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Multi-year assessment (2006–2015) of persistent organic pollutant concentrations in blubber and muscle from Western Arctic bowhead whales (*Balaena mysticetus*), North Slope, Alaska

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ABSTRACT

Blubber and muscle were collected from male bowhead whales (n = 71) landed near Utqiaġvik (Barrow), Alaska, between 2006 and 2015 and analyzed for lipid content and concentrations of persistent organic pollutants (POPs) in order to determine levels and trends over the collection period. Collection year was a significant predictor of blubber concentrations for most classes of POPs, while for a few classes, animal length (proxy for age) was also a significant predictor. This is the first report on levels of PBDEs in bowhead whales; concentrations of these compounds are low (≤55 ng/g wet weight). Blubber concentrations were lower than those reported in samples collected between 1992 and 2000, and many POP classes in blubber declined significantly between 2006 and 2015. Concentrations of POPs in bowhead whale tissues, which are subsistence foods for Native Alaskan communities, appear to be declining at rates comparable with previously reported temporal trends in Arctic biota.

1. Introduction

The bowhead whale (*Balaena mysticetus*) is an ice-associated large (14–18 m) baleen whale with four stocks currently recognized by the International Whaling Commission (IWC) (Cooke and Reeves, 2018). The majority of western Arctic bowhead whales (*syn. Bering-Chukchi-Beaufort* (*BCB*) *stock*; *Bering Sea stock*), the only stock occurring in water of the U.S. Arctic, migrates annually during spring and fall between the northern Bering Sea (wintering area) to the eastern Beaufort Sea (summer area) (Braham et al., 1980; Citta et al., 2015; Moore and Reeves, 1993; Quackenbush et al., 2010). An alternate migration route, taken by a portion of the stock, occurs along the northern coast of Chukotka, Russia (Bogoslavskaya, 2003; Melnikov et al., 2004; Noongwook, 2007). The western Arctic bowhead whale is listed as "endangered" under the U.S. Endangered Species Act (ESA) and designated as "depleted" under the U.S. Marine Mammal Protection Act. Historically all four bowhead stocks were severely depleted because of intense commercial whaling from approximately the 16th to mid-19th century (Bockstoce and Botkin, 1983; Braham, 1984). The longevity of bowhead whales may exceed "*two human lifetimes*" based on traditional ecological knowledge, lens aspartic acid racemization ageing, and recovery of historical weapon fragments (George et al., 1999; George and Bockstoce, 2008). Long-term monitoring of the western Arctic subpopulation via passive acoustic and visual ice-based census counts (Givens et al., 2013) indicates a steady recovery since the end of commercial whaling with an estimated annual rate of increase of 3.4% and a 2011 population estimate of 16,892 (95% confidence interval: 15,704–18,928) (Givens et al., 2013; Givens et al., 2018; Zeh and Punt, 2005).

Bowhead whales have been hunted by Inuit communities of both the Bering Strait and North Slope, Alaska since ~2000 BCE (Carroll, 1976; Marquette and Bockstoce, 1980; Stoker and Krupnik, 1993). Today, as in the past, bowhead meat (muscle) and muktuk (outer blubber layer with black skin attached) are shared throughout the Alaskan whaling communities and beyond and constitute essential nutritional, cultural, and spiritual subsistence resources for northern Arctic communities (Braund, 2018). Similar to other marine mammals (Muir et al., 1988), both blubber and muscle of bowhead whales are effective reservoirs of

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lipophilic environmental contaminants (Hoekstra et al., 2005). The primary waterbodies (Bering, Chukchi, and Beaufort seas) inhabited by the western Arctic stock have been characterized by overall low contaminant levels relative to the rest of the Arctic with the exception of β-hexachlorocyclohexane (HCH), which is found in higher concentrations in the western Arctic compared to other geographical regions (Iwata et al., 1993; MacDonald et al., 2000). Preliminary studies conducted in the 1980s indicated POP exposure in these whales (McFall et al., 1986); however, consistent with the bowhead's lower trophic position within the marine food web (Lowry and Frost, 1984; Lowry et al., 2004), tissue concentrations of POPs were generally low (McFall et al., 1986; Mössner and Ballschmiter, 1997; O'Hara et al., 1999). Processes involved in sex- and age-related bioaccumulation and trophic-related biomagnification of POPs have been elucidated in several integrated contaminant-stable isotope studies of blubber tissues from landed bowhead whales (Hoekstra et al., 2002a, 2003a; Hoekstra et al., 2005; Hoekstra et al., 2003b; Hoekstra et al., 2002b; Hoekstra et al., 2002c; Hoekstra et al., 2003c; Hoekstra et al., 2002d; O'Hara et al., 1999). These studies demonstrated that observations concerning POPs concentrations noted in other cetacean species (Aguilar et al., 1999) also occur in bowhead whales (e.g. tissue levels increase with increasing age in males and senescent females; adult males have higher levels than reproductive females).

The production of many POPs has now been regulated (e.g. Stockholm Convention, EPA, 2017; Vijgen et al., 2011) and their use has either ceased [e.g., open use of PCBs, phase out of polybrominated diphenyl ethers (PBDEs)] or is strongly restricted (e.g., DDTs). Therefore, inputs into the environment are much lower than those that occurred in previous decades and are now predominantly associated with historical use or gradual depletion of existing chemical stocks (Diamond et al., 2010). This has been reflected in trends of POPs in a variety of marine biota (Schell, 2000; West et al., 2008), including Arctic species (Addison et al., 2014; AMAP, 2016; Braune, 2007; Braune et al., 2005; de Wit et al., 2006; Mallory and Braune, 2012; Riget et al., 2019; Riget et al., 2010a; Vander Pol and Becker, 2007; Vander Pol et al., 2004). Major classes of POPs show declining trends in Arctic animals since reaching a peak in the early 1980s, with reductions of ~2–10% per year in marine biota (AMAP, 2016; Riget et al., 2019; Riget et al., 2010a). The PBDEs, a class of flame retardants, has demonstrated a similar propensity to other POPs to bioaccumulate in animals and biomagnify in marine food webs (Ikonomou et al., 2002). Prior to 2005, PBDEs exhibited a rapidly rising trend in marine animals including cetaceans, both in temperate areas (Rayne et al., 2004) and within the Arctic (Hoguet et al., 2013; Ikonomou et al., 2002; Riget et al., 2019). These observations have resulted in a planned phase-out of production of PBDE products in the U.S. (EPA, 2017) and listing under Annex A of the Stockholm Convention (2009 for penta- and octa-BDE commercial products; 2017 for decaBDE). As a result, a declining trend in concentrations in marine biota has also been noted over the last decade (Riget et al., 2019; Ross et al., 2013), although declines have not been uniform (Braune, 2007; Hoguet et al., 2013) and there are expected to be continued emissions due to the stock of products containing PBDEs that are still in use (Abbasi et al., 2015).

