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ARTICLE

Temporal trends of brominated and fluorinated contaminants in Canadian Arctic beluga (*Delphinapterus leucas*)[1]

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Abstract: Limited information exists regarding contemporary and historical emissions for many anthropogenic chemicals, especially for contaminants of emerging concern (CECs). This study examined temporal trends of several perfluoroalkyl substances (PFASs), polybrominated diphenyl ethers (PBDEs), and hexabromocyclododecanes (HBCDs) in three beluga whale (*Delphinapterus leucas*) populations from the Canadian Arctic [Hendrickson Island (HI), NT; Pangnirtung (PG), NU; and Sanikiluaq (SQ), NU] collected from 1982 to 2013. The confounding factors of animal size, age, and sex were included in the analyses, but were only significant for some CECs. The strongest temporal resolution was obtained from HI, which showed increasing trends in PBDEs (4.8%/year) and HBCDs (2.9%/year), and decreases in perfluoroalkylcarboxylic acids (PFCAs, −6.0%/year). Concentrations of perfluorooctanesulfonate (PFOS) were relatively stable between 1984 and 2010, increasing substantially in 2011; trends in perfluorooctanesulfonamide (PFOSA) concentrations were not strictly linear over the study period. PBDE, HBCD, and PFCA trends in the HI population were opposite/different to that of many other arctic animals (e.g., polar bears) for the same time period. Trends were inconsistent among locations, suggesting that regional differences in dietary exposure and/or sources may also impact these trends. The effects of climate-change-driven processes on the exposure and distribution of CECs are currently not well understood, highlighting a need for ongoing contaminant monitoring.

Key words: Beaufort Sea, Cumberland Sound, Western Hudson Bay, organic contaminants, Northern Contaminants Program.

Résumé : Les informations existantes sur les émissions actuelles et historiques de plusieurs produits chimiques anthropiques sont éparses, particulièrement les contaminants nouvellement préoccupants (CNP). Dans le cadre de cette étude, on a examiné les tendances temporelles de plusieurs substances perfluoroalkyliques (SPFA), des éthers diphényliques polybromés (PBDE) et des hexabromocyclododécanes (HBCD) chez trois populations de bélugas (*Delphinapterus leucas*) dans l'Arctique canadien (soit à l'Île Hendrickson (IH), NT; à

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Pangnirtung (PG), NU; et à Sanikiluaq (SQ), NU) collectées de 1982 à 2013. Les variables confusionnelles de taille, d'âge et de sexe d'animal ont été comprises dans les analyses, mais elles étaient seulement significatives pour quelques CNP. La résolution temporelle la plus forte a été obtenue de la population de l'IH, laquelle a montré des tendances croissantes en matière de PBDE (4,8%/an) et de HBCD (2,9%/an) et des diminutions des acides perfluoroalkylcarboxyliques (PFCA, –6,0%/an). Les concentrations en perfluorooctane sulfonate (PFOS) étaient relativement stables entre 1984 et 2010, augmentant considérablement en 2011; les tendances des concentrations en perfluorooctane sulfonamide (PFOSA) n'étaient pas strictement linéaires au cours de la période d'étude. Les tendances des PBDE, des HBCD et des PFCA chez la population de l'IH étaient opposées/différentes de celles chez plusieurs autres animaux arctiques (p. ex. les ours polaires) au cours de la même période. Les tendances étaient inconciliables entres les emplacements, suggérant que des différences régionales de l'exposition et (ou) des sources alimentaires puissent aussi influer au niveau de ces tendances. Les effets des processus dictés par le changement climatique sur l'exposition et la répartition des CNP ne sont pas bien compris en ce moment, mettant en évidence le besoin de surveillance continue des contaminants. [Traduit par la Rédaction]

Mots-clés : mer de Beaufort, baie Cumberland, ouest de la baie d'Hudson, contaminants organiques, Programme de lutte contre les contaminants dans le Nord.

1. Introduction

Since the 1980s, there has been a growing concern amongst the public and scientific communities regarding the spread and accumulation of persistent organic pollutants (POPs) in the Canadian Arctic, and the implications on human and ecosystem health (AMAP 1997; CACAR 1997). With the onset of a changing climate, there has been an increased concern regarding the impacts and effects climate change will have on the distribution, fate, and transport of these contaminants within arctic marine ecosystems (Macdonald et al. 2005; Stern et al. 2012; Loseto et al. 2015). Contaminants of emerging concern (CECs) include highly halogenated brominated flame retardants (BFRs) and per- and polyfluoroalkyl substances (PFASs) due to their persistence, bioaccumulation, and biomagnification in wildlife (Braune et al. 2005; Tomy et al. 2009; Butt et al. 2010; Letcher et al. 2010). Many CECs are now regulated under the Stockholm Convention on Persistent Organic Pollutants (http://chm.pops.int/TheConvention/ThePOPs/TheNewPOPs/tabid/2511/Default.aspx), and have been the focus of initiatives such as the Arctic Monitoring and Assessment Programme (AMAP, https://www.amap.no) and the Canadian Northern Contaminants Program (NCP, http://www.science.gc.ca/eic/site/063.nsf/eng/h_7A463DBA.html).

Beluga whales (*Delphinapterus leucas*) are a high trophic level (TL) species and the most abundant odontocetes in the Canadian Arctic (Brodie 1989), and therefore, play a critical role in the Arctic marine ecosystem. Beluga whales are also an important food source for the subsistent lifestyle of Inuit communities (Usher 2002). As such, beluga whales are an ideal bio-indicator species for tracking the entrance of chemicals into the Arctic marine food web. The NCP has worked with indigenous communities since 1991 to examine human exposure to contaminants through traditional/country foods. As part of this program, samples of beluga spanning 20+ years at three locations across the Canadian Arctic have been collected for contaminant analysis. In the Western Arctic, the Eastern Beaufort Sea beluga sampling program based out of Hendrickson Island (HI) in the Northwest Territories (NT) has been highly successful due in large part to the long-term harvest monitoring program established in the Inuvialuit Settlement Region (ISR) in the 1970s, which has been maintained by the local indigenous communities, as well as co-management boards and Federal Departments (Harwood and Smith 2002). This program has since expanded to include measurements of contaminants such as CECs and mercury (Ostertag et al. 2009; Loseto et al. 2015). The other two monitoring

programs are based out of Pangnirtung, Nunavut (PG, NU, Cumberland Sound population) and Sanikiluaq, NU [SQ, Western Hudson Bay (WHB) population].

