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Spatial and temporal trends of persistent organic pollutants and mercury in beluga whales (*Delphinapterus leucas*) from Alaska

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HIGHLIGHTS

► We examined temporal and spatial differences in pollutants in Alaskan beluga whale.
► Mercury, PCBs, DDTs, and chlordanes were the predominant pollutants observed.
► Most POPs and Hg were higher in Chukchi Sea than Cook Inlet belugas, except BFRs.
► Most legacy POPs and Hg were stable, but PBDEs and α-HBCD were increasing.
► Maternal transfer of most POPs was indicated by sex differences & fetus:mother ratios.

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ABSTRACT

Remote locations, such as the Arctic, are often sinks for persistent contaminants which can ultimately bioaccumulate in local wildlife. Assessing temporal contaminant trends in the Arctic is important in understanding whether restrictions on legacy persistent organic pollutants (POPs) have led to concentration declines. Beluga whale (*Delphinapterus leucas*) tissue samples were collected from two subpopulations (Cook Inlet, Alaska and the eastern Chukchi Sea) between 1989 and 2006. Several POPs (polychlorinated biphenyls (PCBs), dichlorodiphenyldichloroethane and related compounds (DDTs), chlordanes, hexachlorocyclohexanes (HCHs), chlorobenzenes, mirex, polybrominated diphenyl ethers (PBDEs) and semi-quantitatively hexabromocyclododecanes (HBCDs)) were measured in 70 blubber samples, and total mercury (Hg) was measured in 67 liver samples from a similar set of individuals. Legacy POPs (PCBs, chlordanes, DDTs, and HCHs) were the predominant organic compound classes in both subpopulations, with median concentrations of 2360 ng/g lipid for Σ_{80}PCBs and 1890 ng/g lipid for Σ_6DDTs. Backward stepwise multiple regressions showed that at least one of the four independent variables (subpopulation, sampling year, sex, and animal length) influenced the POP and Hg concentrations. ΣPCBs, ΣDDTs, Σchlordanes, Σchlorobenzenes, mirex, and Hg were significantly higher in belugas from the eastern Chukchi Sea than from the Cook Inlet ($p \leq 0.0001$). In contrast, Σ_8PBDE and α-HBCD concentrations were significantly lower in belugas from the eastern Chukchi Sea than from the Cook Inlet ($p < 0.0001$). Significant temporal increases in concentrations of Σ_8PBDE and α-HBCD were observed for both subpopulations ($p \leq 0.0003$), and temporal declines were seen for ΣHCHs and Σchlorobenzenes in eastern Chukchi Sea belugas only ($p \leq 0.0107$). All other POP and Hg concentrations were stable, indicating either a lagging response of the Arctic to source reductions or the maintenance of concentrations by unregulated sources. Sex and length also significantly influenced some concentrations, and these findings are discussed.

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1. Introduction

Although relatively isolated, the Arctic has been shown to be a sink for a variety of contaminants. Atmospheric transport is thought to be the predominant mode of delivery of volatile precursors (Wania and Mackay, 1993; Oehme et al., 1996). Nevertheless, oceanic transport, although somewhat patchy, has also been shown to contribute (Oehme, 1991; Barrie et al., 1992). As a result, the Arctic Monitoring and Assessment Programme (AMAP) was established in 1991 as an international effort to assess Arctic status with respect to target contaminants, including persistent organic pollutants (POPs) and heavy metals. AMAP's initial report (1997) documented the ubiquitous presence of POPs throughout the Arctic. Around the same timeframe, the Stockholm Convention on Persistent Organic Pollutants was signed in 2001 to initiate a

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global ban or restriction on production and use of a number of POPs (Rigét et al., 2010). The effectiveness of this treaty can only be realized by investigating temporal trends in levels of these compounds in the environment and associated biota. Since AMAP's and the Stockholm Convention's inceptions, extensive investigations of marine biota in Canadian and European Arctic regions have been conducted, whereas these have been somewhat lacking in the Alaskan Arctic region.

The beluga whale (*Delphinapterus leucas*) is a circumpolar Arctic cetacean that feeds high in the marine food web, with a diet consisting of various fishes, squid, octopus, and shrimp (Becker et al., 2000). Due to their estimated long life-span (>60 yr) (Stewart et al., 2006), marine-based diet, and high lipid reserves, belugas have been shown to accumulate organic and inorganic contaminants, as do many other Arctic top predators, such as polar bears (*Ursus maritimus*), pinnipeds, and many species of cetaceans (Muir et al., 1992; Skaare, 1995; Kannan et al., 2005). While there have been numerous studies focusing on temporal and spatial trends of these contaminants in Canadian and eastern Arctic belugas (Muir et al., 1999; Braune et al., 2005; Stern et al., 2005; Lebeuf et al., 2007; Kelly et al., 2008), there is limited information on these trends in Alaskan Arctic beluga (Reiner et al., 2011).

Alaskan beluga whales can be divided into two genetically isolated populations: the Bering Sea and Cook Inlet. The Bering Sea population consists of four stocks; the eastern Chukchi Sea, Bristol Bay, the eastern Bering Sea, and the Beaufort Sea (Frost and Lowry, 1990; O'Corry-Crowe et al., 1997; Becker et al., 2000). Cook Inlet belugas (permanent residents of Cook Inlet and the area of the Gulf of Alaska immediately outside Cook Inlet) are geographically separated from the Bering Sea population by the Alaskan Peninsula, while the eastern Chukchi Sea belugas reside between the Russian and United States Arctic (Becker et al., 2000). As the Cook Inlet and eastern Chukchi Sea belugas represent two geographically and genetically isolated populations with tissue samples collected and banked from these two stocks over a span of almost two decades (1989 to 2006), their comparison provides an opportunity to investigate spatial as well as temporal trends in contamination in the Alaskan Arctic.

Beluga whales have been hunted by native Inuit peoples throughout the Arctic. Historical estimates of beluga populations in Cook Inlet and the eastern Chukchi Sea are approximately 1300 and 3700 individuals, respectively (Allen and Angliss, 2010). Although native subsistence hunting is not thought to affect the eastern Chukchi Sea population (Allen and Angliss, 2010), a severe decline in the Cook Inlet population number has led to a halt in this practice (Hobbs et al., 2000). In October 2008, the Cook Inlet belugas were placed on the Endangered Species List in an attempt to bolster dwindling numbers. The National Marine Fisheries Service's most recent survey estimates the population at 284, down approximately 20% from the prior year's estimate of 340 (Hobbs et al., 2011). Genetic isolation, potentially leading to inbreeding and infertility, could impede population growth. However, legacy POPs, as well as emerging contaminants (i.e., brominated flame retardants), have been associated with adverse health effects in marine mammals, including decreased reproduction and offspring survivorship (Schwacke et al., 2002; Wells et al., 2005; Hall et al., 2009) and could also be an impediment to population growth.