Contaminant monitoring of subsistence harvested bowhead whales remains a priority for the Alaska Eskimo Whaling Commission (AEWC), the North Slope Borough Department of Wildlife Management (NSB-DWM), and communities of northern Alaska due to impacts on human and wildlife health and the conservation biology and management of the bowhead whale. The current study contributes to the body of literature on contaminants in baleen whales, which remain less well studied than higher trophic level toothed whale species (Aguilar et al., 2002; O'Shea and Brownell, 1994). This study is the first report on temporal trends of legacy POPs, including hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethane-related pesticides (DDTs), chlordanes (CHLs) and polychlorinated biphenyls (PCBs) in bowhead whales, in a region (the western Arctic) for which fewer temporal trends data sets are available. This study also is the first to report levels of PBDEs in bowhead whales.

2. Materials and methods

2.1. Study area and sample collection

Subsistence hunting of bowhead whales by Alaskan Native hunters has been regulated with annual quotas established by the IWC since 1977 (IWC, 2018). In cooperation with the AEWC and whaling captains from 11 whaling communities, since the mid-seventies bowhead whales have been regularly measured and inspected (post mortem evaluation) by hunters, NSB-DWM staff (i.e., biologists, veterinarians) and collaborators to assess the health status of landed whales and to collect tissue samples for baseline data on life history, natural diseases, and marine threats (Philo et al., 1993; Rolland et al., 2019; Stimmelmayr et al., 2017; Von Duyke et al., 2016). Field collection methods from landed bowhead whales have been previously described (Hoekstra et al., 2002b; O'Hara et al., 1999). For our study, full thickness blubber cores (dorsal midline, ~0.5–1 m caudal to the blowhole) and epaxial muscle samples were collected in the field from whales landed from March–October near Utqiaġvik (Barrow), Alaska (71.2906° N, 156.7886° W) between 2006 and 2015. Life history (i.e. sex, age, reproductive status, history of entanglement, shipstrike, killer whale interaction) and body measurements (i.e., total body length, girth measurements, blubber depth measurements, baleen length, etc.) were recorded for each whale landed. Samples were stored at minus 20 °C at the Barrow Arctic Research Center (Utqiaġvik, AK) and shipped frozen to the Northwest Fisheries Science Center (NWFSC) laboratories in Seattle, Washington, USA, for analysis. Because of offloading of contaminants by reproductive females, only males were included in our study, with the objective of documenting trends of POPs in bowhead whale tissues over the 10-year study period.

2.2. Chemical analysis

Blubber cores (full depth skin to muscle) and muscle subsamples were trimmed under clean laboratory conditions, homogenized, and stored at −80 °C in trace clean glass containers. Samples of full depth blubber and muscle were extracted and analyzed for POPs and lipid content using a previously described method (Sloan et al., 2014). Weighed portions (approximately 0.5 g of blubber or 2 g of muscle) were mixed with sodium sulfate and magnesium sulfate (to remove water) and were then extracted with dichloromethane using automated, pressurized solvent extraction. Polar biogenic compounds were removed from the extracts by filtering them through gravity-flow silica/alumina column, followed by a size-exclusion high-performance liquid chromatography cleanup step to remove lipids and other neutral interfering compounds. The concentrated cleaned up extracts were analyzed for POPs by gas chromatography/mass spectrometry (GC/MS) and the analytes were quantified relative to surrogate standards using multiple concentration levels of GC/MS calibration standards. Percent lipid was determined gravimetrically using a portion of the extract taken prior to the silica-alumina cleanup step.

The POPs analyzed by GC/MS were *o,p′*-DDD, *o,p′*-DDE, *o,p′*-DDT, *p,p′*-DDD, *p,p′*-DDE, *p,p′*-DDT (ΣDDTs); aldrin, endosulfan I, mirex, hexachlorobenzene (HCB), α-, β-, and γ-hexachlorocyclohexane (ΣHCHs); *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, nonachlor III, heptachlor, heptachlor epoxide, and oxychlordane (ΣCHLs); 46 polychlorinated biphenyl congeners (as 40 chromatographic peaks) 17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 82, 87, 95, 99, 101/90, 105, 110, 118, 128, 138/163/164, 149, 151, 153/132, 156, 158, 170, 171, 177, 180, 183, 187/159/182, 191, 194, 195, 199, 205, 206, 208, 209 (ΣPCBs); polybrominated diphenyl ether congeners 28, 47, 49, 66, 85, 99, 100, 153, 154, 155, 183 (ΣPBDEs). The lower limit of quantitation (LOQ) for each analyte was based on sample mass, as

