between PCA projections and biological variables (e.g. length) and $\sum$PCBs.

Fatty acid composition of ringed seal blubber for adult males and subadults was compared between the two PCB source apportionment groupings ('local' versus 'long-range') and across the four sample locations using multidimensional scaling (MDS) and analysis of similarity (ANOSIM) on Bray–Curtis distances (Clarke and Warwick, 1994). Data pretreatment, methods, and rationale for dividing the ringed seals into two source apportionment groupings: 'local' and 'long-range', are described elsewhere (Brown et al., 2014a). A stress value tending towards zero (>0.1) indicates that there is good separation between the groups with high reliability (Clarke and Warwick, 1994).

PCA was used to elucidate differences in PCB patterns in prey species. Samples were standardized to total PCB concentration before multivariate analyses 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 the data set to produce a data set that was unaffected by negative bias or closure (Ross et al., 2004). Data were then autoscaled (scaled to variable mean and standard deviation) to give every variable equal weight before PCA.

Table 2
One-way analysis of similarity (ANOSIM) tests comparing adult male or subadult ringed seal fatty acid compositions across the four marine inlet locations. R-values are presented with *p*-value in parentheses. An asterisk (*) indicates significant difference ($\alpha = 0.05$).

Seal group	Location	Nachvak	Saglek	Okak
Adult male	Saglek	0.155 (0.710)		
	Okak	0.475 (0.036)*	0.772 (0.006)*	
	Anaktalak	0.667 (0.036)*	0.935 (0.022)*	0.750 (0.10)
Subadult	Saglek	0.091 (0.76)		
	Okak	0.274 (0.136)	0.425 (0.005)*	
	Anaktalak	0.055 (0.476)	0.396 (0.008)*	0.234 (0.076)

3. Results and discussion

3.1. Fatty acid composition of ringed seals

The 10 most abundant fatty acids in ringed seal blubber included the saturated fatty acids 14:0, 16:0, and 18:0, the monounsaturates 16:1n7, 18:1n9, 20:1n9, and 22:1n9 and the essential polyunsaturates 20:5n3, 22:5n3, and 22:6n3. Abundant levels of those fatty acids were similar to those found in ringed seals from across the Canadian Arctic (Thiemann et al., 2007), and other pinnipeds (Iverson et al., 1997) and marine mammals (Dahl et al., 2000; Loseto et al., 2008). Age did not differ ($p > 0.05$) between 'local' and 'long-range' adult males and subadult ringed seals. Sex did not differ ($p = 0.51$) between 'local' and 'long-range' subadult ringed seals. The fatty acid composition of adult male and subadult ringed seal blubber did not differ between 'local' and 'long-range' groupings (ANOSIM, $p > 0.05$), but varied significantly across locations (ANOSIM, $p < 0.001$). MDS and Pairwise ANOSIM tests indicated that adult male ringed seals from the two northern inlets (Saglek and Nachvak: ANOSIM $p = 0.07$) and two southern inlets (Okak and Anaktalak: ANOSIM $p = 0.10$) tended to have the most similar fatty acid signatures, whereas those separated by greater distance (northern inlet versus southern inlet) had more distinct signatures (Fig. 2A, Table 2). Principal components analysis confirmed these findings for adult males with the first principal component (p1: 50.8%) clearly differentiating ringed seals from the northern inlets from ringed seals from the southern inlets (Fig. S1). The fatty acids explaining the northern ringed seal distribution to the right of the score plot included 20:1n11, 20:1n9, 14:0, 22:1n9, 20:1n11. Whereas, the fatty acids explaining the southern ringed seal distribution to the left of the score plot included 16:2n4, 18:3n1, 20:4n6, 22:4n6, 22:6n30 (Fig. S1).