In this study, Cook Inlet and eastern Chukchi Sea beluga blubber samples previously collected between 1989 and 2006 as part of the Alaska Marine Mammal Tissue Archival Project (AMMTAP) (Becker et al., 1993) were analyzed for POPs [polychlorinated biphenyls (PCBs), dichlorodiphenyl-dichloroethane and related compounds (DDTs), chlordanes, hexachlorocyclohexanes (HCHs), chlorobenzenes, mirex, polybrominated diphenyl ethers (PBDEs), and semi-quantitatively for hexabromocyclododecanes (HBCDs)]. Blubber is the primary site of POP accumulation in cetaceans, comprising >90% of the whole body burden (Isobe et al., 2009; Yordy et al., 2010a). Beluga liver samples that had been similarly collected were also used for the analysis of total mercury (Hg). The Cook Inlet belugas are thought to be exposed to more local anthropogenic sources coming from the surrounding urbanized area of Anchorage, while the eastern Chukchi Sea belugas, often migrating between the Russian and United States Arctic, should be more exposed to contaminant transported by atmospheric and oceanic mechanisms. There are few reported data sets on POPs in marine mammals from this region and having beluga samples from the Chukchi Sea is a unique opportunity to assess the influences of Asian and Russian production of legacy and emerging POPs to this area of the Arctic. In addition, this information will lead to a more comprehensive account of the Arctic's current and predicted environmental status. Therefore, the main objective of this paper is to provide information on the status and trends of POP and mercury (Hg) concentrations in belugas collected from Alaska.

2. Materials and methods

2.1. Sample collection and preparation

As part of AMMTAP, full-depth blubber and liver tissues were collected from tide-stranded or free-ranging beluga whales during native subsistence hunts in Cook Inlet and the eastern Chukchi Sea, Alaska from 1989 to 2006 (Fig. 1; Supplemental information Table S1). Collection procedures followed standard AMMTAP protocols designed to preserve sample integrity and minimize sample contamination (Becker et al., 1991). Following collection, blubber and liver samples were shipped and stored according to standardized protocols in liquid nitrogen vapor-phase freezers (−150°°C) and maintained at the National Institute of Standards and Technology (NIST) Marine Environmental Specimen Bank (MESB) facility at the Hollings Marine Laboratory in Charleston, South Carolina, as a part of the National Marine Mammal Tissue Bank (NMMTB) (Pugh et al., 2007). The frozen whole tissue sample was then cryogenically homogenized and sub-sampled into fresh frozen powder aliquots (~5 g each) also described in Pugh et al. (2007). Previous studies have analyzed samples from a subset of the same beluga whales for POPs (Becker et al., 1995; Becker et al., 2000; Krahn et al., 2004); however, the data presented in the current study were from new analyses of additional subsamples to ensure consistent methodology for the spatial, temporal, sex and length comparisons.

2.2. Analytical methods

2.2.1. Persistent organic pollutants

2.2.1.1. Extraction and cleanup. Beluga blubber samples from Cook Inlet (n = 30 including 3 fetuses) and the eastern Chukchi Sea (n = 40) were extracted as described in Litz et al. (2007). Briefly, blubber (approximately 1.0 g, exact mass known) was mixed with sodium sulfate and transferred to pressurized fluid extraction (PFE) cells. An internal standard solution containing a suite of ^{13}C-labeled PCB congeners, PBDE congeners, HBCD isomers, and organochlorine pesticides, as well as fluorinated PBDEs, was added gravimetrically to all PFE cells, and samples were extracted with dichloromethane (DCM) using PFE. Lipid content (as expressed by total extractable organics) was determined gravimetrically from a subsample of the PFE extract. The remaining extract was cleaned up using size exclusion chromatography and fractionated using solid phase extraction through acidified silica columns as described in Keller et al. (2009). Most POPs eluted in fraction 1, whereas HBCDs eluted in fraction 2.

2.2.1.2. GC/MS analysis. POPs in fraction 1 were determined by gas chromatography mass spectrometry (GC/MS) using a programmable temperature vaporization (PTV) inlet operated in the solvent vent mode and equipped with a 5 m × 0.25 mm Restek Siltek guard column (Bellefonte, PA) connected to a 0.18 mm × 30 m DB-5MS capillary column, 0.18 μm film thickness (Agilent Technologies, Wilmington, DE). The first injection in the electron impact (EI) ionization mode was used for the determination of the majority of the analytes, including

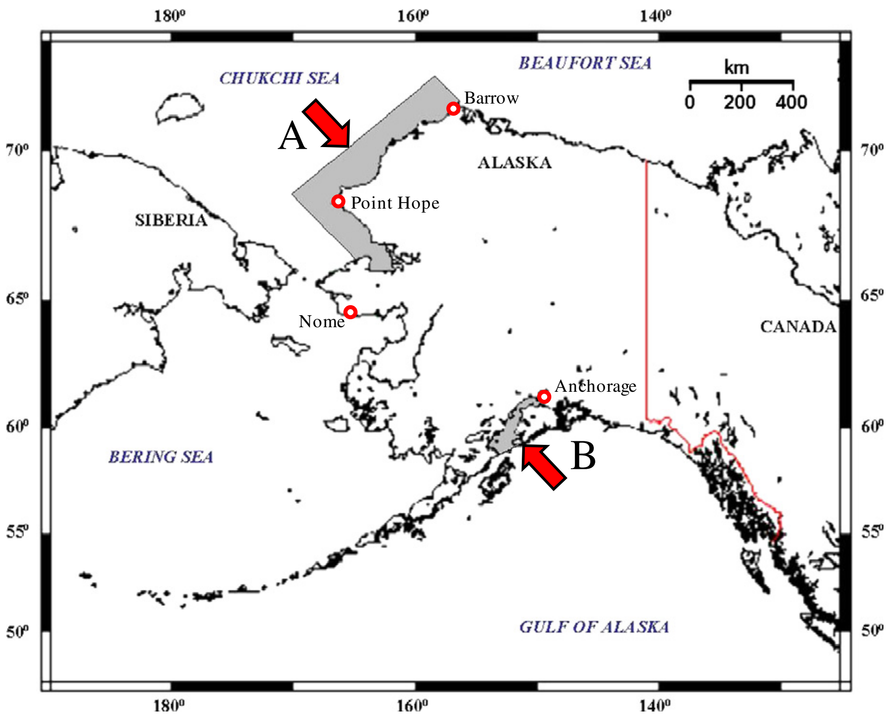

Fig. 1. Map of beluga sample collection sites. "A" represents the eastern Chukchi Sea and "B" represents Cook Inlet, AK. Gray shaded areas are considered the foraging range of each beluga population.