The trophodynamics of BFRs and PFASs has been previously investigated in the Eastern and Western Canadian Arctic (Tomy et al. 2004, 2008*b*, 2009). PFASs including perfluorocarboxylic acids (PFCAs), perfluorooctanesulfonate (PFOS), and its neutral precursor perfluorooctanesulfonamide (PFOSA), have been shown to biomagnify in top TL marine mammals such as beluga. Furthermore, changes in BFR and PFAS food web accumulation may occur as a result of climate-change-driven processes via alteration in fate and transport (direct) or shifts in the food web (indirect) (Macdonald et al. 2005; Ma et al. 2016). Given the data collected in the beluga monitoring programs, we have the opportunity to investigate temporal trends of CECs spanning 20+ years, and to provide information which may be important in future studies examining the effects of climate change on contaminant trends.

Using the data gathered as part of the NCP, we examined the temporal trends of six PBDEs, three hexabromocyclododecane (HBCD) isomers, and three compound classes of PFASs (PFCAs, PFOS, and PFOSA) in beluga whales sampled from the Western Canadian Arctic from 1984 to 2013. The objective of this study was to discern trends in CEC concentrations in the Eastern Beaufort Sea beluga populations over time, within the context of the confounding variables of size, age, and sex. Temporal trends in beluga sampled in the Cumberland Sound and WHB were also examined and discussed as a point of a comparison between the Eastern and Western Arctic beluga populations for selected years; however, due to more limited sample numbers from these locations, these trends should be interpreted with care. In addition, we briefly discuss current knowledge gaps regarding the implications of and potential links to other environmental factors and climate change, and emphasize the importance of continued monitoring of these regions to provide insight into future trends and the impacts to Canada's northern indigenous communities.

2. Materials and methods

2.1. Chemicals

A complete list of the native- and mass-labelled compounds used in this study can be found in the Supplementary Material.[2] Standard reference materials for BDEs in whale blubber (SRM 1945) were obtained from the National Institute of Standards and Technology (NIST, Gaithersburg, MD). No reference materials for HBCD or PFASs were available at the time the analysis was performed.

2.2. Beluga samples

The whale samples selected for study were stored in the archived repository at the Freshwater Institute in Winnipeg. Beluga liver and blubber samples were collected from traditional subsistence hunts from three populations: Eastern Beaufort Sea population samples were collected at Hendrickson Island (HI, NT; 69°32′N, 133°36′W; 1984–2013), the Cumberland Sound population samples were collected in Pangnirtung (PG, NU; 66°08′N, 65°43′W; 1982–2010), and the WHB population samples were sampled near Sanikiluaq (SQ, NU; 56°32′N, 79°13′W; 2003–2013). Where available, 10 male animals ($n = 10$) were selected for study; males were preferred over females to minimize the confounding factors of female lactational and gestational transfers of CECs from mothers to their offspring (Stern et al. 1994). A complete list of animals and their individual CEC concentrations can be found in Supplementary Table S3[2] in the Compiled Beluga Trend Data.xlsx document in the Supplementary Material.[2] Belugas were measured (total length), aged [by counting

[2]Supplementary material is available with the article through the journal Web site at http://nrcresearchpress.com/doi/suppl/10.1139/as-2017-0044.

growth layer groups (GLG) in the dentine] (Campana et al. 2006), and had their sex determined when possible (by examining the genitalia and assessing for the presence of a penis) to account for the confounding variables of size, age, and sex.

2.3. Extraction, clean-up, and instrumental analysis

Details of extraction and clean-up procedures for the analysis of BFRs in beluga blubber, and PFASs in beluga liver tissues have been previously published (Tomy et al. 2004, 2005, 2008*a*, 2008*b*). BDEs were analyzed by gas chromatography electron capture negative ion (ECNI) low-resolution mass spectrometry (GC–LRMS) and HBCDs and PFASs by high-performance liquid chromatography tandem mass spectrometry (HPLC–MS/MS). Details of the methods used are available in the literature (Tomy et al. 2004, 2005, 2008*b*, 2009). BDEs were quantified using external standard calibration standards. Total BDE congeners are based on the sum of BDE-47, -85, -99, -100, -153, and -154. HBCDs and PFASs were quantified by isotope dilution. Total HBCD is based on the sum of the α-, β-, and γ-diastereoisomers. Total perfluorocarboxylic acid (PFCA) content was based on the sum of C_8–C_{12} PFCAs: perfluorooctanoate (C_8: PFOA), perfluorononanoate (C_9: PFNA), perfluorodecanoate (C_{10}: PFDA), perfluoroundecanoate (C_{11}: PFUA), perfluorododecanoate (C_{12}: PFDoDA), perfluorooctanesulfonate (PFOS), and perfluorooctane sulfonamide (PFOSA) are reported separately.

2.4. Statistical analysis

All temporal trend analyses were performed using a robust technique based on log-linear regression including a smoother as an alternative to the regression. The smoother applied here is a simple three-point running mean smoother fitted to the annual geometric mean values. In cases where the regression line is a poor fit, the smoothed line may be more appropriate. The significance of this line was tested by an analysis of variance (ANOVA), where the variance explained by the smoother, and the regression line, was compared with the total variance. This procedure has been used in assessments at the International Council for the Exploration of the Sea (ICES) and is described by Nicholson and co-workers (Nicholson et al. 1998). The statistical power was estimated using a method suggested by Nicholson and Fryer (1992). Multiple regression was used to identify and adjust for potential confounding factors such as whale age, length, and sex, i.e., expressing the concentrations as if these confounders were constant. Temporal trends after adjustments of confounders, when relevant, were assessed using the software package PIA (Bignert 2013). Where trends are significant, the time needed for CECs to double or halve in concentration (i.e., doubling and half times, or t_2 and $t_{1/2}$, respectively) were also determined. Data from HI were not adjusted for sex since almost all (159 of 169) HI whales used were males. The number of whales sampled in PG was considerably lower than in HI (69 vs. 169), which led to an increase in the proportion of females used for the CEC analysis; sex was included in the multiple regression for PG trends to account for this inclusion. In addition, several age and length data were unavailable further reducing the sample size. SQ PBDE data were only available for 2003 and 2013, and HBCD measurements were highly variable for 2005–2011; thus, only PFAS trends were investigated in SQ. Sex was also included in the SQ multiple regression to account for the large number of female whales used in 2011 and 2013. Unless specified, whale lengths were log transformed; details of the treatment of each CEC time-trend can be found in the Supplementary Material[2] and Table S1.[2] Due to the limited sample sizes and missing age and length data for PG and SQ, the confounding variables (sex/age/length) that were statistically insignificant were excluded to increase the available sample size and statistical power. Excluding confounding variables for trend analyses did not result in a statistically significant difference in the predicted trend for any location or contaminant. The student's *t*-test (two-tailed) and one-way ANOVAs were used to examine

differences ($\alpha = 0.05$) between specific years and/or sample sites using SigmaPlot (version 13.0, Systat Software Inc., Chicago, IL, USA), followed by a post-hoc Holm-Šídák pairwise multiple comparison test; when available, CEC concentrations were adjusted to age data prior to these tests. Unless specified, concentrations are presented as the geometric mean (gram) ± the standard error (SE).

