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RESEARCH ARTICLE

PFAS and PBDEs in traditional subsistence foods from Sivuqaq, Alaska

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Abstract

The Arctic is a hemispheric sink for both legacy and current use persistent organic pollutants (POPs). Once in the Arctic, POPs biomagnify in food webs, potentially reaching concentrations in high trophic level animals that pose a health concern for people who subsist on those animals. Indigenous Peoples of the Arctic may be highly exposed to POPs through their traditional diets. The objective of this study was to assess concentrations of polybrominated diphenyl ethers (PBDEs) and per- and polyfluoroalkyl substances (PFAS) in tissues of traditionally harvested foods from Sivuqaq (St. Lawrence Island), Alaska. Community health researchers identified volunteer households and local hunters to donate tissues from traditionally harvested animals. Target species included bowhead whale (*Balaena mysticetus*), Pacific walrus (*Odobenus rosmarus*), ringed seal (*Pusa hispida*), bearded seal (*Erignathus barbatus*), ribbon seal (*Histriophoca fasciata*), spotted seal (*Phoca largha*), and reindeer (*Rangifer tarandus*). PBDEs were frequently detected in all species and tissues. PBDE concentrations tended to be highest in lipid-rich tissues of seals. PFAS were infrequently detected and did not show obvious patterns among species or tissues. This and other studies demonstrate that POPs such as PBDEs are present in tissues of traditional food animals from Sivuqaq, as they are throughout the Arctic, and consumption of these animals likely contributes to exposure among Arctic Indigenous Peoples.

Keywords Arctic · Indigenous · Marine mammal · Persistent organic pollutants · POPs · St. Lawrence Island

Introduction

In the northern hemisphere, the Arctic is a sink for both legacy and emergent persistent organic pollutants (POPs) (Muir and de Wit 2010; Rigét et al. 2019). Indigenous Peoples of the Arctic are exposed to high concentrations of POPs through their traditional diets (Abass et al. 2018; Donaldson et al. 2010). Sivuqaq, the traditional name for St. Lawrence Island, is the largest island in the Bering Sea and is located 61 km from the Chukotka Peninsula of Russia and 322 km from the Alaskan mainland. Sivuqaq is home to approximately 1700 Sivuqaq Yupik residents in the villages of Gambell and Savoonga. The traditional diets of coastal Alaska Natives often include long-lived or high trophic level marine mammals including bowhead whale (*Balaena mysticetus*), Pacific walrus (*Odobenus rosmarus*), and ice seal species such as ringed seal (*Pusa hispida*), bearded seal (*Erignathus barbatus*), ribbon seal (*Histriophoca fasciata*), and spotted seal (*Phoca largha*) (Welfinger-Smith et al. 2011). Previous community-based research in partnership with the leadership and communities on Sivuqaq suggests that residents are exposed to a variety of legacy and current use POPs, such as polychlorinated biphenyls (PCBs), organochlorine pesticides, polybrominated diphenyl ethers (PBDEs), and per- and polyfluoroalkyl substances (PFAS); this exposure occurs through their traditional diet, as well as from local sources of contamination including two formerly

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used defense (FUD) sites dating from the Cold War (Byrne et al. 2017; Carpenter et al. 2005; Jordan-Ward et al. 2022; von Hippel et al. 2018; Welfinger-Smith et al. 2011; Zheng et al. 2020).

PBDEs and PFAS are environmentally ubiquitous POPs (Abbasi et al. 2019; Muir et al. 2019). PBDEs were utilized as additive flame retardants in a variety of commercial products. Their production has been largely phased out and regulated, although they persist in products in use and in waste streams (Abbasi et al. 2015). All three technical formulations, penta-BDE, octa-BDE, and deca-BDE, are globally restricted under the Stockholm Convention on Persistent Organic Pollutants due to evidence of persistence, toxicity, long-range transport, and bioaccumulation in humans and animals (UNEP 2020). However, a large reservoir of PBDEs exists in products manufactured before the phaseout (Abbasi et al. 2015). PBDEs bioaccumulate and biomagnify in Arctic food webs (Sørmo et al. 2006). Due to declines in production and use, most PBDE concentrations are stable or declining in Arctic biota over time (Rigét et al. 2019; Rotander et al. 2012b). PFAS represent a diverse group of more than 4700 recent and current use compounds, some of which are regulated as POPs (OECD/UNEP 2018; UNEP 2020). While perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) concentrations are generally declining over time in the Arctic, unregulated and replacement PFAS contribute to ongoing exposure (Butt et al. 2014; Liu and Mejia Avendaño 2013). Numerous PFAS are present in Arctic biota, and some PFAS have shown increasing trends in the Arctic (Muir et al. 2019).