PCBs, PBDEs, DDTs, chlorobenzenes, and mirex. Extracts were further analyzed using the instrument above in the negative chemical ionization (NCI) mode for chlordanes and HCHs. The column used was either a 0.18 mm × 30 m DB-XLB capillary column, 0.18 μm film thickness (Agilent Technologies) or a 0.18 mm × 30 m DB-5MS capillary column, 0.18 μm film thickness (Agilent Technologies). Extracts from 30 adult males (22 from eastern Chukchi Sea and 8 from Cook Inlet) were screened for higher brominated PBDE congeners (PBDEs 138, 156, 183, 191, 181, 190, 203, 205, 206, and 209) using on-column injection onto a 0.18 mm × 10 m DB-5MS capillary column, 0.18 μm film thickness (Agilent) with NCI mode.

2.2.1.3. LC/MS/MS analysis. HBCD isomers (α, β, and γ) in fraction 2 were determined by liquid chromatography tandem mass spectrometry (LC/MS/MS) equipped with a 3.0 mm × 150 mm × 3.5 μm Eclipse Plus C18 column (Agilent Technologies) connected to the API 4000 (Applied Biosystems, Foster City, CA) electrospray ionization (ESI) source. Solvent A was 2.5 mM ammonium acetate in 12.5% water in methanol (volume fraction), and solvent B was acetonitrile.

2.2.2. Mercury

Beluga liver samples from Cook Inlet (n = 27 including 3 fetuses) and the eastern Chukchi Sea (n = 40) were analyzed for total Hg. The concentration of Hg was determined with a direct Hg analyzer DMA 80 (Milestone Scientific, Shelton, CT). The samples were weighed in nickel sample boats, thermally decomposed, catalytically reduced to Hg^0, and then trapped on a gold amalgamation trap. The Hg was then thermally desorbed and the Hg atomic absorption measured at 254 nm.

2.2.3. Quality control

Blanks and NIST Standard Reference Materials (SRMs) were processed alongside blubber and liver samples for quality control. SRM 1945 Organics in Whale Blubber was used as the control sample in blubber analyses, while QC97LH2 Beluga Whale Liver Homogenate and SRM 1946 Lake Superior Fish Tissue were used as control samples in liver analyses. External calibration curves were prepared using SRMs 2261, 2262, 2274, 2275, and 2257 for blubber analyses; and a matrix matched curve of Hg was built with NIST interlaboratory comparison material QC03LH3 Pygmy Sperm Whale Liver Homogenate for liver analyses. POP and Hg concentrations were calculated using the slope and y-intercept of at least a three point calibration curve that bracketed the peak area ratios for POPs and peak areas for Hg observed in the samples. Reporting limits (RLs) were calculated in each sample as the greater of the nanogram amount of the lowest detectable calibration point or (mean + 3 standard deviations [SD]) nanograms in procedural blanks, all divided by extracted sample mass.

2.2.4. Statistical methods

Statistical tests were performed on lipid-normalized data (ng/g lipid) for the sums of PCB congeners (8, 18, 28, 29, 31, 44, 45, 49, 50, 52, 56, 63, 66, 70, 74, 79, 82, 87, 92, 95, 99, 101, 104, 105, 106, 107, 110, 112, 114, 118, 119, 121, 127, 128, 130, 132, 137, 138, 146, 149, 151, 153, 154, 156, 157, 158, 159, 163, 165, 166, 167, 170, 172, 174, 175, 176, 177, 178, 180, 183, 185, 187, 188, 189, 191, 193, 194, 195, 196, 197, 199, 200, 201, 202, 203, 205, 206, 207, 208, 209), PBDE congeners (28, 47, 49, 99, 100, 153, 154, 155), DDT-related compounds (2,4′- and 4,4′-DDE, DDD, and DDT), chlordanes (oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide), HCHs (α, β, and γ), chlorobenzenes (hexachlorobenzene and pentachlorobenzene), mirex, and α-HBCD, and on wet mass data (μg/g wet mass) for Hg. Data determined to be non-normally distributed were log-transformed. The three fetuses (all female from Cook Inlet) were excluded from statistics except for the fetus:mother ratios. Backward stepwise multiple regressions were performed in JMP 10.0 (SAS Institute, Cary, NC) on the concentrations of the nine contaminant classes with the beluga location, year, sex, and length used as the independent variables. Backward stepwise multiple regressions were repeated after separating the data by location so as to assess the influences of year, sex, and length on contaminant concentrations in the two locations independently. When more than one variable was determined to be significant, those variables were crossed and the backward stepwise multiple regressions were re-run, resulting in interaction terms which indicate whether the variables influence or are independent of each other. For α-HBCD statistical analyses, samples containing detectable β-HBCD or γ-HBCD were excluded (3 Cook Inlet males; 1 Chukchi male) in order to avoid the possible bias from laboratory contamination. When concentrations fell below the reporting limit (RL), which happened for 32% of the samples for α-HBCD only, those values were substituted with half the RL for the multiple regressions in JMP, and all significant differences and relationships were verified using statistics appropriate for censored data in the NADA package of R as recommended by Helsel (2005).

3. Results and discussion

All POP classes and Hg were detected in 100% of the samples, except α-HBCD that was detected less frequently, in only 68% of the samples (Table 1). With both subpopulations and sexes combined, concentrations of ΣPOPs in blubber and Hg in liver concentrations ranged from 685 ng/g lipid to 24.9 μg/g lipid (median: 6503 ng/g lipid), and 0.337 to 158 μg/g wet mass (median: 5.51 μg/g wet mass), respectively. ΣPOP concentrations consisted of 37% ΣPCBs (medians shown in parentheses: 2360 ng/g lipid), 31% ΣDDTs (1890 ng/g lipid), 21% Σchlordanes (1300 ng/g lipid), 6% Σchlorobenzenes (339 ng/g lipid), 4% ΣHCHs (138 ng/g lipid), 0.5% mirex (22.7 ng/g lipid), and 0.4% ΣPBDEs (12.7 ng/g lipid). Higher brominated PBDE congeners (138, 156, 183, 191, 181, 190, 203, 205, 206, and 209) were not detected in the 30 adult males selected for screening. α-HBCD was the predominant HBCD isomer detected, but because of high variability in the control sample, SRM 1945, possible laboratory contamination with β-HBCD and γ-HBCD, and low concentrations in the samples (median near 0.5 ng/g lipid), fewer results are reported.