Significant fine-scale variability was also evident in the subadults where seals from Saglek had a distinct signature from seals from the two southern inlets (Okak and Anaktalak) (Fig. 2B, Table 2). The fatty acids contributing most to the Saglek subadult ringed seals included 22:1n7, 20:1n7, 20:1n11, 22:1n9, 20:1n9 (Fig. S1). These results are consistent with observations in ringed seals from other areas across the Arctic in which fine-scale (<500 km) regional variability (i.e. location) explained the fatty acid signature pattern (Thiemann et al., 2007). There was no relationship ($p > 0.05$) between either sample

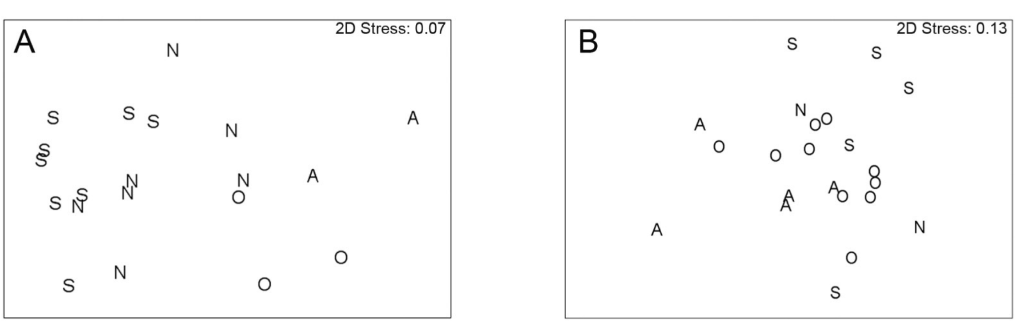

Fig. 2. Multidimensional scaling (MDS) plot of Bray–Curtis similarities of fatty acid profiles of (A) adult male and (B) subadult ringed seals revealed that location had a significant effect on ringed seal fatty acid signatures at the four sites examined along the northern Labrador coast (see Table 2 for R and *p* values). N = Nachvak; S = Saglek; O = Okak; A = Anaktalak.

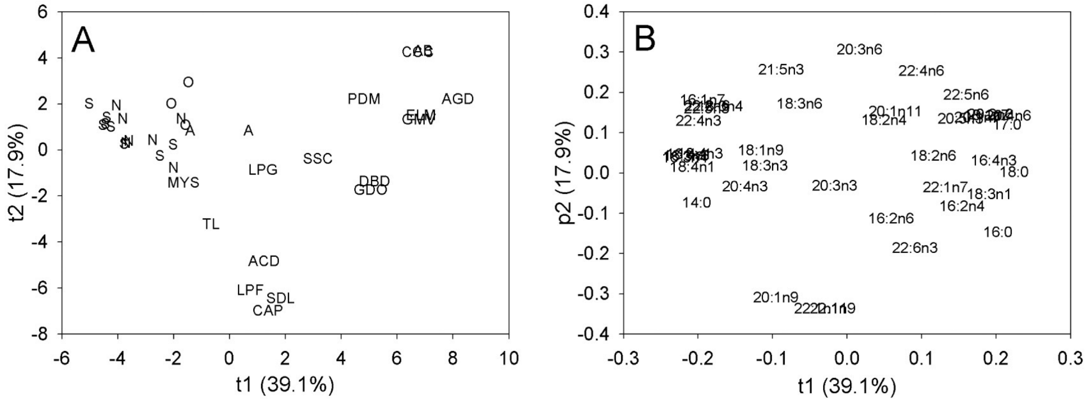

Fig. 3. Principal component analysis (PCA) of the 40 dietary fatty acids measured in adult male ringed seals and their prey. The scores plot (A) reveals that seals from four inlets in Labrador (N = Nachvak; S = Saglek; O = Okak; A = Anaktalak) grouped together, with important prey items identified as a function of their proximity to seals. B) Loadings identify those fatty acids that explain seal and prey profiles. Factor loadings of individual fatty acids are identified by their carbon-chain length: number of double bonds, and position of the first double bond relative to the terminal methyl group. Species abbreviations: (AB = *Astarte borealis*; ACD = *Boreogadus saida*; AGD = *Argis dentata*; CCC = *Clinocardium ciliatum*; DBD = *Leptoclinus maculatus*; ELM = *Eualus macilentus*; GDO = *Gadus ogac*; GMV = *Gymnelus viridis*; LPG = *Liparis gibbus*; LPF = *Lumpenus fabricii*; MYS = *Mysis oculata*; PDM = *Pandalus montagui*; SDL = *Ammodytes* spp.; SSC = *Myoxocephalus scorpius*; TL = *Themisto libellula*).