3. Results

3.1. Size, age, and sex corrections

All temporal trend data were treated with a multiple regression to control for the confounding variables of whale size (total length), age, and sex. Only 10 of 169 whales sampled from HI were female; thus, sex was not included in the multiple regression for this population. Annual contaminant concentrations were not always significantly related to size, age, and/or sex for each location. For example, in the HI population, whale lengths were only significantly ($p < 0.05$) positively correlated to PBDE concentrations. In contrast, the ages of the HI whales were significantly positively correlated ($p < 0.05$) to PBDE, HBCD, PFOS, and PFOSA concentrations. Whale age was negatively correlated ($p = 0.012$) with PBDE concentrations in PG whales, implying that concentrations of PBDEs were lower in older whales. This result could be arising from the large number of female whales in the PG data set (13 of 69) potentially offloading the PBDEs to their offspring, although it is also likely that this is an artefact from the low sample sizes from PG. Sex correlations for PFASs in SQ beluga suggested a slight increase in concentrations in males; however, female whales were only sampled in 2011 and 2013 which were also the lowest years in concentration making the validity of this relation difficult to determine. A complete description of the corrections made for PG and SQ beluga is presented in the Supplementary Material,[2] and a summary is shown in Supplementary Table S1.[2]

Because of the small sample sizes and subsequent variability in the PG whale CEC measurements for most sample years limited the strength of the trend data obtained, and all comparisons must be made with care. Similarly, trend data for the SQ beluga population was limited to PFASs between 2003 and 2013. As such, the discussion will focus primarily on the strong data set from HI (16 years of data over 20 years for BFRs; 17 years over 29 years for PFASs).

3.2. BFRs

3.2.1. Spatial comparisons and congener profile

Lipid-weight (lw)-based concentrations of Σ_6BDEs and Σ_3HBCDs were measured in beluga blubber from three different locations (Fig. 1). A rigorous investigation of spatial trends was beyond the scope of this article and only comparisons of the temporal trends between locations are presented. Total PBDE concentrations were greater than HBCD concentrations in beluga whales from all three locations. Measurements of Σ_6BDEs ranged from 1.0 to 25.9 ng/g lw and 2.2 to 31.6 ng/g lw in HI and PG beluga, respectively (Table 1). Trends in Σ_6BDE concentrations were similar in HI and PG whales, increasing from 3 to 5 ng/g lw in the early 1980s to ca. 20 ng/g lw in 2007 (Fig. 2). Concentrations were only significantly different (i.e., lower in HI whales, $p < 0.001$) in 2008. However, this may be an artefact of the low sample size for PG in 2008 ($n = 4$); PBDE measurements in PG whales post-2008 were not available. SQ Σ_6BDE measurements were only available in 2003 and 2013. In comparison to HI whales, SQ beluga Σ_6BDE concentrations were significantly greater ($p = 0.013$) in both 2003 (22.1 ± 3.9 vs. 11.4 ± 1.6 ng/g lw) and 2013 (27.0 ± 6.8 vs. 13.8 ± 1.7 ng/g lw). 2003 SQ Σ_6BDE concentrations were also significantly greater than PG measurements in 2002 (22.1 ± 3.9 vs. 6.2 ± 2.3 ng/g lw, $p < 0.01$).

Beluga concentrations of Σ_3HBCDs ranged from 0.09 to 6.97 ng/g lw and non-detect (nd) to 9.82 ng/g lw in HI and PG whales, respectively. Temporal trends of HI and PG beluga

Fig. 1. Map of the three beluga sampling sites used in the present study, and the total number of animals sampled over the study period; specifically, Hendrickson Island (HI), 1984–2013; Pangnirtung (PG), 1982–2010; and Sanikiluaq (SQ), 2003–2013. Mapping image created using the Toporama — Mapping Tool (2017) (The Atlas of Canada), and contains information licensed under the Open Government License — Canada.

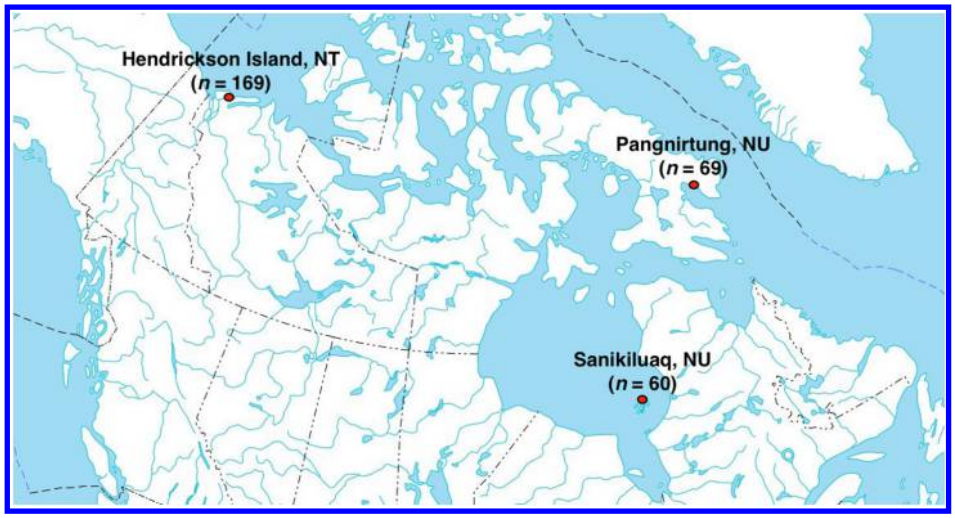


Table 1. Summary of beluga contaminant and morphological data measurements of three populations: Hendrickson Island (HI, NT), Pangnirtung (PG, NU), and Sanikiluaq (SQ, NU). The total range of collection years is shown by location, with the number of years for which data was collected in parentheses. The total number of animals sampled is shown, although data for each contaminant were not available for every individual.