The effects of location, year, sex, and length on contaminant concentrations were examined using backward stepwise multiple regressions (Table 2). All regression models for the nine contaminant classes were statistically significant ($p<0.0001$) when both locations were included. Location and sex were the most consistent variables significantly influencing the contaminant concentrations. Location was significant for all nine contaminants, whereas sex was significantly influencing all contaminant concentrations except ΣHCHs and Hg. Significant temporal trends were noted for ΣHCHs, ΣPBDEs, and α-HBCD (Table 2a), and significant relationships with animal length were apparent for ΣHCHs, mirex, α-HBCD, and Hg. Only six significant interaction terms were observed (e.g., location and year for ΣHCHs), mostly involving location. The multiple regression model for percent lipid was not significant, indicating no location or sex differences and no relationships to year or length (data not shown).

Since location was a strongly influencing variable, additional backward stepwise multiple regressions were assessed with Cook Inlet and eastern Chukchi Sea separated, and fewer significant interaction terms were observed (Table 2b). For these reasons, contaminant concentrations were separated by subpopulation and sex for reporting in Table 1.

3.1. Spatial trends

Significantly higher concentrations of ΣPCBs, ΣDDTs, Σchlordanes, Σchlorobenzenes, ΣHCHs, mirex, and Hg were observed in the eastern Chukchi Sea belugas, while ΣPBDEs and α-HBCD were significantly higher in the Cook Inlet belugas (Tables 1 and 2a). The source of POPs and Hg in the Arctic has typically been explained by two main mechanisms; long-range atmospheric and oceanic transport. However, local release is another potential POP source. The Cook Inlet beluga population is considered to be geographically isolated, staying in Cook Inlet year-round, although they were originally thought to migrate into the Gulf of Alaska when the inlet became heavily iced (Becker et al., 2000). Recent surveys have confirmed the belugas' site fidelity within the inlet (Laidre et al., 2000). The range of Cook Inlet belugas puts them in close proximity to both Anchorage, Alaska and the Kenai Peninsula. Both of these are more urbanized and industrialized areas compared to the remote eastern Chukchi Sea. Therefore, the Cook Inlet belugas could potentially be more affected by anthropogenic sources associated with Upper Cook Inlet, while the more remote eastern Chukchi Sea belugas could be more affected by atmospheric and oceanic transport.

Although beluga migration behavior is not clearly understood, it is thought that some portion of the Bering Sea population (possibly including the eastern Chukchi Sea beluga subpopulation) follow bowhead whale (*Balaena mysticetus*) migration, traversing across the Bering Strait and spending time off the coasts of Russia and Siberia (Burns and Seaman, 1986; Seaman et al., 1986). Neighboring countries, particularly China and Russia, are documented as leading contributors of atmospheric Hg, primarily from fossil fuel combustion and gold mining (AMAP/UNEP, 2008) with the probability of easterly atmospheric transportation of Hg (Strode et al., 2008). Similarly, POPs (i.e., PBDEs, PCBs and organochlorine pesticides (i.e., DDTs and HCHs)) from Russian (Carroll et al., 2008) and Chinese (Tieyu et al., 2005) sources are noted contributors to Arctic contamination, thereby increasing the potential of exposure to these migrating belugas. This may account for higher ΣPCBs, ΣDDTs, Σchlordanes, Σchlorobenzenes, ΣHCHs, mirex, and Hg concentrations in the eastern Chukchi Sea belugas compared to the Cook Inlet belugas.

Table 1
Concentrations of a) POPs (ng/g lipid) in beluga whale blubber samples and b) mercury (μg/g, wet mass) in liver samples.

a.	Cook Inlet				Eastern Chukchi Sea			
	Males (n = 15)		Females (n = 12)		Males (n = 26)		Females (n = 14)	
	Range	Median	Range	Median	Range	Median	Range	Median
% Lipid (blubber)	73.4–91.8	86.3	76.4–94.0	86.3	77.7–91.5	86.6	78.9–89.4	84.4
Analyte								
ΣPCBs	441–4530	1640	260–1960	692	2190–9070	4860	561–5430	1910
ΣDDTs	376–5170	1960	196–1850	626	1350–8240	4310	288–4500	1350
Σchlordanes	156–1390	707	97.1–767	298	1300–6720	3830	242–2980	1150
ΣHCHs	76.6–329	164	53.4–200	92.8	76.0–898	143	89.2–759	221
Σchlorobenzenes	64.5–533	244	47.5–328	138	340–1150	545	71.4–1260	234
Mirex	4.84–40.7	15.2	1.99–14.7	7.85	27.6–156	58.2	12.3–47.5	20.8
ΣPBDEs	6.56–45.6	13.8	7.40–32.0	14.6	4.33–32.2	12.8	1.90–19.4	5.05

b.	Cook Inlet				Eastern Chukchi Sea			
	Males (n = 11)		Females (n = 13)		Males (n = 25)		Females (n = 15)	
	Range	Median	Range	Median	Range	Median	Range	Median
Mercury	2.87–33.7	6.14	0.406–6.68	4.21	0.745–158	16.3	0.337–91.9	6.07

Table 2
Statistical findings using backward stepwise multiple regressions, examining the influences of location, year, sex, and length on contaminant concentrations in Alaskan beluga whales from a) both locations combined and b) each location separated. Significant independent variables were crossed to determine interactions.