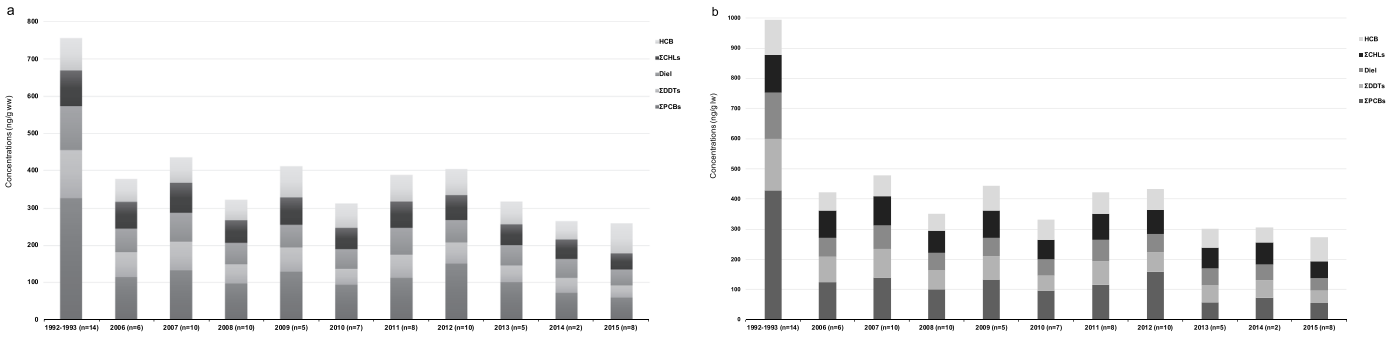

Fig. 1. Fig. 1a. Concentrations (ng/g ww) of POPs in in blubber of bowhead whales collected during Alaskan subsistence hunts 1992–2006, showing proportions of POPs and decline in concentrations over the periods between 1992 and 1993 (O'Hara et al., 1999) and the current data set (2006–2015).
For purposes of comparison, ΣPCB was calculated as the sum of 17 congeners, times two, as reported in O'Hara et al. (1999). All other sums include the same individual POPs.
Fig. 1b. Concentrations (ng/g lw) of POPs in blubber of bowhead whales collected during Alaskan subsistence hunts 1992–2006, showing proportions of POPs and decline in concentrations over the periods between 1992 and 1993 (O'Hara et al., 1999) and the current data set (2006–2015).
For purposes of comparison, ΣPCB was calculated as the sum of 17 congeners, times two, as reported in O'Hara et al. (1999). All other sums include the same individual POPs.

well as the analyte areas in the lowest-level calibration standard, i.e., the LOQ is the concentration that an analyte would have in a given sample if it had a GC/MS peak area equal to the analyte's GC/MS peak area in the lowest level GC/MS calibration standard analyzed concurrently. The LOQs of individual POP analytes ranged from 0.99 to 2.7 ng/g wet weight of blubber and 0.061 to 0.19 ng/g wet weight of muscle.

2.3. Quality assurance for POPs and percent lipid

Quality assurance (QA) guidelines for measuring POPs and percent lipid were followed as described by Sloan et al. (2006). The recoveries of the surrogate standard (PCB 103) ranged from 86 to 109% and met our laboratory criteria (range of 60 to 130%). A sample of the Standard Reference Material® (SRM) 1945 (Organics in Whale Blubber) from the National Institute of Standards and Technology was analyzed with each batch of 12 bowhead blubber samples. In these SRM 1945 samples ($n = 6$), 86 to 98% of the 51 analytes having certified values were within 30% of either end of the 95% confidence interval for the certified value. Percent lipid measured in all the SRM 1945 samples were within the 95% confidence interval for the reference value. A sample of the SRM 1947 (Lake Michigan Fish Tissue) was analyzed with each batch of 12 bowhead muscle samples, as the lipid content of the SRM 1947 fish tissue is more similar to the muscle samples than is SRM 1945. In these SRM 1947 samples ($n = 6$), 94 to 98% of the 48 analytes having certified values were within 30% of either end of the 95% confidence interval for the certified value. Percent lipid measured in all the SRM 1947 samples were within 8% of either end of the 95% confidence interval for the reference value. A solvent (dichloromethane) method blank sample was analyzed with each batch of bowhead blubber or muscle samples. No more than 5 analytes in each method blank sample ($n = 12$) had concentrations greater than two times their respective LOQs.

2.4. Statistical analysis

Mean concentrations of analytes (ng/g wet wt and lipid wt) in bowhead blubber and muscle samples were compared among the sampling years using one-way Analysis of Variance (ANOVA) and the Tukey-Kramer Honestly Significant Difference (HSD) post hoc test for those measurements with significant differences by year (Zar, 1984), with the significance level set at $p < 0.05$. Due to large differences in the variances about the means and left-censored data for muscle samples, all " < LOQ" results for POPs were replaced by zero and the statistical analyses for blubber and muscle POPs were performed on transformed log(concentration + 1) data, while analyses of animal length were performed on log-transformed data and lipid content on arcsin-transformed data to meet assumptions of equal variance and normality (Zar, 1984). Statistical analyses for ANOVAs, and Tukey-Kramer HSD were completed using JMP Statistical Software (version 12.0.1) (SAS Institute, Inc., Cary, NC). In addition, Generalized Linear Model (GLM) analyses were performed in R (3.5.1, 2018-07-02; R Project for Statistical Computing) to evaluate the contributions of collection year, collection month, animal length (a proxy for age), and lipid content to variation of summed POPs classes or individual POPs, as well as selected contaminant ratios. Temporal trends of POPs were estimated by using the slope of the GLM regression vs. collection year, where these regressions were statistically significant. To compare models, we calculated four values for each model: Akaike's Information Criterion (AIC), delta AIC, relative likelihood, and AIC weight. Smaller AIC values indicate "better" models, with delta AIC measuring the difference in AIC between two models (Akaike, 1973; Anderson and Burhham, 2002). The best model was defined as having the lowest AIC, although preference was given to the simplest model(s) if two or more models had a delta AIC of < 2. In addition, in order to compare directly with Arctic Monitoring and Assessment Programme (AMAP) temporal trends assessments, lipid-corrected POPs (blubber) were analyzed (adjusted for animal length) using PIA (Bignert, 2013), a statistical package developed for the International Council for the Exploration of the Sea (ICES) to robustly analyze contaminant temporal trends (Fryer and Nicholson, 1999; Nicholson et al., 1991).

3. Results and discussion

3.1. Overall results and comparisons of POPs levels with previously published data

Arithmetic mean blubber concentrations of selected POPs are reported in Fig. 1 (1a, wet weight (ww); 1b, lipid weight (lw)). We compared these mean concentrations with POP levels determined in bowheads collected in 1992–1993 (O'Hara et al., 1999), using the same analytes and the same method for estimating ΣPCBs, although O'Hara et al. did not report the sex of the animals analyzed for the wet weight data. Generally, it appears that POP concentrations have decreased between 1992 and 1993 and 2006–2015; in particular, mean ΣDDTs and ΣPCBs in blubber from all years of the current study are appreciably lower (2–4 times) than those reported by O'Hara et al. (1999).