scores of the first (t1) or second (t2) principal component and length for adult male and subadult ringed seals. Other marine mammal studies have detected a relationship between diet and length, possibly due to habitat selection relating to a particular size requirement of the animal (Loseto et al., 2008). There was no relationship ($p > 0.05$) between either t1 or t2 and $\sum$PCBs for adult male and subadult ringed seals. These results are consistent with observations above where locally-contaminated ringed seals showed no dietary differences from 'long-range' seals.

3.2. Ringed seal dietary preference

Fifty-seven percent of the variance in the prey and ringed seal fatty acid profiles was explained by the first two PCA axes (PC1: 39.1%, PC2: 17.9%) (Fig. 3). The ringed seals in the prey PCA (Fig. 3) maintained the same positioning as the ringed seals in the seal PCA (Fig. S1). The placement of the mysid *M. oculata* and the amphipod *T. libellula* close to the cluster of adult male ringed seals suggests strong similarities among their fatty acid profiles (Fig. 3). Arctic cod, dusky snailfish and shorthorn sculpin were the next closest prey items to the seals and were plotted on the positive side of the first PCA axis. Prey items furthest from the ringed seals were the two bivalves (*A. borealis* and *C. ciliatum*), which project together on the top right side of the PCA score plot, and the benthic shrimp *A. dentata*. The subadult ringed seal food web PCA showed a similar distribution to the adult male ringed seal food web PCA (Fig. S2), further revealing that *M. oculata* and *T. libellula* were the most important prey items to ringed seals from coastal Labrador. These findings are consistent with stomach content data (B. Sjare pers. comm.) and stable isotope mixing model and isotopic niche size results (Yurkowski et al., in press) for the same seals analyzed in the present study with one exception being arctic cod appeared to dominate the diet more than the amphipod *T. libellula* and the mysid *M. oculata*. A possible explanation for this exception could be due to the small size (5.5 ± 2.9 cm, Table 1) and young year-class (1–2 years, (Matley et al., 2013)) of arctic cod collected in the present study, such that this year-class (and therefore fatty acid composition) may not have been representative of the year-class the seals were feeding on. Generally, adult arctic cod are distributed deeper in the water column than small, juvenile arctic cod, which tend to dominate the pelagic zone and shallow areas (Falk-Petersen et al., 1986; Lonne and Gulliksen, 1989). The fatty acid results from the present study are consistent with previous studies of subadult ringed seals that were shown to mainly forage on zooplankton (e.g. *T. libellula* and *M. oculata*) (Bradstreet and Cross, 1982; Holst et al., 2001). However, our finding that adult males also showed a strong reliance on zooplankton diverged from previous results showing a dominant preference for arctic cod among adult ringed seals (Bradstreet and Cross, 1982; Holst et al., 2001).