a.	Both locations								
	Model					Interactions			
	Overall	Location	Year	Sex	Length	loc × yr	loc × sex	sex × lng	loc × lng
ΣPCBs	**<0.0001**	**<0.0001**	>0.100	**<0.0001**	>0.100	>0.05	>0.05	>0.05	>0.05
ΣDDTs	**<0.0001**	**0.0001**	>0.100	**<0.0001**	>0.100	>0.05	>0.05	>0.05	>0.05
Σchlordanes	**<0.0001**	**<0.0001**	>0.100	**<0.0001**	>0.100	>0.05	>0.05	>0.05	>0.05
ΣHCHs	**<0.0001**	**0.0211**	**0.0138**	>0.100	**0.0005**	**0.0335**	>0.05	>0.05	**<0.0001**
Σchlorobenzenes	**<0.0001**	**<0.0001**	>0.100	**<0.0001**	>0.100	>0.05	>0.05	>0.05	>0.05
Mirex	**<0.0001**	**<0.0001**	>0.100	**0.0003**	**<0.0001**	>0.05	**0.0159**	>0.05	>0.05
ΣPBDEs	**<0.0001**	**<0.0001**	**<0.0001**	**<0.0001**	>0.100	>0.05	>0.05	>0.05	>0.05
α-HBCD	**<0.0001**	**<0.0001**	**<0.0001**	**0.0002**	**0.0283**	**0.0370**	>0.05	**0.0030**	>0.05
Mercury	**<0.0001**	**0.0001**	>0.100	>0.100	**<0.0001**	>0.05	>0.05	>0.05	**<0.0001**

b.	Cook Inlet				Eastern Chukchi Sea				
	Overall	Year	Sex	Length	Overall	Year	Sex	Length	sex × lng
ΣPCBs	**0.0006**	>0.100	**0.0006**	>0.100	**<0.0001**	>0.100	**<0.0001**	>0.100	>0.05
ΣDDTs	**0.0020**	>0.100	**0.0020**	>0.100	**<0.0001**	0.0810	**<0.0001**	>0.100	>0.05
Σchlordanes	**0.0003**	>0.100	**0.0003**	>0.100	**<0.0001**	>0.100	**<0.0001**	>0.100	>0.05
ΣHCHs	**0.0160**	>0.100	**0.0160**	>0.100	**<0.0001**	**0.0001**	>0.100	**<0.0001**	>0.05
Σchlorobenzenes	**0.0039**	>0.100	**0.0039**	>0.100	**0.0001**	**0.0107**	**<0.0001**	**0.0121**	**0.0076**
Mirex	<0.0001	>0.100	>0.100	**<0.0001**	**<0.0001**	>0.100	**<0.0001**	**0.0034**	>0.05
ΣPBDEs	**0.0007**	**0.0003**	0.0822	>0.100	**<0.0001**	**<0.0001**	**<0.0001**	>0.100	>0.05
α-HBCD	**0.0001**	**<0.0001**	**0.0460**	>0.100	**<0.0001**	**<0.0001**	**<0.0001**	**0.0028**	**0.0326**
Mercury	**0.0006**	>0.100	>0.100	**0.0006**	**<0.0001**	>0.100	**0.0157**	**<0.0001**	>0.05

Bolded values indicate statistical significance ($p<0.05$). Values >0.100 indicate an elimination of that variable from the whole model. loc = location; yr = year; lng = length.

Diet and nutrition may also contribute to POP and Hg concentration differences between the Cook Inlet and eastern Chukchi Sea belugas. Although belugas prefer fish, they are opportunistic feeders supplementing their diet with other available prey including a variety of invertebrates (Frost and Lowry, 1990; Becker et al., 2000). Eastern Chukchi Sea belugas frequent areas rich in invertebrates (i.e., clams and crabs) as well as fish (Huntington, 1999). However, close observations of the Cook Inlet belugas indicate a strong preference for seasonally available fish (Huntington, 2000). Regardless, prey availability can become limited throughout the winter with beluga survival relying on utilization of lipid reserves.

An annual thinning of blubber is not uncommon and has been observed in eastern Chukchi Sea belugas (Huntington, 2000). Severe seasonal reductions in blubber thickness have been noted in Cook Inlet belugas, with observations as high as 30 cm thick in the fall and as low as 5–8 cm in the spring (Huntington, 2000). Although utilization of lipid reserves serves an important purpose for these cetaceans, an unfortunate consequence is the concomitant mobilization of lipophilic contaminants (i.e., POPs) closely associated with these lipid reserves. This may result in redistribution into blood and surrounding internal tissues (Debier et al., 2006; Hall et al., 2008; Yordy et al., 2010c), thereby increasing the risk of contaminant-associated health risks (Yordy et al., 2010c). Although speculative, perhaps variable lipid mobilization could contribute to differences in POP concentrations between the two subpopulations.

The brominated flame retardants, ΣPBDE and α-HBCD, were significantly higher in concentration in Cook Inlet than eastern Chukchi Sea belugas. Compared to PCBs with similar physico-chemical properties, PBDEs exhibit lower Arctic contamination potential (de Wit et al., 2010). Lower-brominated congeners tend to degrade atmospherically (Wania and Dugani, 2003) while the less volatile higher-brominated congeners tend to bind to particulates (de Wit et al., 2010). This suggests that exposure is more likely to result from slower oceanic transport or localized inputs. Compared to the eastern Chukchi Sea belugas, the Cook Inlet belugas are both located in lower latitudes (requiring shorter transport of PBDEs) and are in closer proximity to potential local or regional sources year round. One possible source is Anchorage's waste water treatment plant, which serves >200,000 people, with a daily output of approximately 220 million liters. The plant's outfall is located in Knick Arm, a part of Cook Inlet frequented by belugas (Mahoney, personal communication), with many remaining there throughout the summer (Huntington, 2000; Rugh et al., 2000). Studies have detected PBDE congeners in treatment plant effluents (de Boer et al., 2003; Anderson and MacRae, 2006), at concentrations that retain as much as 9% of the original influent concentration (Song et al., 2006). As PBDEs typically bind to sediment and soil, there is the potential to remain in higher concentrations close to the source, although degradation to hydroxylated PBDEs and dioxins has been noted (Steen et al., 2009). Similarly, HBCDs have been detected in sewage influent and effluent with highest concentrations in sludge (Morris et al., 2004). Although the Chukchi Sea belugas are potentially exposed to localized inputs as they migrate between highly urbanized and remote areas, they may not be in direct contact with these sources year round.

3.2. Sex differences and maternal offloading

Based on the multiple regressions separated by subpopulation, contaminants in belugas significantly ($p<0.05$) varied with sex (Table 2b), and when significant, the relationships were always in the expected direction with males having higher concentrations than females (Table 1). This significant sex difference was observed for ΣPCBs, ΣDDTs, Σchlordanes, Σchlorobenzenes, and α-HBCD at both locations; for mirex, ΣPBDEs, and Hg in the eastern Chukchi Sea; and for ΣHCHs in Cook Inlet. This fits the well-documented pattern in cetaceans of organic contaminant transference from mother to offspring, primarily through lactation (Cockcroft et al., 1989; Yordy et al., 2010b).