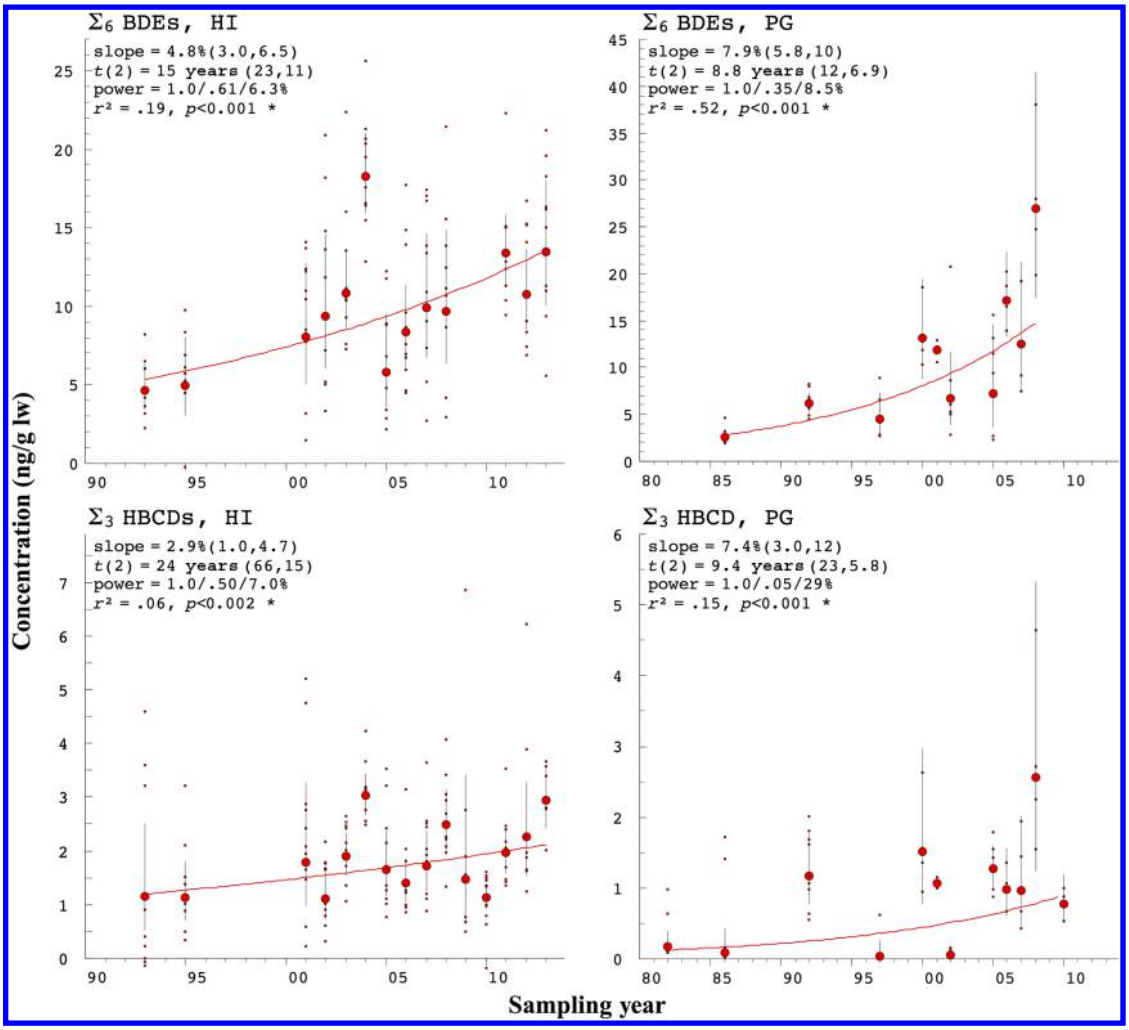
	HI	PG	SQ
Collection years	1984–2013 (17)	1982–2010 (12)	2003–2013 (6)
Sample number	169	69	60
Age range (years)	15–84	3–67	<1–56
Length range (cm)	335–513	194–488	116–467
Σ_6BDEs[a]	1.0–25.9	2.2–31.6	5.1–69.4
Σ_3HBCDs[a]	0.1–7.0	ND–9.8	ND–16.5
Σ_5PFCAs[b]	4.9–72.3	1.1–171.1	11.6–163.3
PFOS[b]	0.1–148.9	0.8–56.2	15.4–200.0
PFOSA[b]	3.3–228.4	4.4–120.4	47.0–920.5

Note: ND, non-detected.
[a]Measured in beluga blubber; units in ng/g lw.
[b]Measured in beluga liver; units in ng/g ww.

Σ_3HBCD concentrations were not significantly different, falling within the 95% confidence intervals. However, the variance in PG measurements (Fig. 2) is large, limiting the detection of differences among trends. In contrast, SQ beluga Σ_3HBCD concentrations in 2003 were significantly greater ($p < 0.05$) compared to both HI and PG (in 2002) whales; however, concentrations in 2013 were not significantly different between the HI and SQ populations.

The PBDE congener profile is shown using arithmetic mean values for both HI and PG beluga (Supplementary Fig. S4).[2] The rank order of PBDEs in HI whales was relatively constant throughout the study period following BDE-47 >> BDE-100 ≈ BDE-99 > BDE-85 ≈ BDE-154 > BDE-153; BDE-85 was not measured in substantial quantities after 2008.

Fig. 2. Temporal trends of BFRs in beluga whales sampled near Hendrickson Island (HI, NT) and Pangnirtung (PG, NU) between 1982 and 2013, with percent annual change (and 95% confidence intervals), doubling times (t_2), and statistical trend power to detect an annual change of 5%/power, if the slope is 5% and number of years is 10/lowest detectable change for a 10-year period with the predicted slope at a power of 0.80. Individual data points are indicated by black dots, annual geometric means in red; error bars depict the 95% confidence intervals. Red lines represent the calculated log-linear regression. One outlier omitted in both HI PBDE and HBCD trends (in 2005 and 2007, respectively).



The congener profile in PG whales was similar although it varied over time, roughly following BDE-47 >> BDE-100 ≈ BDE-154 > BDE-99 > BDE-85 ≈ BDE-153; post-2000 levels of BDE-154 and -100 decreased and increased, respectively, and BDE-85 was not measured in significant quantities after 1997. α- and β-HBCD were the predominant isomers measured in whales from all three locations.

3.2.2. Temporal trends

3.2.2.1. PBDEs

HI beluga Σ_6BDE concentrations increased by 4.8% per year (doubling time, $t_2 = 15$ years) from 1993 (4.7 ± 0.5 ng/g lw) to 2013 (13.8 ± 1.7 ng/g lw) levels (Fig. 2). A summary of the

Table 2. Temporal trends for several CECs measured in Beluga whales in the Canadian Arctic from 1982 to 2013. Annual percent increases/decreases are presented with 95% confidence intervals (CIs) and doubling (t_2) or half times ($t_{1/2}$) (t_2/$t_{1/2}$ 95% CIs presented in parentheses).