This study builds on prior studies of PBDE and PFAS exposure on Sivuqaq (Byrne et al. 2017; Byrne et al. 2018a, b). Previous research on Sivuqaq demonstrated that both PFAS and PBDEs are present in ninespine stickleback (*Pungitius pungitius*, hereafter "stickleback") and Alaska blackfish (*Dallia pectoralis*) collected on Sivuqaq (Byrne et al. 2017; Zheng et al. 2020). The presence of PFAS and PBDEs in fish collected from remote sites on Sivuqaq suggests some exposure is due to atmospheric deposition; however, especially high concentrations were detected in fish collected in Troutman Lake adjacent to the Village of Gambell (Byrne et al. 2017; Zheng et al. 2020). These results suggest that the village, its associated landfill, or the Gambell formerly used defense (FUD) site is a point source of these compounds (Byrne et al. 2017). Research in the Antarctic has confirmed that the built environment acts as a point source of both PBDEs and PFAS in polar regions (Wild et al. 2015). Exposure to POPs through both traditional foods and local hotspots of pollution remains a serious health concern for Indigenous People of the circumpolar Arctic, including the Yupik People of Sivuqaq. Indeed, we found multiple associations between serum PFAS and PBDE concentrations and circulating concentrations of thyroid-stimulating hormone (TSH), thyroxine (T_4), and triiodothyronine (T_3) in Sivuqaq residents, demonstrating a health consequence to exposure (Byrne et al. 2018a, b). The aim of the current study is to assess the presence and concentrations of PBDEs and PFAS in frequently consumed traditional subsistence foods on Sivuqaq to provide residents with the information they need to make informed decisions and to advocate for policies that reduce pollution in the Arctic. This study also contributes to our knowledge of pollutant levels in a relatively poorly studied region of the Arctic.

Methods

This research is part of an ongoing community-based participatory research (CBPR) project (Miller et al. 2013). We collected the subsistence food samples during the months of November and December of 2016. Community health researchers identified volunteer households and local hunters who donated tissues from harvested animals. Harvested animals included bowhead whale, bearded seal, ringed seal, spotted seal, Pacific walrus, and reindeer (*Rangifer tarandus*). No animals were harvested for the purpose of providing samples for this research. We obtained a total of 50 samples of the commonly consumed organs, meat, rendered oils, and blubber of these species (National Marine Fisheries Service Permit No. 18978). Twenty-five samples were collected in Savoonga and 25 in Gambell. Samples included bowhead whale blubber ($n=3$), rendered oil ($n=2$), mangtak (skin + blubber; $n=8$), muscle ($n=1$); reindeer fat ($n=5$), fat and muscle ($n=5$), muscle ($n=3$); bearded seal blubber ($n=1$), muscle ($n=1$); spotted seal blubber and meat ($n=4$), muscle ($n=1$); ringed seal blubber and muscle ($n=3$), muscle ($n=3$); rendered oil from unidentified seal species ($n=6$); and spotted seal liver, heart, intestine, and kidney ($n=1$ of each).

Strict precautions were taken to prevent contamination from the tools used for sampling. Samples of traditional foods were resected with a clean stainless steel knife to access tissue that had not previously been exposed to the external environment. Aluminum foil was used to cover cutting surfaces. Samples were wrapped in clean aluminum foil, placed in a sealable plastic bag, and frozen at − 20 °C. Some samples were traditionally prepared subsistence foods rather than tissue samples. Bowhead whale samples included a combination of skin and blubber (mangtak), and rendered oil from whale and seal blubber was also analyzed. Rendered oil was produced in the homes of Sivuqaq residents. Rendered oil is typically produced by allowing blubber to sit in a cool dark location in the home or through heating the blubber. Rendered oil included a combination of blubber from multiple seal species. Community health researchers completed a sample record sheet for every sample. The record sheet

included the following: species and tissue, date of harvest, approximate location of harvest, method of transport for the harvested sample, and any information on animal condition. In addition, the date of sample collection, the weight of the sample, and information on previous storage conditions were collected. If the sample was a traditional preparation (rendered oil), then information about the preparation was also recorded.

Samples were stored at − 20 °C while we completed the process of getting a CITES permit to export the samples from Anchorage, Alaska to SGS AXYS Analytical Services Ltd. (AXYS) in British Columbia, Canada. The 50 samples were exported to AXYS in Canada in March 2017. PFAS concentrations were quantified using reverse-phase high-performance liquid chromatography–mass spectrometry on a Waters 2690 (Waters, Milford, MA) coupled to a Micromass Quattro Ultima MS/MS (Waters, Milford, MA), using AXYS method MLA-043 (SGS-AXYS Analytical Services Ltd 2017a). Briefly, samples were homogenized and isotopically labeled surrogate standards were added prior to extraction with methanolic potassium hydroxide solution, with acetonitrile, and finally with methanolic potassium hydroxide solution, each time collecting the supernatants. The supernatants were combined, treated with ultrapure carbon powder, and evaporated to remove methanol. The resulting solution was diluted with water and cleaned up by solid-phase extraction (SPE) on a weak anion exchange sorbent. The eluate was spiked with recovery standards and analyzed on an ultrahigh performance liquid chromatography reversed-phase C18 column using a solvent gradient. The column was coupled to a triple quadrupole mass spectrometer run at unit mass resolution in the multiple reaction monitoring (MRM) in negative electrospray ionization mode. The instruments were calibrated with a minimum of five calibration standards. At minimum, blanks and matrix spike samples were analyzed with each analytical batch of 20 samples. Sample-specific method detection limits were defined as the concentration associated with 3 times the instrumental noise, or the lowest calibration standard. Detection limits for PFAS were typically < 1 ng/g and not above 2 ng/g.