ANOVA analyses for bowhead blubber samples collected from 2006 to 2015 showed significant differences based on collection year for percent lipid ($p < 0.0001$), HCB ($p = 0.026$), ΣCHLs ($p = 0.0076$),

Table 1
Wet weight mean concentrations (ng/g) of POPs in male bowhead blubber and muscle collected during Alaskan subsistence hunts 1997–2016.

	n		Body length (m)	% Lipid	HCB	ΣHCHs	ΣCHLs[a]	Dieldrin	ΣDDTs[b]	ΣPCBs[c]	ΣPBDEs
1997–1999[d]	71	Blubber		76 ± 1.6	100 ± 7.0	203 ± 13	152 ± 9.2	84 ± 4.3	331 ± 55	410 ± 29	
	5	Muscle		2.39 ± 1.85	1.60 ± 0.96	2.74 ± 1.68	2.32 ± 1.17		1.71 ± 1.01	1.87 ± 0.90	
1999–2000[d]	25	Blubber	10.9	70 ± 2.6		282 ± 25	260 ± 928		437 ± 40	541 ± 45	
2005–2009[d]	3	Blubber			23.82 ± 2.14				6.29 ± 1.16	317.61 ± 87.43	
	3	Rendered oil			16.91 ± 0.76				26.43 ± 14.06	354.06 ± 201.22	
	4	Muscle			0.58 ± 0.28				(3) 0.27 ± 0.28	27.20 ± 50.42	
2006	6	Blubber	10.8 ± 1.1	79 ± 2.0	60 ± 5.8	97^{A} ± 1.7	84 ± 7.4	62 ± 14	$65^{A,B}$ ± 7.6	78 ± 7.1	< LOQ
	5	Muscle		0.86 ± 0.27	0.85 ± 0.29	1.65 ± 0.36	0.63 ± 0.42	0.76 ± 0.28	(4) 0.54 ± 0.28	4.1 ± 0.43	(2) 0.18 ± 0.04
2007	10	Blubber	10.7 ± 0.74	81^{A} ± 0.79	66 ± 4.7	106^{A} ± 5.7	93^{A} ± 7.4	73 ± 7.7	71^{A} ± 11	$91^{A,B}$ ± 7.7	(5) 1.1 ± 0.064
	10	Muscle		0.78 ± 0.29	0.89 ± 0.37	1.6 ± 0.36	(7) 1.1 ± 0.67	0.79 ± 0.54	(5) 0.58 ± 0.68	5.0 ± 0.54	(3) 0.13 ± 0.005
2008	10	Blubber	9.6 ± 0.51	83^{A} ± 1.6	52 ± 7.7	85^{A} ± 6.9	67 ± 9.0	53 ± 7.4	48 ± 6.1	$65^{B,C}$ ± 6.7	(3) 0.99 ± 0.67
	10	Muscle		0.90 ± 0.21	0.69 ± 0.12	1.4 ± 0.25	(9) 0.60 ± 0.17	0.53 ± 0.13	(5) 0.54 ± 0.08	5.2 ± 0.43	(1) 0.13
2009	5	Blubber	8.9 ± 0.39	84^{A} ± 0.82	81^{A} ± 9.0	105^{A} ± 9.1	87 ± 9.3	58 ± 7.7	64 ± 8.8	$89^{A,B}$ ± 11	(1) 1.3
	5	Muscle		0.58 ± 0.07	0.58 ± 0.09	1.4 ± 0.22	0.50 ± 0.23	0.37 ± 0.15	(2) 0.25 ± 0.05	3.7 ± 0.20	< LOQ
2010	7	Blubber	8.4 ± 0.53	85^{A} ± 1.1	61 ± 12	79 ± 11	62 ± 9.8	48 ± 9.2	$39^{B,C}$ ± 7.0	$61^{B,C}$ ± 11	< LOQ
	7	Muscle		0.84 ± 0.11	0.68 ± 0.19	1.1 ± 0.25	(6) 0.50 ± 0.23	0.45 ± 0.14	(4) 0.28 ± 0.17	4.1 ± 0.21	(3) 0.092 ± 0.004
2011	8	Blubber	9.9 ± 0.79	82^{A} ± 1.2	68 ± 6.5	91^{A} ± 7.6	80 ± 9.5	63 ± 10	55 ± 9.2	$74^{B,C}$ ± 9.1	(1) 0.95
	8	Muscle		0.64 ± 0.13	0.64 ± 0.13	1.2 ± 0.23	(6) 0.47 ± 0.10	(7) 0.36 ± 0.09	(5) 0.23 ± 0.11	3.9 ± 0.26	(2) 0.12 ± 0.003
2012	10	Blubber	9.2 ± 1.2	85^{A} ± 0.59	67 ± 4.6	94^{A} ± 6.5	78 ± 6.6	57 ± 6.1	54 ± 4.3	93^{A} ± 5.0	(2) 1.5 ± 1.4
	9	Muscle		0.73 ± 0.55	0.65 ± 0.29	0.95 ± 0.32	(5) 0.59 ± 0.36	(8) 0.46 ± 0.15	(2) 0.60 ± 0.38	4.8 ± 64	< LOQ
2013	5	Blubber	9.7 ± 1.3	79 ± 0.76	61 ± 5.8	72 ± 6.0	64 ± 6.3	53 ± 6.4	43 ± 5.5	$68^{A,B}$ ± 5.8	(1) 0.68
	4	Muscle		1.1 ± 0.42	1.1 ± 0.48	1.3 ± 0.47	0.49 ± 0.60	0.63 ± 0.34	(3) 0.37 ± 0.41	5.7 ± 0.46	(1) 0.12
2014	2	Blubber	10.7 ± 1.3	70^{B} ± 0.86	49 ± 6.0	69 ± 4.5	62 ± 6.5	50 ± 4.5	40 ± 5.0	59 ± 4.0	(1) 0.73
	1	Muscle		0.39	0.42	0.89	< LOQ	< LOQ	< LOQ	5.1	< LOQ
2015	8	Blubber	10.4 ± 0.61	80^{A} ± 1.7	46^{B} ± 2.6	59^{B} ± 3.9	51^{B} ± 3.3	40 ± 3.7	32^{C} ± 2.4	47^{C} ± 2.2	(4) 7.0 ± 14
	7	Muscle		0.58 ± 0.16	0.40 ± 0.081	0.90 ± 0.092	(4) 0.38 ± 0.032	0.27 ± 0.054	(3) 0.17 ± 0.019	5.5 ± 0.17	(5) 0.15 ± 0.046

Geometric means ± SE for 2006–2015, all males. The number in parentheses before the mean indicates the number of measurements above the LOQ, if different from *n*.
ΣPBDEs = sum of BDEs 47, 49, 66, 85, 99, 100, 153, 154, 155, 183.
Capital letter superscripts denote difference levels by Tukey-Kramer HSD for each measured quantity by year (2006–2015); quantities without superscripts are not significantly different from quantities with any superscript.

[a] ΣCHLs = sum of cis-chlordane, trans-chlordane, cis-nonachlor, trans-nonachlor, oxychlordane, and heptachlor epoxide (Hoekstra et al.) and including nona III (this study).