Location		Years	Sample No.	Slope (%)	95% CI (%)	t_2/$t_{1/2}$ (years)	Significance
HI	Σ_3HBCDs	1993–2013	129	2.9	1.0, 4.7	24 (66, 15)	$p < 0.002$
	Σ_6BDEs	1993–2013	114	4.8	3.0, 6.5	15 (23, 11)	$p < 0.001$
	Σ_5PFCAs	1984–2013	158	−6.0	−7.5, −4.4	12 (9.2, 16)	$p < 0.001$
	PFOS[a]	1984–2013	140	5.4	3.8, 7.0	13 (18, 9.9)	$p < 0.001$
	PFOSA[a]	1984–2013	142	−3.0	−5.2, −1.1	22 (13, 63)	$p < 0.003$
PG	Σ_3HBCDs	1982–2010	60	7.4	3.0, 12	9.4 (23, 5.8)	$p < 0.001$
	Σ_6BDEs	1986–2008	49	7.9	5.8, 10	8.8 (12, 6.9)	$p < 0.001$
	Σ_5PFCAs	1982–2010	63	6.7	4.2, 9.2	10 (17, 7.5)	$p < 0.001$
	PFOS	1982–2010	63	1.6	0.031, 3.2	43 (NA[b], 22)	$p < 0.043$
	PFOSA[a]	1982–2010	60	−3.0	−4.7, −1.2	23 (15, 56)	$p < 0.001$
SQ	Σ_5PFCAs	2003–2013	59	−12	−15, −8.7	5.7 (4.5, 7.9)	$p < 0.001$
	PFOS[a]	2003–2013	59	−5.4	−9.1, −1.6	13 (7.6, 45)	$p < 0.007$
	PFOSA[a]	2003–2013	58	−6.0	−11, −0.91	11 (6.3, 76)	$p < 0.021$

[a]Non-linear trend components present.
[b]t_2 = 2228 years based on an annual increase of 0.031%; not significant.

temporal trends by location and contaminant is shown in Table 2. Similarly, Σ_6BDE concentrations increased in PG whales by 7.9% annually (t_2 = 8.8 years) from 1982 (3.2 ± 0.3 ng/g lw) to 2008 (21.5 ± 3.4 ng/g lw).

3.2.2.2. HBCDs

A small but statistically significant increase in Σ_3HBCD concentrations (2.9% per year, t_2 = 24 years) was measured in HI beluga between 1993 (0.4 ± 0.6 ng/g lw) and 2013 (3.1 ± 0.3 ng/g lw) (Fig. 2). The overall trend in PG whale Σ_3HBCD concentrations increased by 7.4% per year (t_2 = 9.4 years) between 1982 (0.2 ± 0.1 ng/g lw) and 2010 (1.4 ± 2.3 ng/g lw), although concentrations appear to plateau after 1992 (Fig. 2); concentrations between 1992 and 2010 were not statistically different to each other ($p > 0.05$), except in 1997 and 2002 when concentrations were uncharacteristically low.