PBDE concentrations were determined using isotope dilution high-resolution gas chromatography high-resolution mass spectrometry with a Hewlett Packard 5890 (Hewlett Packard, Palo Alto, CA) coupled to a Micromass Ultima (Waters, Milford, MA), according to EPA Method 1614A (SGS-AXYS Analytical Services Ltd 2017b). The instruments were calibrated with a minimum of six calibration standards. Sample matrices were extracted using the same procedures that are used for the extraction of PCB congeners by EPA Method 1668A. Prior to extraction, each sample was spiked with an aliquot of a surrogate standard solution containing a suite of $^{13}C_{12}$-labeled BDE. Homogenized samples were mixed with anhydrous sodium sulfate and extracted with methylene chloride in a Soxhlet extractor. The extract was spiked with an aliquot of cleanup surrogate solution also containing $^{13}C_{12}$-labeled BDE prior to chromatographic cleanup procedures. The extract was cleaned up on a BE 8 g 44%/Florisil column and alumina column. An aliquot of recovery standard solution containing $^{13}C_{12}$-labeled BDEs was added to the extract prior to analysis by high-resolution GC/MS. All analysis procedures were conducted under low light levels. Aluminum foil shields were used to protect the samples from ambient lighting. Lipid content was determined gravimetrically from the sample extract prior to cleanup processes. The lipid content was assessed in duplicate and reported as the average of the percent lipid from the two analyses. Sample-specific method detection limits were defined as the concentration associated with 2.5 times the instrumental noise. Detection limits for most PBDEs were < 2 pg/g ww. Heavily brominated BDEs (206, 207, 208) had higher detection limits, typically below 10 pg/g ww, and BDE-209 had sample-specific detection limits typically below 40 pg/g and never above 100 pg/g.

This study assessed the presence of 13 PFAS and 40 PBDEs in traditional subsistence foods. Analyzed PFAS include perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoA), perfluorobutanesulfonic acid (PFBS), perfluorohexanesulfonic acid (PFHxS), perfluorooctanesulfonic acid (PFOS), and perfluorooctanesulfonamide (PFOSA). Analyzed PBDE congeners include BDE 7, BDE 8 + 11, BDE 10, BDE 12 + 13, BDE 15, BDE 17 + 25, BDE 28 + 33, BDE 30, BDE 32, BDE 35, BDE 37, BDE 47, BDE 49, BDE 51, BDE 66, BDE 71, BDE 75, BDE 77, BDE 79, BDE 85, BDE 99, BDE 100, BDE 105, BDE 116, BDE 119 + 120, BDE 126, BDE 128, BDE 138 + 166, BDE 140, BDE 153, BDE 154, BDE 155, BDE 181, BDE 183, BDE 190, BDE 203, BDE 206, BDE 207, BDE 208, and BDE 209. With the exception of a single matrix spike recovery of 126% for PFDoA, recoveries of PFAS from matrix spike samples were all ± 20%, and most were ± 10%. For PBDEs, two matrix spike samples had 126% recovery for BDE 209. One matrix spike sample had a 121% percent recovery for BDE 47. The remaining matrix spike samples had recoveries all ± 20%. Descriptive statistics were calculated in Excel (Microsoft Corporation, Redmon, Washington) and R using tidyverse (R Development Core Team 2010; Wickham et al. 2019). Figures were produced with ggplot2 and ggbreak (Xu et al. 2021). Non-detects were treated as zero for the purposes of descriptive statistics. Contaminant concentrations are presented in wet weight (ww). When making comparisons to studies that only present lipid weight (lw) results, we convert ww to lw using the lipid content of the sample.

Results

At least one PBDE congener was detected in every traditional food sample. BDEs 28/33, 47, 99, 100, 153, 154, and 209 were detected in 100% of traditional food samples; BDE 183 was present in 95% of samples (Table 1). These congeners also tended to make up relatively large percentages of the total PBDE concentration in each sample. BDEs 49, 66, 206, and 208 were present in at least 58% of traditional food samples, with the remaining PBDEs detected less frequently. In marine species, lipid-rich tissues tended to have a greater number of detectable PBDEs and higher concentrations of PBDEs. Reindeer muscle tissue had relatively high concentrations of PBDEs (Fig. 1).

Sample sizes for seals were small, and no clear trends emerged between seal species (Suppl. Table 1). Therefore, data for seal species are reported together. Collectively, the seal species had the highest median PBDE concentration compared to other species. Eight BDE congeners were present in all seal muscle samples (BDE 28/33, 47, 99, 100, 153, 154, 183, and 209). BDEs 47, 99, and 209 often made up the majority of overall PBDE concentrations in seal tissues (Fig. 1). Blubber and rendered oil contained the highest median (2394.5 pg/g ww) and maximum (4700.8 pg/g ww) Σ-PBDE concentrations of the seal tissues (Table 1). A greater diversity of PBDEs was present in these lipid-rich tissues. Only eight BDEs were detected in muscle samples, compared to a median of 29 and a maximum of 32 congeners detected in blubber and rendered oil.

In bowhead whales, PBDE concentrations were highest in blubber and rendered oil, followed by mangtak, and finally muscle (Table 1). A median of 22 and a maximum of 26 congeners were detected in bowhead whale samples. The median Σ-PBDE concentrations in muscle, mangtak, and blubber or rendered oil were 320.4, 312.7, and 650.7 pg/g ww, respectively. Lipid-rich tissues tended to have higher concentrations of PBDEs than muscle or mangtak (Fig. 1). The bowhead muscle sample had the greatest number of detectable PBDEs (26 congeners), but the Σ-PBDE concentration was below the median value for other bowhead tissue samples. The highest Σ-PBDE concentration in bowhead whale samples was 2089.3 pg/g ww in a rendered oil sample.