[b] ΣDDTs = sum of *o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, and *p,p′*-DDD, except Welfinger-Smith et al. (2011), which reports *p,p′*-DDE + cb85.

[c] ΣPCBs based on the sum of 126 congeners (1997–1999 and 1999–2000), 101 congeners (2005–2009) or 46 congeners (this study, 2006–2015).

[d] Means ± SE for blubber 1997–1999 from Hoekstra et al. (2002b) Table 1; means ± SE for blubber 1999–2000 from Hoekstra et al. (2003b) Table 1; means ± SD for muscle from Hoekstra et al. (2005) Table 1; means ± SD for 2005–2009 from Welfinger-Smith et al. (2011) Tables 2 and 3.

Table 2
Best fit General Linear Model parameters and coefficients for POP classes and selected POP ratios in blubber of male bowhead whales landed at Utqiaġvik (Barrow), Alaska 2006–2015.

Variable	Model description	AIC	Parameter	Estimate	SE	p-val
Dieldrin	Year + length	627.22	**Intercept**	79.201	29.496	**0.0091**
			Collection year	−0.0377	0.0147	**0.0124**
			Body length	0.0601	0.2058	**0.0048**
ΣCHLs	Year + length	648.24	**Intercept**	93.749	26.083	**0.0006**
			Collection year	−0.4467	0.013	**0.0098**
			Length	0.0376	0.0182	**0.0427**
ΣDDTs	Year + length	616.65	**Intercept**	129.877	29.824	**4.6e−05**
			Collection year	−0.06297	0.0148	**6.75e^{-05}**
			Body length	0.06886	0.0208	**0.0015**
ΣHCHs	Year	637.56	**Intercept**	100.535	20.657	**6.91e^{-06}**
			Collection year	−0.04778	0.01028	**1.55e^{-05}**
ratio ppDDE:ΣDDTs	Year + length	−340.03	**Intercept**	−39.397	5.5837	**1.13e^{-09}**
			Collection year	0.0191	0.00278	**2.27e^{-09}**
			Body length	−0.0123	0.0039	**0.0023**
ratio aHCH:bHCH	Year	−133.977	**Intercept**	153.034	14.417	**3.7e^{-16}**
			Collection year	−0.07645	0.0072	**3.15e^{-16}**
ΣPCBs (46 cbs)	Month	681.04	**Intercept**	4.572	0.145	**2.0e^{-16}**
			Collection month	−0.0222	0.0176	0.211
	Year	681.07	Intercept	45.638	31.436	0.151
			Collection year	−0.0205	0.0156	0.194
	Lipid	681.43	**Intercept**	3.435	0.7854	**4.5e^{-05}**
			Percent lipid	0.0118	0.00957	0.223
	Length	681.99	**Intercept**	4.231	0.2202	**2e^{-016}**
			Body length	0.0169	0.2171	0.439
HCB	Year	619.30	Intercept	46.3155	26.736	0.0877
			Collection year	−0.2098	0.0133	0.1194
	Lipid	619.44	**Intercept**	3.0879	0.6782	**2.21e^{-05}**
			Percent lipid	0.01298	0.0083	0.121
	Length	621.04	**Intercept**	4.3011	0.1903	**2e^{-16}**
			Body length	−0.0149	0.0188	0.429

See Materials and methods section for selection criteria of best fit models; significant parameter *p*-values are bolded.

ΣDDTs ($p = 0.0013$), ΣHCHs ($p < 0.0001$), and ΣPCBs ($p < 0.0001$). Whale length did not differ significantly by year ($p = 0.1732$). Tukey-Kramer HSD was performed post hoc for variables with significant differences by ANOVA by year to determine difference levels (Table 1). In contrast to blubber, muscle percent mean lipid values and mean contaminant concentrations did not vary significantly by collection year; and overall, the POPs levels were low (many analytes were < LOQ). For example, 47% of muscle samples had no detectable DDTs, and 26% had no detectable chlordanes (Table 1). While most muscle samples had measurable concentrations of HCB, dieldrin, ΣHCHs, and ΣPCBs, these concentrations were very low (< 10 ng/g, ww). Because there were no significant differences by year for any POPs class in muscle samples, no further statistical analyses (e.g., GLM, PIA analysis) on muscle contaminants were performed. Overall, ANOVAs found that mean percent lipid values did vary significantly by collection season ($p = 0.0054$), with animals collected in spring (March and April) having very slightly higher mean lipid content compared with animals collected in fall (September and October) (84% vs 81%). In contrast, the mean POP concentrations did not vary significantly by season ($p > 0.05$). A linear regression of lipid content by collection month also found that lipid was higher in animals harvested in the spring (March and April) ($p = 0.0158$).

Percent lipid content and wet weight means of blubber and muscle of POP classes are presented in Table 1. Blubber and muscle concentrations reported by Hoekstra et al., 2002b, 2003b, 2005 and Welfinger-Smith et al. (2011) are shown for comparison. Blubber concentrations of POPs in bowheads collected between 1997 and 2000 are higher than those collected 2006–2015. Rank order of POPs concentrations in bowhead whale blubber collected 2006–2015 was ΣHCHs > ΣCHLs ~ HCB > ΣPCBs > ΣDDTs ~ dieldrin, which is different from the order reported for bowhead blubber samples collected from 1997 to 2000 (Hoekstra et al., 2005; Hoekstra et al., 2003b; Hoekstra et al., 2002b; O'Hara et al., 1999) for which the order was ΣPCBs > ΣDDTs ≥ ΣHCHs ≥ ΣCHLs. In muscle samples, our study showed lower levels, on a wet weight basis, of all contaminants compared with the previous studies, with the exception of ΣPCBs, for which our levels were 2–3 times higher, while we also observed muscle lipid levels about 2–3 times lower than those reported by Hoekstra et al. (2005). The reasons for these discrepancies are unknown. Concentrations of ΣPBDEs in blubber and muscle, which have not been previously reported for bowhead whales, are shown in Table 1. Most samples did not have measurable levels of PBDEs, but a few samples had somewhat higher concentrations, particularly blubber of two whales harvested in 2015 that had concentrations of 40 and 55 ng/g, ww. For the remaining blubber and muscle samples, ΣPBDE concentrations were ≤2 ng/g ww, of similar magnitude to concentrations in blubber of ringed seals collected in the Beaufort Sea region between 1998 and 2013 (Houde et al., 2017). For those samples with measurable concentrations of PBDEs, BDE 99 was the predominant congener, followed by BDE 47, with lesser amounts of BDEs 100, 153, 154, 49, and 155, in order of prominence.