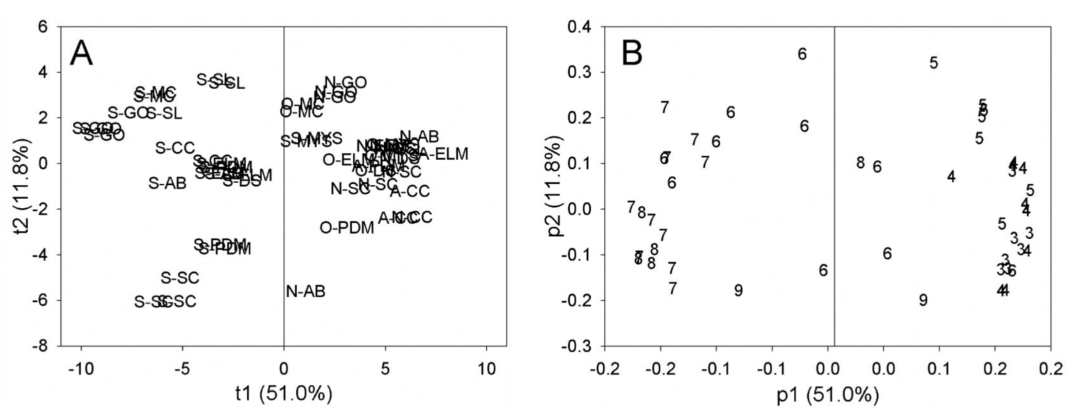

Fig. 4. Principal components analysis (PCA) of the 50 polychlorinated biphenyl (PCB) congeners reveal that prey on the right of the scores plot are dominated by heavier congeners, consistent with exposure to the local Saglek source (A: symbols represent prey from the four inlets in Labrador: N = Nachvak; S = Saglek; O = Okak; A = Anaktalak). Numbers in the loadings plot identify the degree of chlorination of each PCB congener (B). Species abbreviations: (AB = *Astarte borealis*; CC = *Clinocardium ciliatum*; DBD = *Leptoclinus maculatus*; ELM = *Eualus macilentus*; GDO = *Gadus ogac*; MC = *Macoma calcarea*; MYS = *Mysis oculata*; PDM = *Pandalus montagui*; SDL = *Ammodytes* spp.; SSC = *Myoxocephalus scorpius*).

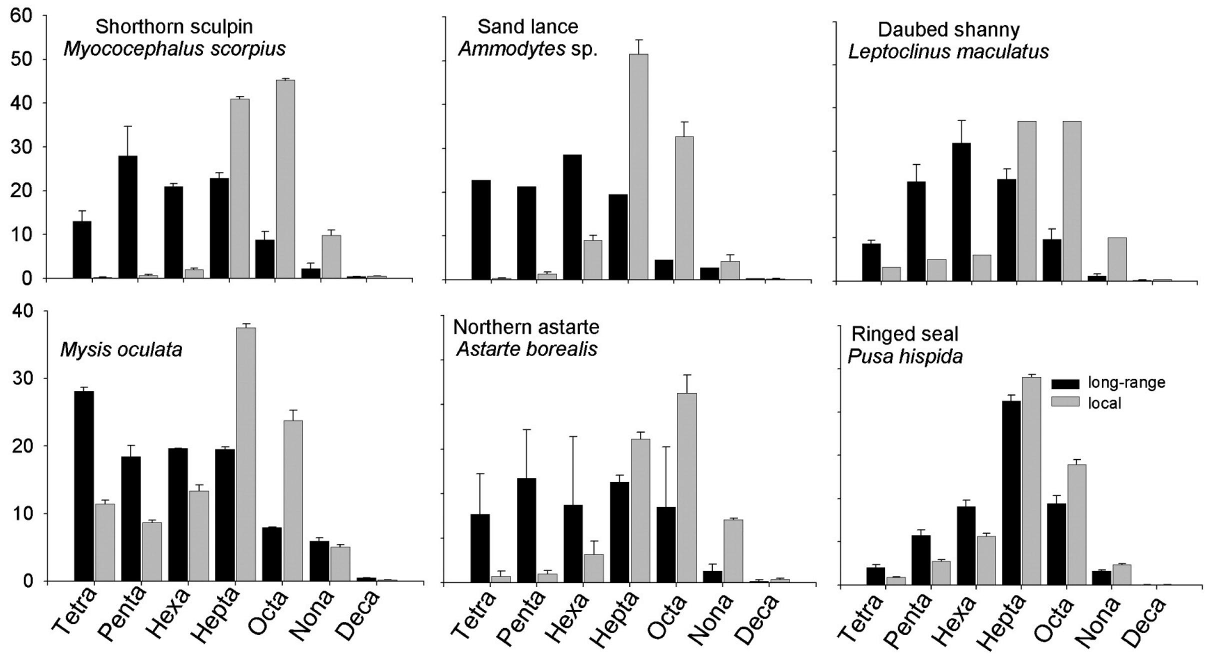

Fig. 5. Polychlorinated biphenyl (PCB) homolog group (i.e. degree of chlorination of congeners) patterns in 'local' (gray bars) and 'long-range' (black bars) adult male ringed seals from Brown et al. (2014a) and their preys from the present study are similar across food web members.