A relatively small number of PBDEs were detected in reindeer samples as compared to marine mammal samples. Seven PBDE congeners were present in all reindeer samples regardless of tissue (BDE 28/33, 47, 99, 100, 153, 154, and 209). BDE-183 was present in 11 of 12 reindeer samples, but not detected in one fat sample. A maximum of eight PBDE congeners was detected in reindeer samples. Median concentrations of Σ-PBDEs in reindeer muscle, muscle and fat, and fat were 611.5, 268.4, and 325.1 pg/g ww, respectively. While concentrations of PBDEs in reindeer were not uniformly higher than in other species, one reindeer muscle and fat sample contained a BDE 209 concentration of 35,600 pg/g ww, contributing to a Σ-PBDE concentration of 36,240.7 pg/g ww (Fig. 1). This was the highest Σ-PBDE concentration detected in the study.

Of 13 analyzed PFAS, only nine were detectable in any of the samples. Overall PFAS concentrations were highest in seal tissues (Fig. 2). In seal tissue samples, PFPeA was the most commonly detected PFAS, and was present at the highest median concentration of any PFAS (20.2 ng/g ww) (Table 1). PFPeA was also the compound present at the highest maximum concentration (36.8 ng/g ww), which occurred in a mixed blubber and muscle sample from a spotted seal. PFAS were most frequently detected and present at the highest concentrations in seal tissues compared to other species (Fig. 2).

PFPeA, PFHxA, and PFUnDA were all detected in the single muscle tissue sample from a bowhead whale. PFHxA was present at the highest concentration of the three (7.54 ng/g ww). PFPeA was detected in 25% of bowhead whale mangtak samples. No PFAS were detected in bowhead whale blubber or rendered oil (Table 2).

In reindeer samples, PFOS, PFNA, PFOA, and PFPeA were sporadically detected, without a clear pattern. PFOA and PFOS were detected in 33% of muscle samples, with a maximum of 2.56 and 1.02 ng/g ww, respectively. PFPeA was detected in 20% of fat and mixed muscle and fat samples, with a maximum concentration of 3.54 ng/g ww in fat. PFNA was detected in a single mixed muscle and fat sample at a concentration of 9.85 ng/g ww (Table 2).

Multiple tissues (heart, liver, kidney, and intestine) from a single juvenile spotted seal allowed the comparison of PFAS and PBDE concentrations across tissues (Fig. 3). Concentrations of PBDEs in organs of the juvenile seal were substantially lower than those found in samples of other tissues from adult seals (Table 3). BDEs 47, 99, and 209 were the predominant congeners in the juvenile samples (Table 3). Long-chain perfluoroalkyl acids (PFAAs), those with ≥ 7 perfluorinated carbons, were present at relatively high concentrations in the juvenile seal organs compared to tissues from other seals (Table 3). PFOS, PFHxA, PFNA, and PFUnDA were the predominant compounds in seal organ samples; PFPeA was not detected in seal organs. However, total PFAS concentrations were higher in seal muscle than in other tissues due to the relatively high concentrations of PFPeA (Fig. 2).

Discussion

PBDE concentrations in traditional food samples from this study are within the range of those reported elsewhere in the Arctic. The median Σ-PBDE concentration in seal blubber

Table 1 PBDE concentrations (pg/g ww) in tissues of traditional food animals from Sivuqaq, Alaska

	BDE-28 + 33	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	Σ-PBDE
Reindeer									
Muscle [$n=3$]: median (range)	1.9 (1, 3.2)	32.8 (30.5, 221)	37.7 (31, 214)	6.6 (5.4, 42.2)	6.4 (5.9, 18.8)	3.1 (2.7, 15.1)	0.9 (0.5, 2)	96.3 (28.9, 1730)	611.5 (116.4, 1809.8)
% detect	100	100	100	100	100	100	100	100	100
Reindeer fat and muscle [$n=5$]: median (range)	3.2 (0.7, 5.9)	88.2 (24, 267)	74.9 (24.8, 259)	14.5 (5.6, 50.9)	10.8 (4.4, 28.5)	5.1 (2.7, 19.9)	1.8 (0.9, 9.5)	72.3 (44.7, 35,600)	268.4 (129.2, 36,240.7)
% detect	100	100	100	100	100	100	100	100	100
Reindeer fat [$n=5$]: median (range)	2.8 (2.5, 6)	85.8 (68.1, 105)	84.9 (69.6, 101)	16.2 (12.8, 18.8)	12.7 (11.5, 16.3)	6.3 (5.3, 7.7)	2.4 (0, 2.6)	116.0 (86.7, 127)	325.1 (295.2, 355.5)
% detect	100	100	100	100	100	100	100	100	100
Seal									
Muscle [$n=5$]: median (range)	10.4 (7.7, 39.4)	321.0 (272, 555)	70.5 (13.2, 642)	39.9 (15.6, 124)	9.5 (4.7, 63.4)	12.4 (3.9, 51.4)	0.7 (0.6, 2.5)	41.5 (18.1, 133)	694.5 (342.6, 1426.5)
% detect	100	100	100	100	100	100	100	100	100
Seal muscle and blubber [$n=7$]: median (range)	30.9 (9.6, 86.8)	890.0 (215, 1070)	72.8 (29.4, 236)	55.4 (16.8, 92.3)	16.1 (4.6, 56)	13.8 (2.9, 176)	1.8 (1, 5.6)	80.7 (66.6, 412)	1343.4 (439.3, 2140.5)
% detect	100	100	100	100	100	100	100	100	100
Seal blubber or rendered oil [$n=7$]: median (range)	49.7 (17.8, 133)	1340.0 (381, 2750)	287.0 (52, 1130)	122.0 (19.8, 291)	44.8 (9.8, 131)	64.1 (3.9, 156)	4.7 (2.3, 8)	118.0 (111, 255)	2394.5 (628.6, 4700.8)
% detect	100	100	100	100	100	100	100	100	100
Bowhead whale									
Muscle [$n=1$]: median (range)	1.6 (NA)	102.0 (NA)	123.0 (NA)	22.2 (NA)	13.4 (NA)	9.8 (NA)	0.9 (NA)	31.3 (NA)	328.4 (NA)
% detect	100	100	100	100	100	100	100	100	100
Mangtak (blubber and skin) [$n=8$]: median (range)	4.7 (2.4, 6.1)	52.7 (48.9, 97.8)	28.1 (23, 68.2)	7.3 (6.1, 16.6)	3.2 (2.4, 7.8)	3.2 (2.2, 11.5)	1.7 (ND, 2.7)	146.0 (77.7, 345)	312.7 (210.9, 621.4)
% detect	100	100	100	100	100	100	87.5	100	100
Blubber or rendered oil [$n=5$]: median (range)	20.6 (10.1, 22.7)	233.0 (125, 537)	121.0 (62.4, 642)	30.6 (16.6, 128)	10.5 (5.1, 62.3)	11.2 (6.2, 52)	3.7 (1.8, 4.9)	151.0 (105, 382)	656.7 (436.9, 2089.3)
% detect	100	100	100	100	100	100	100	100	100