To further investigate parental offloading, contaminant concentrations from three fetus–mother pairs were compared to examine transplacental transfer (Table 3). Ratios of each fetus–mother pair were calculated by dividing the contaminant concentration of the fetus by the contaminant concentration of the mother. Ratio values >1.0 indicated a high amount of offloading, while values <1.0 indicated less offloading from mother to fetus. Accordingly, ΣHCHs and Σchlorobenzenes demonstrate strong offloading tendencies, with mean ratios of 1.93 and 1.52, respectively. Conversely, mirex and Hg show weak tendencies towards offloading with mean ratios of 0.229 and 0.153, respectively, as did

Table 3
Concentrations of POPs (ng/g lipid) and mercury (Hg) (μg/g, wet mass) in matched beluga whale fetus and mother pairs from Cook Inlet. Ratios indicate contaminant offloading to fetus from mother.

	ΣPCBs	ΣDDTs	Σchlordanes	ΣHCHs	Σchlorobenzenes	Mirex	ΣPBDEs	Hg
Fetus-1	871	874	452	167	222	2.26	15.8	0.434
Mother-1	1140	936	440	120	209	9.45	14.3	2.02
Ratio-1 (fetus to mother)	0.764	0.934	1.027	1.40	1.06	0.240	1.10	0.215
Fetus-2	937	1020	385	317	249	2.46	21.9	0.514
Mother-2	1070	1100	414	140	168	11.4	18.3	5.62
Ratio-2 (fetus to mother)	0.876	0.927	0.930	2.26	1.48	0.216	1.19	0.0915
Fetus-3	264	250	105	113	95.7	1.70	15.2	0.474
Mother-3	391	308	120	53.4	47.5	7.31	15.0	n.m.
Ratio-3 (fetus to mother)	0.675	0.810	0.874	2.12	2.02	0.233	1.02	
Mean ratio	0.772	0.890	0.944	1.93	1.52	0.229	1.11	0.153

n.m. = not measured.

α-HBCD with a mean ratio of 0.447 from two pairs. Beluga (Desforges et al., 2012) and sea lion (Greig et al., 2007) studies have demonstrated similar findings for organochlorine contaminants, concluding significant negative correlations between log K_{ow} and fetus:mother ratios as well as a propensity for transfer of less lipophilic, lower molecular mass congeners to fetuses (Desforges et al., 2012). The low log K_{ow} and molecular mass of HCHs (3.76 on average, 290.8 g/mol) and chlorobenzenes (5.24 on average, 268 g/mol) support these findings. Additionally mirex's weak offloading tendency coupled with its relatively high log K_{ow} and molecular mass (6.89, 545.5 g/mol) further support this rationale. Mercury transfer studies also support this study's findings, demonstrating limited transfer of Hg from mother to fetus in dolphin species (Law et al., 1992; Storelli and Marcotrigiano, 2000); however, strong transfer tendencies have been seen in humans (Santos, et al., 2007) indicating the importance of species-specific investigations. Transfer efficiencies are different for the various mercury forms (organic vs. inorganic), but the current study only measured total Hg.

3.3. Length relationships

Relationships between contaminant concentrations and another life history trait, animal length, were also assessed in the multiple regressions (Table 2b). Mirex and Hg concentrations in both locations significantly increased with animal length, while ΣHCH concentrations in only the eastern Chukchi Sea significantly decreased with length (Fig. 2). The relationships of Σchlorobenzenes and α-HBCD with length were more complicated, emphasized by the significant interaction term between sex and length in the eastern Chukchi Sea (Table 2b). Σchlorobenzene concentrations did not change with length in Cook Inlet; however, they increased with length in males and decreased with length in females in the eastern Chukchi Sea (Fig. 2). Similarly, α-HBCD increased with length in eastern Chukchi Sea males, and no relationship could be determined in the females from this location because they were all (n = 14) below detection. The length relationship with Σchlorobenzenes is supported by its high offloading ratio, allowing larger females, which presumably have had more chances to calve, a greater ability to deplete their Σchlorobenzene burdens. A previous study has shown that select POPs, including ΣHCH, HCB and PBDE congeners 47, 99, 100, 153 and 154 decrease with length in male white-sided dolphins (Tuerk et al., 2005), while another study conducted in Alaskan Arctic bowhead whales reported an increase of select POPs, including *cis*-chlordane, *trans*-nonachlor, ΣPCB, and DDT-related compounds with length in males but not in females (O'Hara et al., 1999). Furthermore, a study in Svalbard belugas noted no significant trends in POPs with length (Andersen et al., 2001). These contradictory findings further emphasize the importance of studying species-specific and location-specific accumulation of POPs as well as the environmental behavior of the pollutants.

3.4. Temporal trends

Temporal trends in contaminant concentrations were similarly identified using the backward stepwise multiple regressions (Table 2b). ΣHCH and Σchlorobenzene concentrations significantly declined from 1989 to 2000 in the eastern Chukchi Sea belugas (Fig. 3), but no significant trend was noted in the Cook Inlet belugas for these compounds. ΣPBDEs and α-HBCD significantly increased in both locations (Fig. 3). Temporal trends of α-HBCD could only be observed in three of the four groupings of beluga whales in Fig. 3, because all samples of the eastern Chukchi Sea females (n = 14) were non-detectable. No other compounds demonstrated a significant temporal trend, suggesting that concentrations of most compounds have remained stable through time.

Current-use compounds (i.e., PBDEs and HBCDs) are of rising concern, yet the status of legacy compounds in the environment should likewise remain at the forefront of monitoring. Although temporal trends for both Arctic biota (Lebeuf et al., 2007; Rigét et al., 2010) and air concentrations (Hung et al., 2010) suggest a general decline in legacy POP concentrations, this study suggests legacy POPs, except HCHs and chlorobenzenes, are in a steady state in the Alaskan Arctic, perhaps due to lag times in atmospheric and oceanic transport. This study demonstrates significant temporal increases in ΣPBDE and α-HBCD concentrations with doubling times in males only for ΣPBDEs of 4.6 years in Cook Inlet and 6.1 years in the Chukchi Sea, and for α-HBCD of 2.4 years in Cook Inlet and 5.0 years in the Chukchi Sea. Similarly, penta- and hexa-BDE concentrations demonstrated a doubling time of 4.7 and 4.3 years, respectively, in ringed seals (*Phoca hispida*) collected from 1981 to 2000 (Ikonomou et al., 2002). Additionally, ΣPBDE concentrations in harbor seals (*Phoca vitulina* Richardsi) collected between 1989 and 1998 had a doubling rate every 1.8 years (She et al., 2002). Increasing HBCD concentrations have been documented for marine mammals outside of the Arctic, including a doubling rate of approximately 2 years in California sea lions from 1993 to 2003 (Stapleton et al., 2006) and exponential increases in harbor porpoises from the United Kingdom from 1994 to 2003 (Law et al., 2006).