3.3. PFASs

3.3.1. Spatial comparisons and PFCA profile

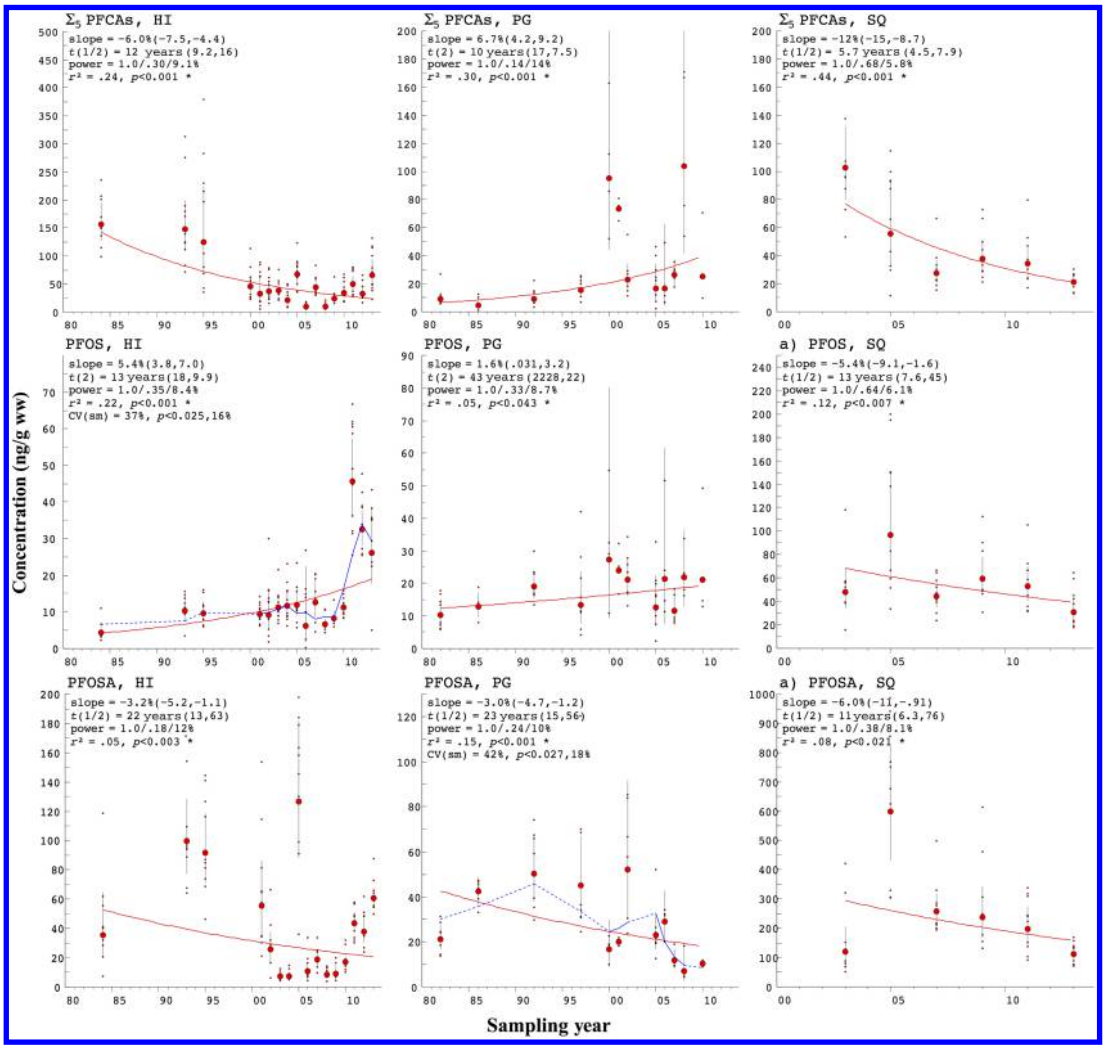
3.3.1.1. PFCAs

Wet-weight (ww) concentrations of Σ_5PFCAs, PFOS, and PFOSA were measured in beluga liver tissues from the three study locations. Ranges of CEC concentrations by location are shown in Table 1. Σ_5PFCA concentrations in HI beluga in 1984 (157 ± 15 ng/g ww) were significantly greater by orders of magnitude ($p < 0.001$, length normalized) than concentrations in PG beluga in 1982 (9.3 ± 2.5 ng/g ww). Temporal trends in Σ_5PFCA concentrations were opposite between the HI and PG whales, reaching approximately the same final concentrations (30–40 ng/g ww) in 2010. Concurrent with the decline in HI whales, Σ_5PFCAs decreased in the SQ population between 2003 and 2013. However, Σ_5PFCA concentrations were significantly larger in the SQ population than in HI beluga in 2003 (103 ± 12 vs. 38.7 ± 4.7 ng/g ww, $p < 0.001$), whereas the opposite was true in 2013 (20.9 ± 1.7 vs. 65.4 ± 11 ng/g ww, respectively, $p < 0.001$).

3.3.1.2. PFOS

PFOS concentrations between 1982 and 2010 were relatively constant in both HI and PG whales ca. 10–20 ng/g ww; concentrations in SQ whales were three to six times greater throughout the study period between 2003 and 2013 (Fig. 3). A significant ($p < 0.001$)

Fig. 3. Temporal trends of PFASs in beluga whales sampled at three locations: Hendrickson Island (HI, NT), Pangnirtung (PG, NU), and Sanikiluaq (SQ, NU) between 1982 and 2013, with percent annual change (and 95% confidence intervals), doubling and half times (t_2 and $t_{1/2}$), and the statistical trend power to detect an annual change of 5%/power, if the slope is 5% and number of years is 10/lowest detectable change for a 10-year period with the predicted slope at a power of 0.80. Individual data points are indicated by black dots, annual geometric means in red; error bars depict the 95% confidence intervals. Red lines represent the calculated log-linear regression. A three-point running geomean smoother is indicated by a blue line where significant ($p < 0.05$). Outliers removed in the HI PFOS (1× 1995, 2007), HI PFCA (1× 1995), SQ PFCA (PFUA excluded from one animal, 2003), and PG PFOSA (2× 2002, 1× 2005) trends.



increase in PFOS concentrations was measured in HI whales in 2011, which was not observed in the SQ population. Whales from PG post-2010 were not available, thus no comparison could be made.

3.3.1.3. PFOSA

Concentrations of PFOSA were significantly greater ($p = 0.032$, length normalized) in HI whales in 1984 in comparison to measurements of PG whales in 1982. Similarly, PFOSA

concentrations in the 1990s in HI beluga were roughly twice that measured in PG whales (ca. 100 vs. 50 ng/g ww). However, the overall temporal trends of PFOSA concentrations were similar between the HI and PG populations, increasing during the 1990s with a subsequent decline in the 2000s until 2010, reaching ca. 10 ng/g ww in both populations (Fig. 3). In contrast, SQ beluga PFOSA concentrations were 10 to 20 times greater ($p < 0.001$) than measurements in either HI or PG whales between 2003 and 2013, and were also still greater (3–10×) than either of the HI and PG whales in the 1990s.

3.3.1.4. PFCA profiles

The profiles of PFCAs (C_8–C_{12}) varied both over the study period and by location, and are depicted in Supplementary Fig. S5.[2] In HI beluga, pre-2007 levels followed approximately PFUA > PFOA > PFNA ≈ PFDA > PFDoDA, whereas post-2007 levels followed PFUA > PFDA > PFNA ≈ PFDoDA > PFOA. Levels of PFOA declined from ca. 30% of the total PFCAs in 1984 to negligible levels in 2007. Concurrently, PFUA increased from ca. 25% in 1984 to 60% in 2002 where it remained for the rest of the study period, and PFDA and PFDoDA remained relatively constant at ca. 20% and 10% for the entire study, respectively. Between 1984 and 2007, PFNA varied from ca. 0% to 38% of the total PFCAs; the post-2007 levels were remained consistent at ca. 8%. The PFCA profile in PG beluga was more varied, with no discernable trend over the study period. The relative abundances of the individual PFCAs were also different than in HI, on average following PFUA > PFDA > PFNA > PFOA ≈ PFDoDA between 1982 and 2010. The distribution of PFCAs in SQ whales was consistent between 2005 and 2013 following PFUA >> PFDA > PFDoDA > PFNA >> PFOA, whereas the 2003 distribution followed PFUA >> PFOA > PFNA ≈ PFDA > PFDoDA. The relative abundances were dissimilar between all locations investigated.

3.3.2. Temporal trends

3.3.2.1. PFCAs

HI beluga Σ_5PFCA concentrations decreased by 60% (6.0% per year, half-time $t_{1/2} = 12$ years) between 1984 (157 ± 15 ng/g ww) and 2013 (65 ± 11 ng/g ww) (Fig. 3). In contrast, Σ_5PFCA concentrations in PG beluga increased by 6.7% annually ($t_2 = 10$ years) from 9.3 ± 2.5 ng/g ww in 1982 to 25 ± 18 ng/g ww in 2010 (Fig. 3). Measurements of PFCAs in PG whales were uncharacteristically high in 2000, 2001, and 2008; however, the sample size was low for these years (2–4 whales/year) and their removal does not significantly change the observed trend. Σ_5PFCA concentrations in SQ whales decreased more rapidly than in the HI population, declining by 12% per year ($t_{1/2} = 5.7$ years) between 2003 (103 ± 12 ng/g ww) and 2013 (20.9 ± 1.7 ng/g ww).

3.3.2.2. PFOS

Levels of PFOS remained stable at approximately 10 ng/g ww in HI beluga whales between 1984 and 2010 before increasing precipitously to 44.9 ± 5.2 ng/g ww in 2010 (Fig. 2). A subsequent decline in concentrations followed in 2012 (35.7 ± 2.2 ng/g ww), and again in 2013 (27.9 ± 3.9 ng/g ww). A slight but statistically significant increase (1.6% per year, $t_2 = 43$ years, Fig. 3) in PFOS in PG whales was measured between 1982 and 2010, although the lower 95% CI is near zero percent (0.031%, Table 2). SQ beluga PFOS concentrations doubled between 2003 (47.7 ± 8.3 ng/g ww) and 2005 (96 ± 20 ng/g ww), but gradually decreased back to 2003 levels by 2013 (Fig. 3). The overall 10-year SQ-PFOS trend followed a 5.4% annual decline ($t_{1/2} = 13$ years, Fig. 3); however, omitting 2003 produces an 8-year trend with a steeper decline of 10% ($t_{1/2} = 6.7$ years, Supplementary Fig. S1[2]) from 2005 to 2013. Similarly, omitting 2005 produces a 10-year trend with no significant trend in PFOS

concentrations ($p = 0.138$). Thus, it is believed that the overall 2003–2013 trend in PFOS concentrations in SQ belugas is insignificant (i.e., concentrations stable), or follows a trend break starting in 2005; however, the applied three-point running mean smoother did not suggest any statistically significant non-linearity from 2005, and the lack of additional samples from other years (particularly prior to 2005) prevents a conclusive interpretation.