3.3. Prey contaminant patterns

PCB congener patterns reveal strong differences between the Saglek prey items and the reference inlets prey items with the first principal component explaining 51% of the total variance (Fig. 4). The Saglek fjord prey items have a greater proportion of the more heavily chlorinated congeners than the reference inlets prey items which have a greater proportion of the lighter (less-chlorinated) congeners. The heavy PCB signature is typical of a local source signal, in which more chlorinated congeners dominate the composition of PCBs, whereas the light signature is more characteristic of a long-range atmospheric transport signal (Wania and Mackay, 1993), a result of the favoring of lighter, more volatile congeners during volatilization and atmospheric transport. These divergent PCB signatures are consistent with the PCB profiles observed in 'local' and 'long-range' ringed seals from northern Labrador (Brown et al., 2014a) and further corroborate previous conclusions that the local contamination is the dominant source for the 'heavier' signature observed in 'local' ringed seals. Based on the divergent PCB congener profiles, the prey were divided into the 'local' and 'long-range' groups. Prey items to the left of the t1 axis, which were collected in the Saglek Bay area, hereafter will be referred to as 'local', and prey items to the right of the t1 axis, which were collected in the reference inlets, hereafter will be referred to as 'long-range'. While the dominant prey species *M. oculata* collected from Saglek fell just right of the t1 axis (Fig. 4), the results for this species consistently showed a heavier PCB pattern and elevated PCB concentrations when compared with *M. oculata* collected from reference inlets (Fig. 5; Table 3). For this reason, *M. oculata* collected in Saglek Bay will also be referred to as 'local'.

We compared the homolog PCB patterns in five prey species to the patterns observed in adult male ringed seals to further investigate foodweb PCB dynamics. As expected, homolog patterns diverged between prey contaminated by 'local' or 'long-range' PCB sources, with the divergent pattern consistent with observations in the two groups of ringed seals (Fig. 5). Although metabolic processes can deplete lighter PCBs in aquatic organisms (Boon et al., 1994; Desforges et al., 2013; Yunker et al., 2011) they cannot explain the distinct pattern differences observed between 'local' and 'long-range' groups of seals and their prey in our study.

Table 3
Arithmetic means ± SE and ranges [ng/g wet weight] of PCB concentrations in 'local' and 'long-range' prey items. n.a., not applicable.

Latin name	Abbrev	Local	Long-range	Fold difference
Fish				
Ammodytes spp. ($n = 3, 1$)[b]	SDL	115 ± 58 (32–227)[c]	8 n.a.	14
Gadus ogac ($n = 4, 3$)[b]	GDO	625 ± 83 (535–838)[c]	10 ± 2[a] (7–14)[c]	60
Leptoclinus maculatus ($n = 1, 2$)[b]	DBD	72 n.a.	32 ± 2[a] (29–35)[c]	2
Myoxocephalus scorpius ($n = 3, 3$)[b]	SSC	561 ± 149 (409–859)[c]	12 ± 1[a] (10–14)[c]	47
Pelagic invertebrates				
Eualus macilentus ($n = 4, 3$)[b]	ELM	20 ± 3 (14–30)[c]	3 ± 0.5[a] (2–4)[c]	7
Mysis oculata ($n = 2, 2$)[b]	MYS	5 ± 1 (5–5)[c]	3 ± 0.1[a] (3–3)[c]	1.5
Pandalus montagui ($n = 2, 2$)[b]	PDM	22 ± 0.03 (22)[c]	5 ± 3[a] (2–8)[c]	4
Benthic invertebrates				
Astarte borealis ($n = 2, 2$)[b]	AB	239 ± 115 (124–354)[c]	27 ± 9 (18–36)[c]	9
Clinocardium ciliatum ($n = 2, 4$)[b]	CCC	97 ± 58 (39–155)[c]	12 ± 1[a] (9–14)[c]	8
Macoma calcarea ($n = 2, 2$)[b]	MC	1090 ± 4 (1080–1090)[c]	2 ± 0.06[a] (2–2)[c]	544

[a] $p < 0.05$ compared with 'local' group.
[b] Number of samples in the 'local' group followed by the number of samples in the 'long-range' group.
[c] PCB concentration range.