Increases of current-use compounds may not be surprising, but there are recent concerns for further release of legacy POPs due to climate change. While sea ice and entrained particulates can contain relatively low levels of contaminants (Muir et al., 1992), in some areas, sea ice can contain equal if not greater levels of PCBs, DDTs, and heavy metals relative to surface sea water (Pfirman et al., 1995). With trends in increased sea ice melt, the potential for legacy compounds to be reintroduced into these Arctic regions should be of concern. Therefore, continued monitoring of this area should be considered.

3.5. Comparison to other beluga studies and implications

Previous studies on POPs in male belugas demonstrate that the Alaskan belugas analyzed here have similar contaminant concentrations

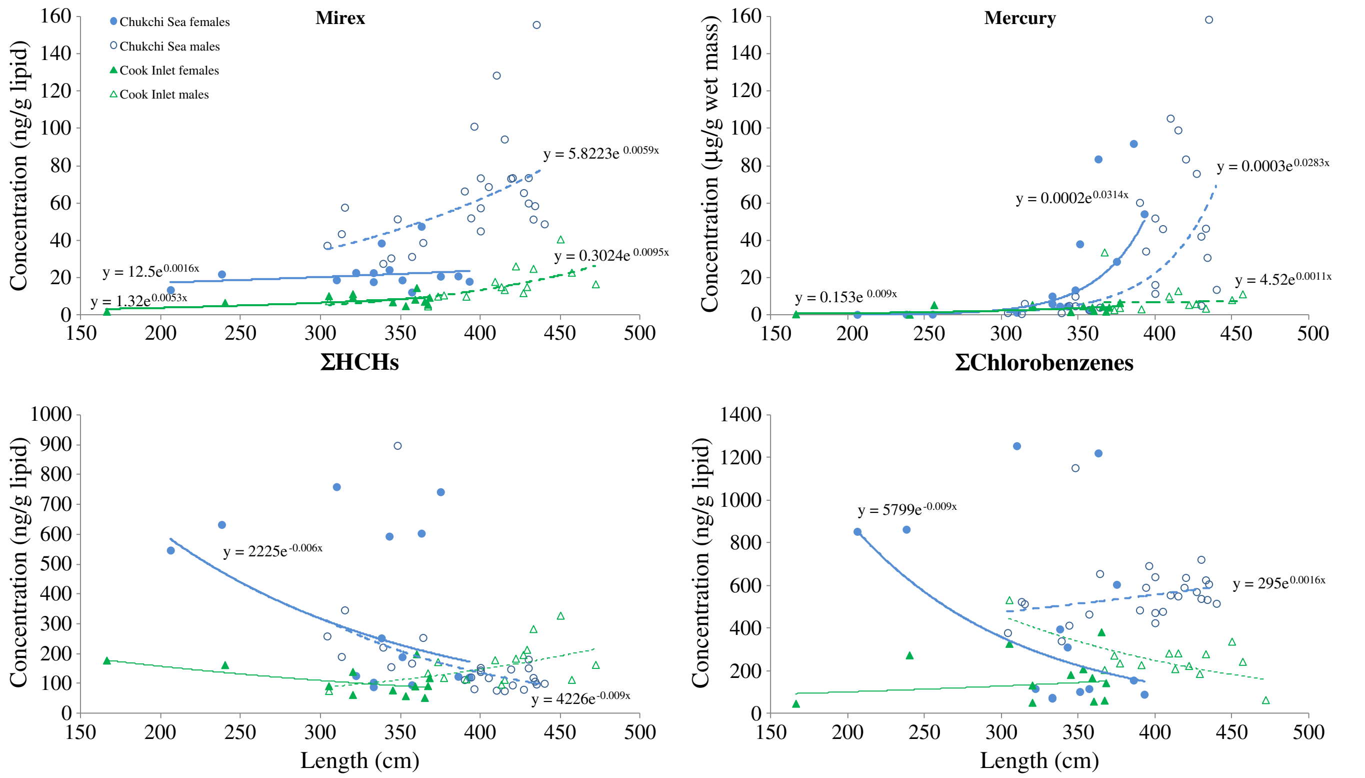

Fig. 2. Relationships between Alaskan beluga whale length and concentrations of mirex, ΣHCHs, and Σchlorobenzenes in blubber and mercury in liver. Blue and green lines are exponential fit trendlines through the Chukchi Sea and Cook Inlet beluga concentrations, respectively. Solid lines are through female data; dashed lines are through male data. Equations are given when length was a significant variable in the backward stepwise multiple regression for that location.