3.3.2.3. PFOSA

Although non-linear components were not determined to be significant in HI whale-PFOSA trends using the three-point running mean smoother, the regression poorly fits the available data ($R^2 = 0.05$, Fig. 3). However, PFOSA measurements in whales between 1984 and 1993 doubled in concentration ($p < 0.009$), and excluding 1984 from the overall regression produces a better fitting trend ($R^2 = 0.35$, Supplementary Fig. S2[2]) with an annual decline of −13% per year (−15%, −9.6%; $t_{1/2} = 5.5$ years, $p < 0.001$). After this decline, a significant increase ($p < 0.05$) in PFOSA concentrations is observed between 2010 and 2013, concurrent with the increase of PFOS concentrations during the same period for the same population (HI). PFOSA trends in HI and PG whales were similar, with an increase during the 1990s followed by a decline in the 2000s until 2010 (−3.0% per year, $t_{1/2} = 23$ years, Fig. 3). SQ beluga PFOSA trends were similar to SQ PFOS trends, increasing between 2003 (115 ± 22 ng/g ww) and 2005 (630 ± 70 ng/g ww), and decreasing back to 2003 levels in 2013 (106 ± 9 ng/g ww). The overall 10-year trend shows a decrease in SQ beluga PFOSA by 6% per year ($t_{1/2} = 11$ years), while omitting 2003 from the regression produces an 8-year trend with a steeper decline of 18% ($t_{1/2} = 3.9$ years) between 2005 and 2013 (Supplementary Fig. S3).[2] Again, similar to PFOS trends in the SQ whales, omitting 2005 produces a 10-year trend that is insignificant ($p = 0.736$), although the extent of the increase between 2003 and 2005 and the more gradual decline thereafter (Fig. 3) lend support for the trend break in 2005 and 18% annual decline to 2013 (Supplementary Fig. S3).[2]

4. Discussion

4.1. Concentrations and temporal trends of CECs in Arctic beluga

4.1.1. BFRs

Concentrations of Σ_6BDEs and Σ_3HBCDs steadily increased in both HI and PG beluga whales between the 1990s and 2010. Interestingly, belugas from HI possess trophic magnification factors for both PBDEs and HBCDs below unity (Tomy et al. 2009). Concentrations and trends of BFRs in the HI and PG populations were similar, and may provide support for similar fate/transport processes occurring circumpolarly. It is unclear what the specific drivers or causes of these higher concentrations are, but the greater Σ_6BDE concentrations in SQ whales points to higher sources that may be unique to the location, or food web/exposure differences (e.g., feeding at a higher trophic level).

4.1.2. PFASs

Temporal trends in PFASs varied both by compound and location. Despite possessing trophic magnification factors greater than unity (Tomy et al. 2009), Σ_5PFCA concentrations were shown to decrease in both the HI and SQ beluga populations. Increasing Σ_5PFCA concentrations in the PG whales may suggest higher sources or exposures unique to PG. The greatest decline in Σ_5PFCA concentrations in the HI population occurred between 1984 and 2004, whereas concentrations in the SQ population declined continuously from 2003 to 2013. Since concentrations in 2003 were roughly 2.5× greater in SQ whales than those from HI, it is possible that exposures/sources of PFCAs in the WHB were greater in earlier years, taking longer to decline to levels comparable to the HI population.

The similar concentrations and trends (i.e., mostly stable year-to-year) in PFOS between the HI and PG beluga populations support similar fate/transport/exposure. However, concentrations in 2011 rose dramatically in the HI population; unfortunately, no data was available post-2010 for PG whales for comparison. Furthermore, although a slight but significant increase of PFOS in PG whales was measured, the lower 95% CI lies near 0% annual change, and thus, this trend should be interpreted with caution. Although a statistically significant increasing trend was observed in PFOS in SQ belugas between 2003 and 2013, it is entirely dependent on the comparably high PFOS concentrations in 2005, and these results should be interpreted with caution. It is also possible that there was no overall trend in SQ whale PFOS concentrations, or that there was a non-linear trend with a significant increase in 2005 followed by a decline to 2013 levels.

None of the beluga PFOSA concentrations followed a strictly linear trend making direct comparisons among populations difficult. Of note, however, is that concentrations in SQ beluga were 10–20× greater than measurements made in either HI or PG whales, suggesting substantially greater exposures and/or sources of PFOSA in the WHB area and associated food web. In general, concentrations of PFOSA were similar or greater than PFOS across all three populations. Interestingly, this contrasts measurements of perfluorinated sulfonates made in other arctic marine mammals such as polar bears and ringed seals, both of which possess substantially higher levels of PFOS in comparison to PFOSA (Letcher et al. 2009; Tomy et al. 2009). Tomy et al. (2009) previously posited that this (specifically with regards to ringed seals) may suggest a greater efficacy for the biotransformation of PFOSA to PFOS by ringed seals in comparison to beluga whales.

4.2. Circumpolar trends in comparable biota

4.2.1. BFRs

The observations of increasing concentrations of Σ_6BDEs and Σ_3HBCDs in HI beluga contrasts temporal trends reported in the Arctic Monitoring and Assessment Programme (AMAP) 2015 assessment of Arctic temporal trends in POPs. The assessment found that most biota BDE-47 and -99, and α-HBCD time-series increased prior to 2000, but are now in a state of decline due to their decreased use and recent inclusion in the Stockholm Convention. In the present study, examining the trends in BDE-47 and -99 concentrations alone produced similar results to the Σ_6BDE trend in HI beluga. For example, increasing levels of PBDEs and HBCD in the early-mid 2000s followed by a plateau or decline into the 2010s has been reported for both ringed seal (*Phoca hispida*, PBDEs and α-HBCD in the East Baffin and Eastern Beaufort Sea populations, respectively), and in the eggs of thick-billed murre (*Uria lomvia*) and northern fulmar (*Fulmarus glacialis*) near Prince Leopold Island (Braune et al. 2015*b*; Houde et al. 2017). The authors suggest that the recent decrease in egg concentrations is presumably linked to the phase out and regulation of the octa- and penta-BDE, and HBCD commercial products. Similarly, increasing trends of 13% annually were reported in WHB polar bear (*Ursus maritimus*) Σ_{37}BDE concentrations between 1991 and 2007 (increases matched with BDE-47, -99, -100, and -153) (McKinney et al. 2010). More recently Letcher et al. (2018) reported decreases in both PBDE and HBCD concentrations in bears sampled between 2013 and 2014 from both the Southern and Western Hudson Bay (SHB and WHB) polar bear populations compared to 2007–2008 levels (with the exception of PBDEs in adult males from the WHB which increased) (McKinney et al. 2011*b*). The continued increase in Σ_6BDEs and Σ_3HBCDs in the HI (and PG) beluga population suggests that their phase-out has not had an immediate and direct effect on beluga exposure. This may point to growing BFR concentrations in the beluga food web; however, to our knowledge, no BFR (nor PFAS) trend measurements have been produced for beluga prey and food web species (e.g., arctic cod). Although some