3.2. Trends of POPs and POPs ratios in bowhead blubber collected 2006–2015

Because animal age and lipid content are variables that can influence tissue levels of POPs in cetaceans (Aguilar et al., 1999), GLM on blubber wet weight POP concentrations was carried out to assess the interactions of collection year, collection month, age (using body length as a proxy), and lipid content for each contaminant class (HCB, dieldrin ΣHCHs, ΣCHLs, ΣDDTs, ΣPCBs). In addition, particular ratios of POPs provide information on relative length in time since significant inputs into the environment, and are a measure of degradation of the initial products used. Thus, several ratios of individual analytes to sums of their contaminant classes or to other analytes were calculated (e.g. oxychlordane and *trans*-nonachlor to ΣCHLs, CB153 to ΣPCBs, α-HCH to β-HCH and *p,p′*-DDE to ΣDDTs). The ratios of α-HCH to β-HCH and

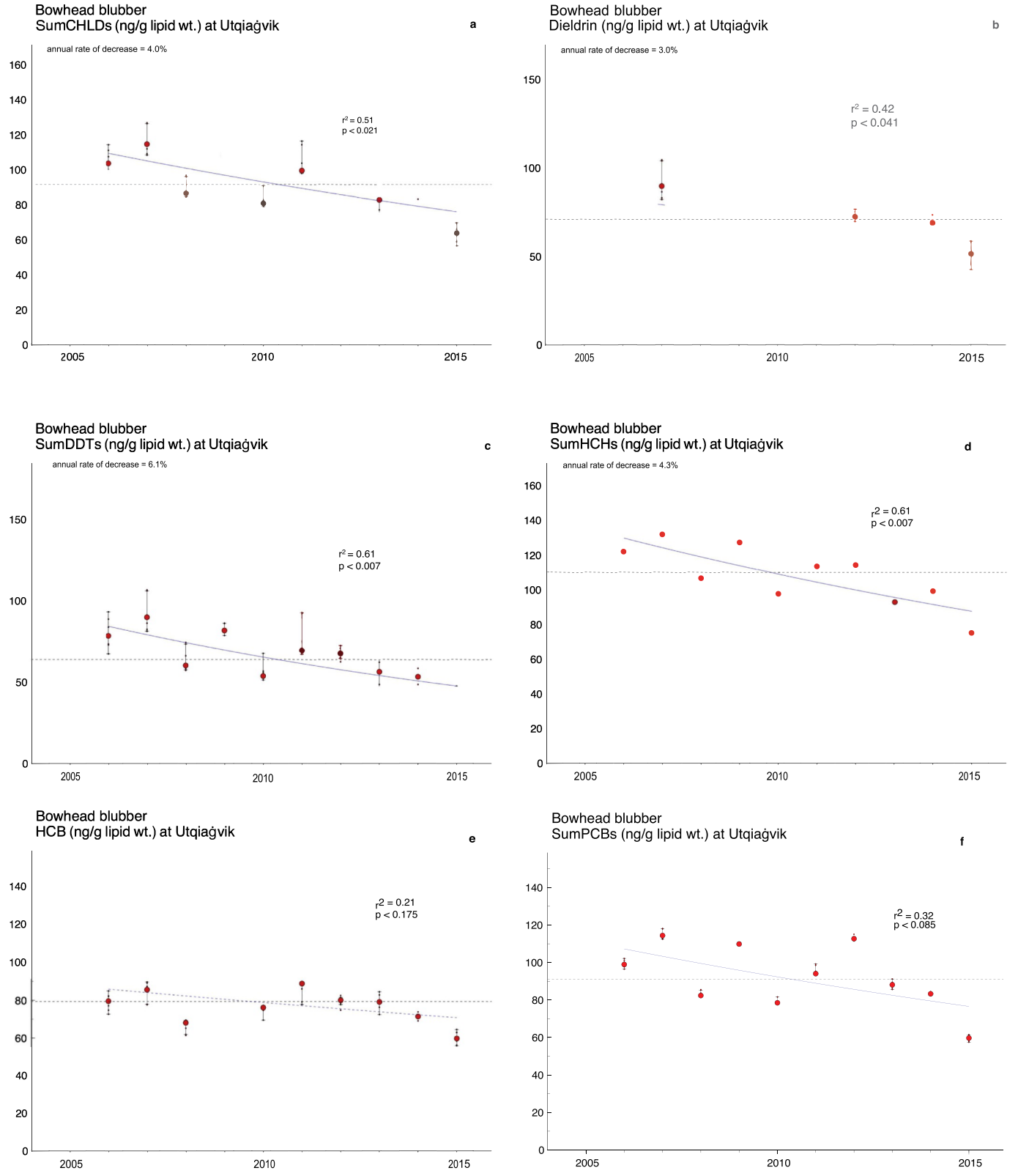

Fig. 2. Time series of POPs in blubber of male bowhead whales landed at Utqiaġvik, Alaska, 2006–2015, adjusted for body length, as assessed by PIA trends analysis package.
Points represent annual median values. PIA trends analysis package available from [www.amap.no/documents/doc/pia-application-version-051113/1026].

p,p′-DDE to ΣDDTs demonstrated differences by collection year, and as a result these were also included as dependent variables in the GLMs. The best-fit models obtained from GLM analyses are summarized in Table 2. Collection year was determined to be the best predictor of concentrations for most POP classes, though in some instances (dieldrin, ΣCHLs, ΣDDTs and *p,p′*-DDE/ΣDDTs) there were significant interactions with length (age). Seasonality was investigated by including collection month in the GLMs, but the only POP for which collection

Table 3
Annual trends of POPs in blubber of male bowhead whales landed at Utqiaġvik (Barrow), Alaska 2006–2015.