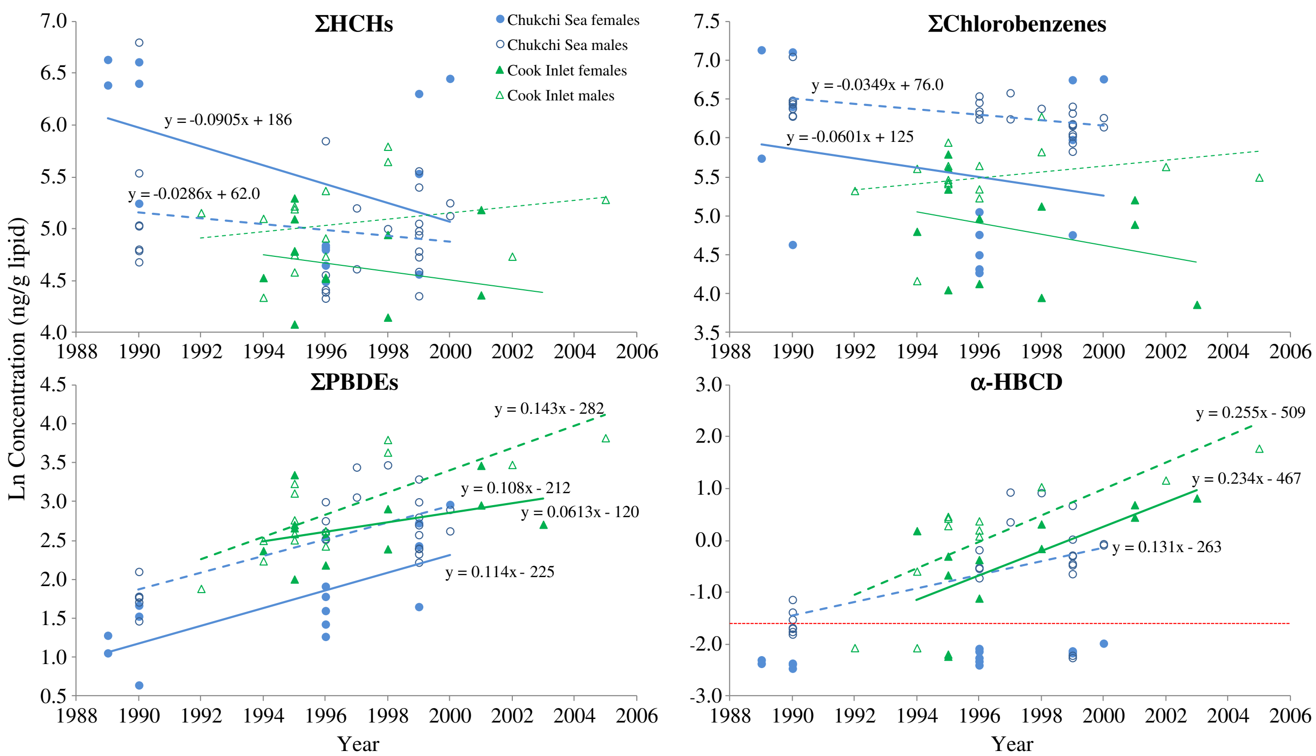

Fig. 3. Temporal trends of natural log, lipid-normalized concentrations of ΣHCHs, Σchlorobenzenes, ΣPBDEs, and α-HBCD in beluga whale blubber from Alaska. Blue and green lines are linear trendlines through the Chukchi Sea and Cook Inlet beluga concentrations, respectively. Solid lines are through female data; dashed lines are through male data. Red line indicates the reporting limit for α-HBCD. Equations are given when year was a significant variable in the backward stepwise multiple regression for that location.

Table 4
Median concentrations (and ranges) of POPs (ng/g lipid) in male beluga blubber samples from various locations.

	Cook Inlet southern Alaska n = 15	Eastern Chukchi Sea northeastern Alaska n = 26	Svalbard Norway n = 10	Eastern Hudson Bay, Canada n = 21	Igloolik Nunavut, Canada n = 22	Sanikiluaq, Hudson Bay, Canada n = 20	Hendrickson Island, Canada n = 10	St. Lawrence River Estuary n = 15	St. Lawrence River Estuary n = 9	St. Lawrence River Estuary n = 15
	This study	This study	Andersen et al. (2001)	Kelly et al. (2008)[a]	Stern et al. (2005)[b]	Stern et al. (2005)[b]	Tomy et al. (2009)[c]	Muir et al. (1996a)[d]	Muir et al. (1996b)[d]	Lebeuf et al. (2004)[d]
Years	1992–2005	1989–2000	1995–1997	1999–2003	1995, 1997	1994, 1998	2007	1987–1990	1993–1994	1997–1999
ΣPCBs	1640 (441–4530)	4860 (2190–9070)	4680 (3198–10,075)		4257	3649		78,900 (8330–412,000)	79,200 (49,300–135,000)	
ΣDDTs	1960 (376–5170)	4310 (1350–8240)	5083 (3272–6770)		5432	2190		81,100 (3360–389,000)	47,600 (20,100–63,700)	
Σchlordanes	707 (156–1390)	3830 (1300–6720)	2566 (2099–6143)		1886	2168		8400 (1430–28,400)	11,200 (8150–15,500)	
ΣHCHs	164 (76.6–329)	143 (76.0–898)	111 (68–510)		213	483		468 (202–900)	345 (275–513)	
Σchlorobenzenes	244 (64.5–533)	545 (340–1150)	430 (324–1423)		712	523		1350 (242–5460)	975 (487–1890)	
Mirex	15.2 (4.84–40.7)	58.2 (27.6–156)						334 (1.3–6800)	1010 (660–1930)	
ΣPBDEs	13.8 (6.56–45.6)	12.8 (4.33–32.2)		34 (13–96)			9.27 ± 1.86			430 (170–780)

[a] Data presented as geometric means and 95% confidence intervals.
[b] Data presented as geometric mean converted with arithmetic mean of % lipid.
[c] Data presented as geometric means and one standard error.
[d] Data presented as means and ranges.

as other high latitude belugas but much lower levels than St. Lawrence River male belugas (Table 4). This comparative finding is true across all legacy POPs and the brominated flame retardants. ΣPBDE concentrations in male St. Lawrence River beluga are 33 times higher than the Alaskan belugas (Lebeuf et al., 2004).

The median α-HBCD concentrations observed in male Alaskan belugas were approximately 1 ng/g lipid, which is similar to the geometric mean of 1.13 ng/g lipid reported for belugas from Hendrickson Island, in northwest Canada (Tomy et al., 2009).

The Stockholm Convention was implemented to substantially reduce the input of deleterious compounds in the environment thereby protecting environmental and human health. Arctic marine mammals present a direct pathway of contaminant exposure to humans through consumption. Belugas, in particular have been a staple in the Alaskan Inuit diet, with blubber plus skin (maktaaq) being regularly consumed (Becker et al., 2000). POPs, including PCBs (Dewailly et al., 1993), DDTs (Tieyu et al., 2005), and PBDEs (Fängström et al., 2005) have been documented in the breast milk of Inuit women who regularly consume whale blubber. Although beluga whale is an important nutritional staple in the Inuit diet, consideration should be used as exposure to these contaminants has been linked to a variety of deleterious toxicological effects (Longnecker et al., 1997; McDonald, 2002).

4. Conclusions

This study demonstrates that even within a relatively small region of the Arctic, there exist significant spatial and temporal differences in potential exposure to various deleterious compounds. Although contaminant exposure sources can be speculated, it is not completely clear whether these differences are likely due to geographic variations in long-range atmospheric transport of contaminants, oceanic transport, localized releases, such as sewage treatment plants, feeding habits and nutrition, or climate changes. Typical sex differences and interesting length relationships were observed in the Alaskan beluga whales. Temporal trends generally indicated stable concentrations of legacy POPs and Hg, except for a few declines in HCHs and chlorobenzenes, and increasing concentrations of the brominated flame retardants, ΣPBDEs and α-HBCD. Although the toxicological implications of the concentrations measured in the blubber of beluga whales are unknown, their potential impact on both beluga whale health and human health still exists and should be considered.

Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Conflict of interest

None.

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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.2013.01.072.

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