Individual data for PBDEs detected in 90% of samples. Σ-BDE includes all detected PBDEs. *NA* not applicable. *ND* not detected (< method detection limit)

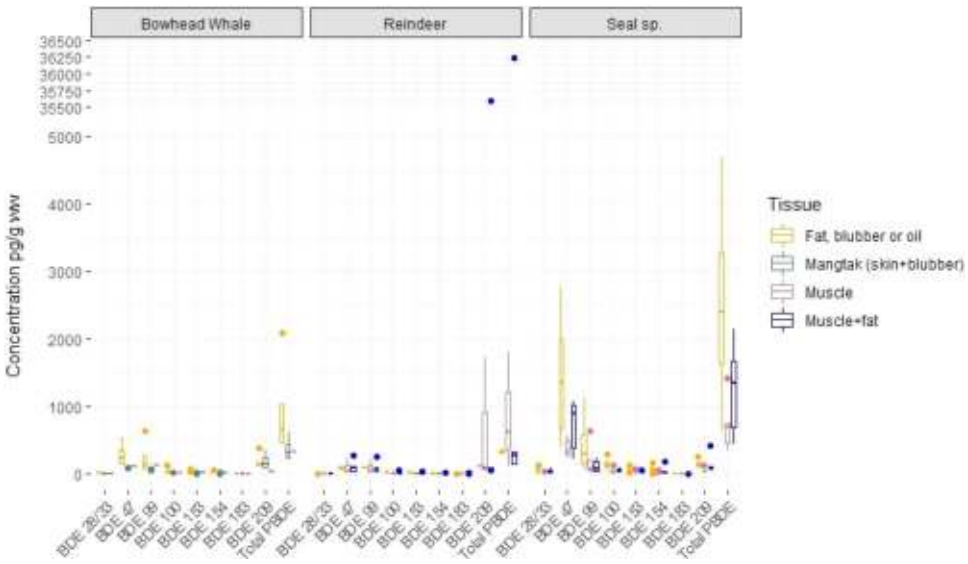

Fig. 1 PBDE concentrations (pg/g ww) in tissues of traditional food animals from Sivuqaq, Alaska. Boxplots present median and interquartile range (IQR) and whiskers extend to 1.5 × IQR; data outside 1.5 × IQR are plotted as individual data points

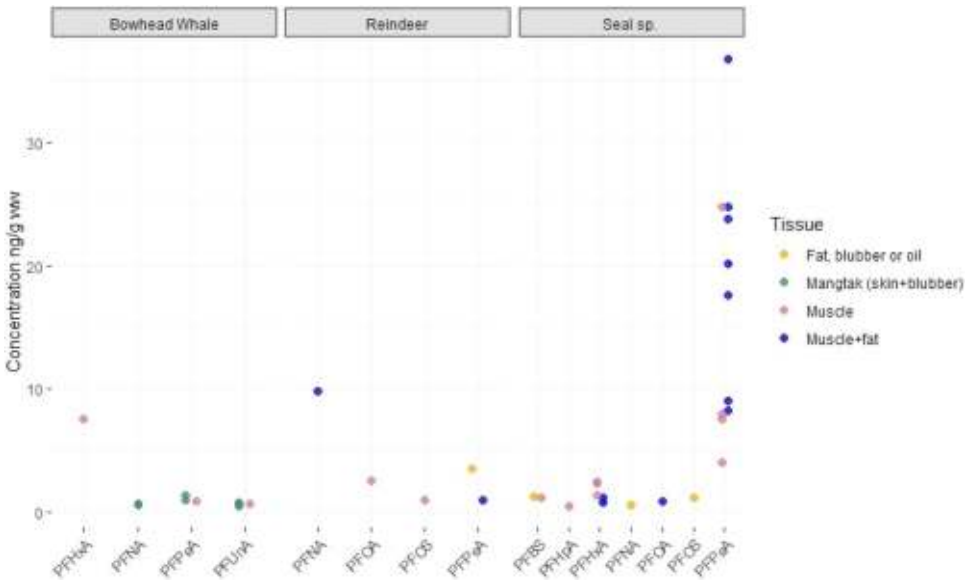

Fig. 2 PFAS concentrations (ng/g ww) in tissues of traditional food animals from Sivuqaq, Alaska. All concentrations greater than the limit of detection presented