POP	Trend	PIA analysis			GLM analysis	
		Annual rate (%)	95% CI	*p*	Annual rate (%)	*p*
ΣDDTs	↓	**−6.1**	−9.9, −2.2	**< 0.007**	**−6.3**	**< 0.0001**
ΣHCHs	↓	**−4.3**	−6.9, −1.5	**< 0.007**	**−4.8**	**< 0.0001**
ΣCHLs	↓	**−4.0**	−7.0, −0.78	**< 0.021**	**−4.5**	**< 0.01**
Dieldrin	↓	**−3.0**	−5.9, −0.15	**< 0.041**	**−3.8**	**0.0124**
HCB	–	−2.0	−5.3, 1.2	< 0.175	−2.1	0.1194
ΣPCBs	–	−2.6	−8.5, 3.6	< 0.350	−2.1	0.194

PIA analysis: Lipid weight POPs, adjusted for animal length, as assessed by PIA statistical package (Bignert, 2013).
GLM: From slope of wet weight POPs regression vs. collection year in GLMs.
Bolded values are significant based on p values < 0.05.

month figured in any of the best-fit models was ΣPCBs (Table 2).

Estimated annual trends of POPs using the PIA statistical package were in good agreement with GLM regressions vs. collection year; PIA program output for lipid-corrected chlordanes, HCHs, dieldrin, DDTs, HCB and PCBs (adjusted for animal length) are shown in Fig. 2 and the temporal trends estimated using both PIA and GLMs are reported in Table 3. The annual rates reported in the following text refer to the output from the PIA analysis to facilitate comparison with trends reported by AMAP and temporal assessments used by AMAP (Riget et al., 2019; Riget et al., 2010a). Trends of these POPs over the 10-year period of the current data set appear to agree with previous reports of POPs trends in Arctic air and biota, with decreases of approximately 2–10% per year for many POPs, especially before the year 2000. Previous studies indicate that the most rapid decrease in concentrations of most POPs was observed between 1980 and 2000 (AMAP, 2016), and reflected international action restricting the production and use of POPs beginning in the 1970s and 1980s (i.e. the Stockholm Convention). As concentrations of POPs such as DDTs, dieldrin, and PCBs continued to decline, decreases in concentrations of HCHs and PBDEs were observed after 2000 due to cessation of production (e.g., addition to Annex A of the Stockholm Convention) and phase-out of use (EPA, 2017; Riget et al., 2010a; Vijgen et al., 2011), though PBDEs may still be increasing in some Arctic biota (Hoguet et al., 2013; Riget et al., 2019).

An annual decrease of 4.2% has been reported for ΣDDT concentrations (Riget et al., 2019) from long-term time series of Arctic biota, and for studies that reported *p,p′*-DDE as well, the rate of decrease for this isomer was not different from ΣDDT (Riget et al., 2019). In Alaskan murre eggs, *p,p′*-DDE decreased significantly between the 1970s and 1999–2000 (Vander Pol et al., 2004). For the bowhead whale blubber samples in the current study, robust trend analysis indicates that ΣDDTs decreased by 6.1% annually ($p < 0.007$) (Table 3). In addition, the ratio of *p,p′*-DDE to ΣDDTs is increasing, and both collection year and length appear to be significant predictors of the ratio value in the GLMs. Because *p,p′*-DDE is a recalcitrant metabolite, the ratio is expected to increase with time as historical releases of the technical DDT product are degraded over time. This trend has been observed in other Arctic biota (e.g. in ringed seals, Addison et al. (2014)). However, while the slope for collection year is positive ($p = 0.0015$) (i.e., the ratio increases over time) in the GLMs indicating a decline in "fresh" inputs to these bowhead whales, the slope for animal length is negative ($p = 0.00232$), indicating that older animals are maintaining a relatively "fresher" pattern. Bowhead whales have one of the lowest metabolic rates of any mammal (Keane et al., 2015; West et al., 2002), and health assessments of these bowheads indicated that they were all in good condition with ample blubber, which would tend to limit the amount of circulating DDTs available for biotransformation. Given the long lifespans and low metabolic rates of bowhead whales, the relatively lower ratio of *p,p′*-DDE to ΣDDTs in blubber of older animals might reflect exposure not only over some portion of the whale's lifetime, but also over the entire historical span of use of DDTs and other POPs, and indicate that relatively low levels of DDTs or other POPs may be stored within blubber lipids without undergoing extensive biotransformation after they are incorporated.

There was no significant decline in ΣPCBs over the 10-year study period. GLMs showed that collection month, lipid, and length were significant with respect to the intercept but not predictive of trends. However, comparing with data from bowhead whales collected in the 1990s, it is clear that concentrations of ΣPCBs have declined over this longer period (Fig. 1, Table 1). In addition, as levels of all POPs have declined since those previous data, the concentration ranking has also changed since the 1990s data sets, as ΣPCBs are no longer the predominant POP. Riget et al. (2019) reported ΣPCBs declines of approximately 3.7% for long-term time series data, but that short-term time series have shown a slowing of declines since 2000 (AMAP, 2016). A slowing of the rate of decline after 2000 could help explain the decrease observed in ΣPCB levels measured in whales sampled in the 1990s compared to 2006–2015 whales but no significant trend being detected over the 2006–2015 period. Furthermore, the rates of decline in PCB concentrations may be less pronounced in the western Arctic compared with areas further east (Addison et al., 2014).

In long-term time series, chlordanes (Riget et al., 2019) are decreasing at a mean annual rate of 4.6%, while we observed an annual 4.0% decrease in ΣCHL concentrations ($p < 0.021$) (Table 3). In bowhead blubber in the current data set, ΣCHLs was composed primarily of the more recalcitrant *trans*-nonachlor and heptachlor epoxide, which made up 37 and 24% of the sum, respectively, with no apparent change in the ratios of these contaminants to ΣCHLs over the sampling period. A temporal analysis showed that *cis*-nonachlor decreased significantly in Alaskan seabird eggs collected in the 1970s vs. 1999–2000 (Vander Pol et al., 2004). In the GLMs, collection year + animal length was a significant predictor of ΣCHL concentrations. GLMs showed collection year + animal length was also a significant predictor of dieldrin concentrations in the best-fit model (Table 2). Riget et al. (2019) reported a mean annual decrease of 3.0% in long-term time series data, but no detectable trends in short-term series (since 2000). However, we found a significant annual decrease in dieldrin of 3.0% ($p < 0.041$) determined by PIA analysis (Table 3).