high TL arctic marine mammals (e.g., polar bears and beluga whales) have been shown to be important in the metabolic depletion of PBDEs (particularly the highly brominated forms) (Tomy et al. 2009; McKinney et al. 2011*a*; Desforges et al. 2013), beluga concentrations continued to increase to 2013. Beluga feed at a slightly higher TL than ringed seals and are thus subject to higher burdens (Tomy et al. 2009) which may result in more time needed to metabolize these compounds. As such, more time may be necessary before a decline in concentrations is observed.

4.2.2. PFASs

The temporal trends of PFCAs in other Arctic animals have shown to either increase or not significantly change over time (Butt et al. 2007*a*, 2007*b*; Braune et al. 2010; AMAP 2016; Letcher et al. 2018) in contrast to the declining trend in HI belugas, although more recent measurements in the eggs of both thick-billed murre and northern fulmar eggs showed decreasing PFCA concentrations from 2008 and 2010, respectively, to 2012 (Braune and Letcher 2014). Because PFCAs decreased in our SQ time series but increased in the PG beluga population, these differences may be the result of differences in source exposure and/or diets among locations. Concentrations of PFASs in polar bears from the Amundson Gulf (AG) area (near our Eastern Beaufort Sea beluga population) have been reported for 2001 (Smithwick et al. 2005) and 2008 (Letcher et al. 2009). In their 2009 report, Letcher et al. observed a slight (but not significant) decline in PFOS concentrations in bears from the AG population between 2008 and 2001, but to date no statistical analyses has yet been performed on PFCA concentrations, preventing a comparison to PFCAs in our HI beluga population. To our knowledge, no other temporal trend studies have yet been done on PFASs in arctic marine animals in the Eastern Beaufort Sea area for comparison.

PFOS trends in several other North American Arctic fresh water and marine animals showed plateaus or declines during the 2000s (Butt et al. 2010; Braune and Letcher 2014) similar to that reported here for the HI beluga population prior to 2011. Due to the lack of data for other species into the 2010s, comparisons with other trends are difficult. Braune and Letcher (2014) reported decreases in PFOS levels in both northern fulmars and thick-billed murres from 2009 and 2008, respectively, to 2012, although the authors state that this may be an artefact of the variability in their dataset. Recently, Letcher et al. (2018) reported a significant decrease (59%) in polar bear (SHB) PFOS levels in 2013–2014 compared to bears from 2002. An increase in beluga PFOS levels was only measured in HI, which may suggest that this increase was localized to the Eastern Beaufort Sea population. This beluga population winters south of the Beaufort Sea in the Bering Sea (Citta et al. 2016; Hauser et al. 2017), and as a result, their exposure may be linked to Pacific drivers and systems sources. It is understood that PFOSA can degrade both biotically and abiotically to form PFOS (Butt et al. 2010), and as such, it is possible that the increase in beluga PFOSA concentrations from 2011 to 2013 are directly linked to the concurrent increase of PFOS (and potentially, the plethora of PFOS precursors that could be present but not measured) in 2011. Although animal trend data of PFOSA concentrations are limited, ringed seal from the WHB and Lancaster Sound showed maximal concentrations between 1998 and 2000 with subsequent declines to 2005 (Butt et al. 2007*b*), similar to the trend in both the HI and PG beluga populations. This may indicate a rapid response by the HI beluga and beluga food web to the phasing out of perfluorooctane-sulfonyl-fluoride-based compounds in 2001 by 3M, although it remains uncertain as to what may have resulted in the spike in HI PFOSA levels in 2011–2013.

4.3. Climate change and future trends

Increases in the global air and ocean temperatures are creating a shift in Arctic food webs (Grebmeier et al. 2006; Post et al. 2009), and the associated sea ice decline may

influence food webs, diet, and therefore, contaminant exposures of arctic species (McKinney et al. 2009; Gaden et al. 2012). For instance, McKinney et al. (2012) demonstrated that increased numbers of transient and subarctic species may alter the contaminant dynamics in the Cumberland Sound (NU) food web. Although substantial work has been done examining the connections between climate-change-induced effects and arctic biota mercury concentrations/speciation (Gaden and Stern 2010; Stern et al. 2012; Braune et al. 2015*a*; Loseto et al. 2015), there remains a gap in knowledge for the effects of these climate-induced changes on organic contaminants distribution in arctic marine mammals.

There has been a growing concern regarding the decline in the body condition of animals in the Beaufort Sea ecosystem (Harwood et al. 2015). Beluga whales from the Eastern Beaufort Sea in particular have been reported to have significantly declined in body size-at-age between 1989 and 2008 due to reductions in prey availability that may have come as a result of climate-induced ecosystem shifts (Harwood et al. 2014). Changes in the beluga food web as a result of ongoing climate-induced ecosystem shifts may result in significant changes in the exposure of belugas to CECs. Belugas are believed to feed primarily on arctic cod (*Boreogadus saida*) (Loseto et al. 2009; Quakenbush et al. 2015) which are currently the most abundant fish in the Beaufort Sea (Majewski et al. 2017). However, arctic cod may be displaced in the future by increases in the boreal Pacific sand lance (*Ammodytes hexapterus*) populations as the ice-free season continues to lengthen (Falardeau et al. 2014); such changes towards a more diverse boreal ecosystem have already been measured in the Hudson Bay (Chambellant et al. 2012; Gaston et al. 2012; Falardeau et al. 2014). As a result, there is a need for continued monitoring of CECs in the context of climate-induced changes (e.g., changes in sea-ice coverage), particularly for those contaminants continuing to increase. In addition, because many northern Indigenous communities depend on beluga as a country food, monitoring will continue to aid in the evaluation of contaminant exposure in these communities.

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