or rendered oil from this study (2394.5 pg/g ww) was lower than the concentration reported in harbor seals collected in Alaska in 2000–2002 (14,600 pg/g ww) (Wang et al. 2012). Similarly, blubber from harp seals (*Pagophilus groenlandicus*) and hooded seals (*Cystophora cristata*) stranded in the Northeastern USA between 2000 and 2010 had mean Σ-PBDE concentrations of 70,550 and 94,2800 pg/g ww, respectively (Soulen et al. 2018). Comparatively lower concentrations in the current study may be due in part to overall decreasing trends of PBDE concentrations in the Arctic resulting from global regulation. In addition, POP concentrations are generally higher on the North Atlantic coast of North America as compared to central Canada or Alaska (Braune et al. 2005). The mean Σ-PBDE concentrations in ringed seals collected from several locations in the Canadian Arctic in 2013 included 1.44 ng/g lw in Sachs Harbour, 5.96 ng/g lw in Resolute Bay, and 5.05 ng/g lw in Nain (Houde et al. 2017). These are similar values to the mean Σ-PBDE concentration of 2.37 ng/g lw in seal blubber or rendered oil from the current study.

Bolton et al. (2020) reported mean Σ-PBDE concentrations of 7 ng/g ww in bowhead whale blubber and 0.15 ng/g ww in muscle tissue collected in 2015 from Utqiaġvik (formerly Barrow), Alaska, with detection rates of 50% and 71%, respectively. Comparatively, mean Σ-PBDE concentrations in bowhead whale blubber or rendered oil were under 1 ng/g ww in the current study, despite higher detection rates and a larger number of target PBDEs. BDEs 47 and 99 were the predominant congeners in bowhead whales harvested from Utqiaġvik. Bolton et al. (2020) did not determine BDE 209

Table 2 PFAS concentrations (ng/g ww) in tissues of traditional food animals from Sivuqaq, Alaska

	PFPeA	*PFHxA*	*PFHpA*	*PFOA*	*PFNA*	*PFDA*	*PFUnDA*	*PFBS*	*PFOS*
Reindeer									
Muscle [$n=3$]: median	ND	ND	ND	ND	ND	ND	ND	ND	ND
(range)	ND	ND	ND	(ND, 2.56)	ND	ND	ND	ND	(ND, 1.02)
% detect	ND	ND	ND	33.33	ND	ND	ND	ND	33.33
Reindeer fat and muscle	ND	ND	ND	ND	ND	ND	ND	ND	ND
[$n=5$]: median	(ND, 0.94)	ND	ND	ND	(ND, 9.85)	ND	ND	ND	ND
(range)									
% detect	20	ND	ND	ND	20	ND	ND	ND	ND
Reindeer fat [$n=5$]: median	ND	ND	ND	ND	ND	ND	ND	ND	ND
(range)	(ND, 3.54)	ND	ND	ND	ND	ND	ND	ND	ND
% detect	20	ND	ND	ND	ND	ND	ND	ND	ND
Seal (bearded, spotted, ringed)									
Muscle [$n=5$]: median	7.54	1.42	ND	ND	ND	ND	ND	ND	ND
(range)	(ND, 24.8)	(ND, 2.48)	(ND, 0.53)	ND	ND	ND	ND	(ND, 1.15)	ND
% detect	80	60	20	ND	ND	ND	ND	20	ND
Seal muscle and blubber	20.2	ND	ND	ND	ND	ND	ND	ND	ND
[$n=7$]: median	(8.22, 36.8)	(ND, 1.15)	ND	(ND, 0.89)	ND	ND	ND	ND	ND
(range)									
% detect	100	28.57	ND	14.29	ND	ND	ND	ND	ND
Seal blubber or rendered oil	ND	ND	ND	ND	ND	ND	ND	ND	ND
[$n=7$]: median	ND	ND	ND	ND	(ND, 0.6)	ND	ND	(ND, 1.29)	(ND, 1.14)
(range)									
% detect	ND	ND	ND	ND	14.29	ND	ND	14.29	14.29
Bowhead whale									
Muscle [$n=1$]: median	0.85	7.54	ND	ND	ND	ND	0.67	ND	ND
(range)	(NA)	(NA)	(NA)	(NA)	(NA)	(NA)	(NA)	(NA)	(NA)
% detect	100	100	ND	ND	ND	ND	100	ND	ND
Mangtak (blubber and skin)	ND	ND	ND	ND	ND	ND	ND	ND	ND
[$n=8$]: median	(ND, 1.38)	ND	ND	ND	(ND, 0.66)	ND	(ND, 0.74)	ND	ND
(range)									
% detect	25	ND	ND	ND	25	ND	37.5	ND	ND
Blubber or rendered oil [$n=5$]:	ND	ND	ND	ND	ND	ND	ND	ND	ND
median	ND	ND	ND	ND	ND	ND	ND	ND	ND
(range)									
% detect	ND	ND	ND	ND	ND	ND	ND	ND	ND

PFBA, PFDoA, PFHxS, PFOSA analyzed but not detected. *NA* not applicable. *ND* not detected (< method detection limit)

concentrations; however, in whales harvested from Sivuqaq, BDE 209 was frequently present at concentrations comparable to BDEs 47 and 99. The mean Σ-PBDE concentration of 1063 pg/g lw in bowhead whale blubber from Sivuqaq is below previously reported Σ-PBDE concentrations in North Atlantic fin whale blubber, which ranged from 41,000 to 82,000 pg/g lw (Rotander et al. 2012a).