Similar to ΣPCBs, we did not see a significant trend in concentrations of HCB over the study period, though levels in bowhead blubber appear to have decreased since the 1990s (Fig. 1, Table 1). GLMs showed that lipid, and animal length were significant with respect to the intercept but not predictive of trends. Riget et al. (2019) reported a mean annual decrease in levels of HCB in long-term time series of 2.6%, but short-term time series (since 2000) showed no trend in HCB levels, and noted the decline for HCB overall has been slower than for many other POP classes. Other studies have also reported mixed or no trends in HCB concentrations in Arctic biota, depending on the location and species (Addison et al., 2014; AMAP, 2016; Braune, 2007; Braune et al., 2005; Riget et al., 2010a). A significant declining trend was found, however, for chlorobenzenes (hexa- and pentachlorobenzene) in beluga

whales from the Chukchi Sea collected between 1989 and 2006 (Hoguet et al., 2013).

A significant declining annual trend of 4.3% ($p < 0.007$) (Table 3) was found for ΣHCHs, even though ΣHCHs are now the predominant POP measured in bowhead blubber from the western Arctic. A significant declining trend has also been found for ΣHCHs in beluga whales from the Chukchi Sea collected between 1989 and 2006 (Hoguet et al., 2013). This decline is expected to continue since HCHs have now been added to the Stockholm Convention and production has all but ceased (Vijgen et al., 2011).

The ratio of α-HCH to β-HCH is decreasing by collection year ($p < 0.0001$) (Table 2). This ratio is a measure of the length since significant fresh input of this POP class to the environment, as β-HCH is more resistant to degradation (Willet et al., 1998). The ratio is also indicative of changing inputs to the Arctic (air vs. water transport), because α-HCH is transported more rapidly via air, while β-HCH is transported predominantly via water currents, with an approximately 10-year delay (Li and Macdonald, 2005). Hoekstra et al. (2002b) reported that α-HCH was the predominant HCH isomer in 1997–1999 bowhead samples, while β-HCH was the most prominent isomer in all samples collected from 2006 through 2015, indicating a likely shift to sea current transport as the driver of input for HCHs to the Arctic since 2000.

Contrary to the trend detected with *p,p′*-DDE/ΣDDTs, where older animals showed a relatively "fresher" pattern in their blubber, animal length (age) did not appear as a factor in best-fit GLMs of α-HCH/β-HCH. The increasing trend in the ratio of β-HCH to α-HCH or to ΣHCH has been demonstrated in other studies of Arctic biota. For example, Riget et al. (2019) reported diverging trends between α-HCH and β-HCH, with α-HCH declining at 9.9% per year in biota since 2000, while β-HCH showed has shown both decreasing and increasing trends, depending on the study. Lindane (γ-HCH) showed a similar trend to α-HCH of more rapid declines (AMAP, 2016), reflecting the fact that both α-HCH and γ-HCH are more easily degraded than β-HCH and that inputs of these isomers to the Arctic are also less recent.

An analysis of seasonal variation of POPs in bowhead whales (Hoekstra et al., 2002b) showed that β-HCH fluctuated in bowhead blubber samples between spring and fall (i.e. β-HCH was higher in spring as bowheads migrated from their Bering Sea winter feeding areas). Stable isotopes of carbon were also found to fluctuate seasonally in bowhead muscle (i.e. $\delta^{13}C$ was less depleted in spring) while nitrogen stable isotope ratios remained constant, indicating that there were probably no significant seasonal trophic shifts that would account for these fluctuations (Hoekstra et al., 2002a). Hoekstra et al. (2002b) noted that PCA loadings of recalcitrant POPs such as some higher chlorinated PCB congeners and *p,p′*-DDE showed higher proportions in bowhead whales collected in spring, and hypothesized that this difference may be due to factors affecting seasonal bioaccumulation of these contaminants, such as differences in sea water concentrations of POPs between winter and summer feeding areas, or seasonal differences in lipid storage, which have been noted in other studies of seasonal factors affecting bioaccumulation of POPs in Arctic biota (Borgå et al., 2004; De Laender et al., 2010; Fisk et al., 2001). We did, in fact, find slightly higher lipid levels in bowheads collected in spring months, which may support this hypothesis. However, we did not see any significant seasonal variation in any POPs, though collection month did appear in one of the best-fit general linear models for ΣPCBs (Table 2).

It has been hypothesized that as the Arctic has undergone dramatic climatic changes over the past decade, there is now a possibility for altered pathways for introduction of POPs, as well as other environmental contaminants, into the Arctic environment. Changes in patterns of prevailing wind currents, precipitation, melting of ice containing POPs deposited over decades of atmospheric transport (Macdonald et al., 2005; McKinney et al., 2015), and increased biotransport of contaminants from migratory species are just some of possible processes affecting levels of POPs in Arctic biota with continuing climate change. Increases in anthropogenic inputs of contaminants due to increased human activity in the region may also occur (Ma et al., 2016). In addition, ecological changes to marine and terrestrial food webs are currently being observed (Cabrerizo et al., 2018; Gaden et al., 2012; McKinney et al., 2009; McKinney et al., 2015; Riget et al., 2010b), though not all of these changes are anticipated to result in increased levels of POPs. Investigation of the effects of climate variability on POPs measured in bowhead whales was beyond the scope of our study and as noted earlier, the only significant trends in POPs levels in this bowhead whale population during 2006–2015 were declines.

Concentrations of most POP classes measured in Arctic biota, including bowhead whales, have declined significantly since peak concentrations of most POPs were reached between ~1985–2005 (AMAP, 2016). International action taken to restrict the use and production of many POPs (e.g. Stockholm Convention) has been effective in driving this decline. For Arctic Inuit consumers of bowhead muktuk and meat, it is important that concentrations of POPs are quite low, and that levels are declining or stable. Levels of POPs in bowhead blubber and muscle meat are now approximately one half to one quarter what they had been in the 1990s, and many classes of these contaminants appear to be continuing to decline.

CRediT authorship contribution statement

Jennie L. Bolton: Investigation, Formal analysis, Data curation, Visualization, Writing - original draft. **Gina M. Ylitalo:** Conceptualization, Formal analysis, Data curation, Visualization, Writing - original draft, Project administration, Funding acquisition. **Paul Chittaro:** Formal analysis, Data curation, Visualization, Writing - original draft. **J. Craig George:** Investigation, Writing - review & editing. **Robert Suydam:** Investigation, Writing - review & editing. **Brian T. Person:** Investigation, Writing - review & editing. **Jonelle B. Gates:** Investigation, Writing - review & editing. **Keri A. Baugh:** Investigation, Writing - review & editing. **Todd Sformo:** Investigation, Writing - review & editing. **Raphaela Stimmelmayr:** Conceptualization, Writing - original draft, Project administration, Funding acquisition.

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Declaration of competing interest

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