3.4. Prey PCB concentrations

Despite temporal (1998–2006) declines in PCB concentrations in the marine sediments and two indicator species (shorthorn sculpin; *M. scorpius* and black guillemot; *Cepphus grylle*) at the Saglek Anchorage (Brown et al., 2009), PCB concentrations in eight of the ten ringed seal prey species collected in the present study were greater in the 'local'

Table 4
Toxic equivalents (TEQs) to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) for polychlorinated biphenyls (PCBs) in 'local' and 'long-range' prey items. Arithmetic means ± SE in the local prey items exceed the Canadian tissue residue guideline (0.79 ng TEQ kg^{-1} wet weight) for PCBs for the protection of mammalian consumers of aquatic biota, whereas the long-range prey items do not.

Latin name	Abbrev	Local prey items	Long-range prey items
		ng TEQ kg^{-1} wet weight	
Gadus ogac ($n = 4, 3$)[b]	GDO	5.44 ± 2.6	0.10 ± 0.03[a]
Macoma calcarea ($n = 2, 2$)[b]	MC	4.81 ± 0.39	0.07 ± 0.04[a]

[a] $p < 0.05$ compared with 'local' group.
[b] Number of samples in the 'local' group followed by the number of samples in the 'long-range' group.

group compared with the 'long-range' group (Table 3). There was no difference ($p > 0.05$) in average PCB concentrations between the two groups for the sand lance (*Ammodytes* spp.) and the bivalve, *A. borealis*, however this is likely attributable to the small sample size (Table 3). Overall, these results are consistent with previous observations in ringed seals, with the 'local' seals having higher PCB concentrations relative to the 'long-range' seals (Brown et al., 2014a).

The deposit feeding bivalve (*M. calcarea*) from the 'local' group had the highest PCB concentrations compared with all other benthic invertebrates and fish and pelagic species (Table 3). Average PCB concentrations in the 'local' *M. calcarea* were up to 10- and 2-fold greater than concentrations in suspension feeding bivalves (*A. borealis* and *C. ciliatum*) and the most contaminated benthic feeding fish (*M. scorpius* and *G. ogac*), respectively. The elevated levels in the deposit feeding bivalve *M. calcarea* compared with all other species, is likely due to different feeding strategies. Deposit feeders take up PCBs passively through physical contact with porewater (bioconcentration) as well as actively through ingestion of sediment particles (Zhang et al., 2013), whereas suspension feeders take up PCBs via physical contact with porewater as well as by pumping water to obtain particulate organic matter utilized as food (Bjork, 1995). Lower concentrations in the benthic feeding fish were likely due to them feeding on invertebrates located both within the contaminated sediments and in areas with lower concentrations of PCBs. PCB concentrations in the dominant prey species *M. oculata* collected from Saglek was 1.5 fold greater than background levels measured in this species (Table 3). PCB concentrations in the two 'local' pelagic shrimp species (*E. macilentus* and *P. montagui*) were elevated above 'long-range' group background levels of the shrimp and most of the fish and bivalve species (Table 3). Pelagic shrimp accumulate PCBs both from water and from food and in some instances do this more rapidly than fish (Borga et al., 2005). Furthermore, these species tend to respond more quickly than their predators to fluctuations of organochlorine contaminants in the water column (Bettinetti et al., 2012), due largely to a comparatively short life span and the ability to equilibrate with contaminant concentrations in the water column (Larsson, 1989). These observations suggest that the sediment contamination at Saglek continues to impact the benthic-associated biota and early-warning indicator species of the pelagic food web. These results also suggest that the 'local' pelagic food-web, which includes the two dominant ringed seal prey species: *T. libellula* and *M. oculata* has contributed to the elevated PCB levels in 'local' ringed seals.