PBDE concentrations in reindeer from Sivuqaq are within the range of those reported elsewhere in the Arctic. In semi-domesticated Norwegian reindeer, BDE 47 concentrations ranged from non-detect to 1.05 ng/g ww in muscle, and non-detect to 4.15 ng/g ww in fat, which are above the concentrations reported in the current study (Hassan et al. 2013). Mean Σ-PBDE concentrations of 60 pg/g ww were reported in Finnish reindeer muscle collected in 2006 (Suutari et al. 2011), which is lower than those in the current study. The mean Σ-PBDE concentration in semi-domesticated reindeer muscle, collected between 2006 and 2013 in Finland and Russia, was also 60 pg/g ww (Holma-Suutari et al. 2016).

Although seals tended to contain the highest concentrations of PBDEs, one sample of reindeer fat and muscle contained a BDE 209 concentration of 35,600 pg/g ww, contributing to the single highest total PBDE concentration of 36,240 pg/g ww in any tissue from any species examined in this study (Fig. 1). We found that BDEs 47, 99, and 209 tended to make up the majority of the Σ-PBDE concentration, and were often equivalent in concentration. However, BDE 209 made up more than 98% of the Σ-PBDE concentration in the outlier reindeer sample. This is similar to muscle tissue from semi-domesticated reindeer harvested in Finland,

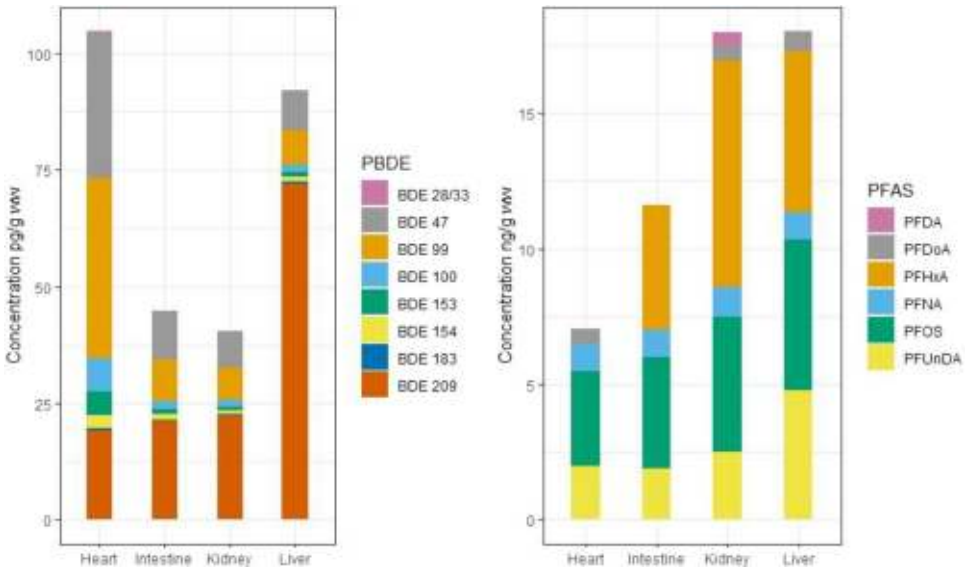

Fig. 3 Relative concentrations of PBDEs and PFAS in organ tissues of a single juvenile spotted seal

Table 3 Concentrations and relative percentage of PBDEs and PFAS in organ tissues of a single juvenile spotted seal

	Kidney		*Liver*		*Heart*		*Intestine*	
PBDE (pg/g ww)								
BDE-28 + 33	ND	0.0%	ND	0.0%	0.44	0.4%	0.47	1.0%
BDE-47	7.77	19.2%	8.61	9.3%	31.10	29.7%	10.00	22.3%
BDE-99	6.91	17.1%	7.30	7.9%	38.70	36.9%	8.88	19.8%
BDE-100	1.54	3.8%	1.61	1.7%	7.13	6.8%	1.95	4.3%
BDE-153	0.78	1.9%	1.02	1.1%	4.87	4.6%	0.92	2.0%
BDE-154	0.72	1.8%	1.01	1.1%	2.88	2.7%	1.25	2.8%
BDE-183	0.35	0.9%	0.65	0.7%	0.43	0.4%	0.23	0.5%
BDE-209	22.30	55.2%	71.90	78.1%	19.20	18.3%	21.20	47.2%
Σ-*PBDE*	40.37		92.10		104.74		44.89	
PFAS (ng/g ww)								
PFHxA	8.37	46.5%	5.92	32.8%	0.00	0.0%	4.59	39.5%
PFNA	1.08	6.0%	1.01	5.6%	1.01	14.4%	1.04	9.0%
PFDA	0.51	2.8%	ND	0.0%	ND	0.0%	ND	0.0%
PFUnDA	2.50	13.9%	4.77	26.5%	1.97	28.0%	1.89	16.3%
PFDoA	0.55	3.1%	0.74	4.1%	0.53	7.6%	ND	0.0%
PFOS	4.99	27.7%	5.58	31.0%	3.52	50.1%	4.09	35.2%

NA not applicable. *ND* not detected. % is column percentage, or relative contribution within tissue

in which BDE 209 made up over 90% of the total concentration (Holma-Suutari et al. 2014). The Σ-PBDE concentrations in the Finnish reindeer tissues were in the low ng/g lw range, similar to the median Σ-PBDE concentration of 7 ng/g lw in the current study (Holma-Suutari et al. 2014).