Within the Saglek Anchorage area, average PCB concentrations in *M. scorpius* have decreased approximately 5-fold ($\Delta_{2007-2011} = 2380$) since the last marine investigation in 2007 (Table 3; (Brown et al., 2009)). Average PCB concentrations in the benthic invertebrates have decreased 10- to 50-fold since the initial marine investigations were conducted in 1998 (Table 3; (Kuzyk et al., 2005b)). Despite these declines and those in the sediment (Brown et al., 2009), biota across all trophic levels continue to exceed background concentrations.

The $\sum$PCB TEQs of two of the 'local' prey items were approximately 55- to 70-fold higher than the 'long-range' prey items (Table 4). These results may indicate a PCB-related induction via the aryl hydrocarbon receptor (AhR) of detoxifying enzymes in the more contaminated seals. These findings are in agreement with a previous study that found a relationship between hepatic *Ahr* mRNA levels and PCB concentrations in ringed seals from this study area (Brown et al., 2014c). TEQs were not calculated for ringed seals in Brown et al., 2014a, because less than half (42%) of the dioxin-like PCBs were analyzed using GC-ECD.

To further assess potential health risks associated with dietary exposure in seals feeding on contaminated prey from Saglek, we compared prey item $\sum$PCB (Table 3) and $\sum$PCB TEQs (Table 4) concentrations with published estimated dietary threshold concentrations. Long-range prey PCB concentrations were below all dietary thresholds. Whereas, 40% of the average 'local' prey (*G. ogac*, *M. scorpius*, *M. calcarea*, *A. borealis*) PCB concentrations were above a PCB dietary threshold of 140 ng/g diet wet weight (wt) for immune and reproductive impairment in seals (Kannan et al., 2000) based on semi-field feeding studies (Boon et al., 1987; Brower et al., 1989; De Swart et al., 1994; Reijnders, 1986; Ross et al., 1995) (Table 3). Three (33%) of the 'local' prey items (*G. ogac*, *M. scorpius*, *M. calcarea*) were above the 250 ng/g wet wt threshold for reproductive toxicity in mink (*Mustela vison*) established in laboratory feeding studies (Kannan et al., 2000). All of the 'local' prey items and two of the 'long-range' prey items (*A. borealis* and *L. maculatus*) exceeded the dietary threshold of 20 ng/g wet wt for

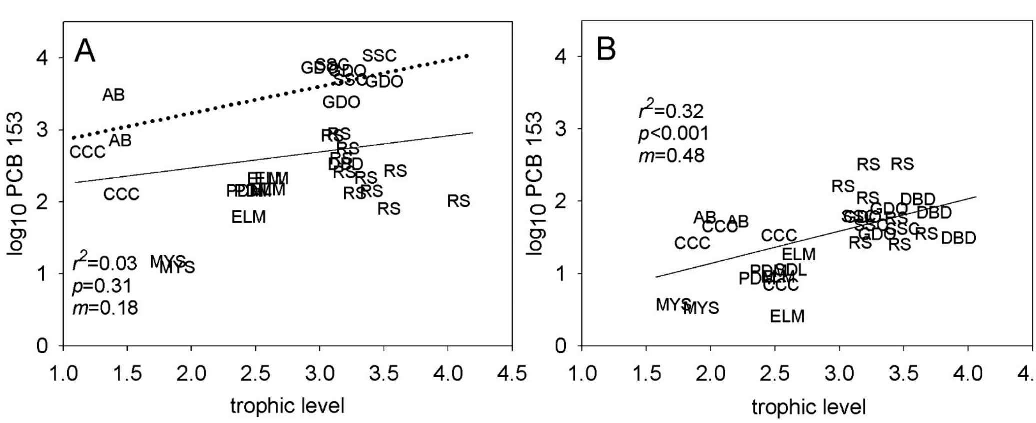

Fig. 6. PCB 153 concentration (ng/g, lipid corrected) trophic level relationships for the local (A) and long-range (B) food web. Solid lines are linear regressions for each group. Using the slope from the 'long-range' group we were able to predict (A, dotted line) PCB 153 concentrations in adult male ringed seals feeding exclusively in the Saglek Anchorage. Species abbreviations: (AB = *Astarte borealis*; CCC = *Clinocardium ciliatum*; DBD = *Leptoclinus maculatus*; ELM = *Eualus macilentus*; GDO = *Gadus ogac*; MC = *Macoma calcarea*; PDM = *Pandalus montagui*; RS = *Pusa hispida*; SDL = *Ammodytes* spp.; SSC = *Myoxocephalus scorpius*).

vitamin A disruption in the European otter (*Lutra lutra*) (Kannan et al., 2000) established in semi-field studies (Leonards et al., 1997; Murk et al., 1998; Smit et al., 1996).

The 'long-range' ∑PCB TEQs for the bivalve *M. calcarea* and the benthic-feeding fish *G. ogac* fell below the Canadian PCB tissue residue guidelines for the protection of mammalian wildlife consumers of aquatic biota (0.79 ng TEQ/kg diet wet wt, CCME, 1999, 2002). Whereas, the 'local' ∑PCB TEQs calculated for these two species exceed this guideline by 6–7-fold (Table 4). These observations substantiate previous findings of adverse effects in some ringed seals from this study area (Brown et al., 2014c).

Trophic magnification factors were calculated for the 'local' and 'long-range' food-webs to evaluate the PCB accumulation in each of the two food webs. Log PCB 153 concentrations increased as trophic level in the 'long-range' food web increased ($r^2 = 0.2$, $p = 0.02$), but not in the 'local' food web ($r^2 = 0.01$, $p = 0.67$; Fig. 6). The ringed seals in the 'local' food web had lower PCB concentrations than the benthic-feeding fish species, *G. ogac* and *M. scorpius*. This observation is consistent with previous results with the benthic associated food-web at Saglek Bay being more contaminated than the locally-contaminated ringed seals (Table 3, (Brown et al., 2014a; Kuzyk et al., 2005b)). These marine mammals have a home range that is far greater than the spatial extent (~10 km^2) of the contamination at Saglek Bay and are therefore feeding on contaminated prey but also on less contaminated 'long-range' prey from elsewhere. These findings are consistent with observations from a recent space-use study which showed 'locally' contaminated ringed seals from Labrador traveling and feeding (2281 km^2) only within the marine inlets located within and directly surrounding Saglek Fjord (Brown et al., 2014b). The TMF calculated for the 'long-range' food web was similar to that observed in other arctic food webs (Borga et al., 2011). Using the slope from the 'long-range' food web and a conversion factor (25%) for PCB 153 to ∑PCBs, we predict that adult male seals that restrict their feeding to Saglek Bay would have an average ∑PCB concentration of approximately 16,300 ng/g lipid wt (Fig. 6). This concentration is similar to those measured in ringed seals from the contaminated Baltic Sea where increased phase I enzyme activity and endocrine effects have been reported (Routti et al., 2008, 2010) and where a history of reproductive and developmental abnormalities exist (Bergman and Olsson, 1985; Helle et al., 1976a,b).

4. Conclusions

The results from the present study strongly suggest that habitat use (i.e., geographic foraging range) rather than differences in prey selection is the primary mechanism explaining the divergent PCB patterns in Labrador ringed seals. These findings are consistent with a previous study that explained the mode of exposure as a function of space use (i.e. habitat use), whereby locally-contaminated seals displayed a strong preference to inlets located both within and directly surrounding Saglek Fjord (Brown et al., 2014b). To our knowledge, this is the first time that fatty acid signatures have been used to determine regional or local source apportionment of a contaminant in a marine mammal. The present study demonstrates that food web tracers, such as fatty acids, and contaminant pattern analysis on prey, can be used to inform the contribution of point source pollution to exposure in mobile marine animals. Furthermore, the present study shows that locally-contaminated prey exceeded wildlife consumption guidelines for PCBs, which further supports the previous finding that ringed seals foraging within the contaminated area are at risk of adverse health effects, not withstanding recent temporal declines in sediment PCB concentrations (Brown et al., 2009; ESG, 2013).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.02.019.

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