PFAS concentrations documented in this study are on the low end of the ranges reported in other studies of Arctic biota. The Σ-PFAS concentration in East Greenland ringed seal blubber collected in 2011–2012 was 1 ng/g ww, with higher detection rates for long-chain PFAAs (Boisvert et al. 2019). Historically, high production volume compounds (PFOA, PFOS) as well as long-chain PFAAs were frequently detected in Arctic biota (Butt et al. 2010; Muir et al. 2019). However, the congener composition of PFAS in seal samples from this study is different than those reported elsewhere in the Arctic. Specifically, PFPeA was the most frequently detected and highest concentration PFAS in any seal tissue, followed by PFHxA. We are not aware of any other study reporting PFPeA as a predominant compound in Arctic biota. In general, shorter chain carboxylated PFAAs tend to be less bioaccumulative than those with longer chains (Wen et al. 2019). Zhang et al. (2019) noted relatively high

bioaccumulation factors for PFPeA within plankton which they attributed to exposure to precursor compounds such as 6:2 fluorotelomer sulfonate. Seals may accumulate PFPeA as the result of biotransformation of precursors. Research in harbor seals (*Phoca vitulina*) suggests that PFAS differentially partition into tissue compartments (Van de Vijver et al. 2005). It may be that PFPeA is preferentially accumulating in muscle; however, relatively little is known about the partitioning dynamics of PFAS within tissues (De Silva et al. 2021). It is also possible that locally harvested animals are impacted by a point source of PFPeA on Sivuqaq, such as the unlined landfill.

In addition, the composition of PFAS compounds in seal samples was dissimilar to the relative concentrations of PFAS in human sera or stickleback tested on Sivuqaq (Byrne et al. 2017). Specifically, both humans and stickleback appear to accumulate longer chain PFAS and show a tendency to accumulate PFAS with an odd number of carbon atoms in the alkyl chain at higher concentrations than the next even-numbered chain (e.g., PFNA > PFOA). This pattern has been seen in other Arctic biota (Martin et al. 2004; Reiner et al. 2011) and is thought to occur as the result of atmospheric transport of PFAA precursors such as fluorotelomer alcohols (Ellis et al. 2004). This pattern is present in the seal organ tissues, but not muscle or fat, so tissue-specific partitioning may play an important role in the relative concentration patterns for PFAS (i.e., this pattern may only be apparent in blood and organ tissues). PFPeA was notably absent from seal organ samples.

We believe this is the first report of PFAS concentrations in bowhead whales. Bowhead whale mangtak as well as muscle show a tendency to accumulate PFNA and PFUnDA. PFNA and PFUnDA were detected in 25% and 37.5% of mangtak samples, respectively, although concentrations did not exceed 1 ng/g for either compound. Short-chain compounds such as PFPeA were less frequently detected, but were occasionally the predominant compounds (Table 2). PFHxA was present at a concentration of over 7 ng/g ww in the bowhead whale muscle sample. Research on PFAS concentrations in cetaceans is limited. Concentrations of legacy and long-chain PFAAs were broadly similar between bowhead whales from Sivuqaq and fin whales collected from Iceland in 2009 (Rotander et al. 2012a). Several PFAS were detectable in muscle at concentrations below 1 ng/g ww in both studies. However, PFHxA was not analyzed in the fin whale samples.

PFAS in reindeer from Sivuqaq were largely non-detectable, and detections did not follow a clear pattern. Caribou muscle collected from two locations in Canada in 2007 and 2008 had a mean Σ-PFAS concentration of 0.25 and 0.53 ng/g ww (Muller et al. 2011). These means are within the range of PFAS concentrations observed in reindeer on Sivuqaq. In Norwegian reindeer muscle collected in 2014, PFDoDA and PFTrDA were the predominant compounds with concentrations of approximately 300 pg/g ww; however, shorter chain compounds including PFHxA, PFHpA, and PFHxS were detectable (Muir et al. 2019). PFAS concentrations in reindeer from Sivuqaq appear to be comparable or perhaps slightly higher than those reported in other studies; however, studies reporting PFAS concentrations in reindeer muscle or fat are rare.

While most long-term data suggest declining trends of legacy PFAS in the Arctic, increasing trends are apparent for some compounds such as perfluorocarboxylic acids (PFCAs) in specific species and regions (Muir et al. 2019). This is possibly due to the continued use of compounds such as fluorotelomer alcohols, perfluoroalkyl phosphates, and other compounds that degrade into PFCAs (Buck et al. 2011). Additionally, the relatively small number of frequently measured PFAS comprises only a fraction of the total organofluorine compounds present in the tissues of marine mammals (Spaan et al. 2020). Most biomonitoring for PFAS utilizes blood or liver samples, and muscle tissue often has lower concentrations of PFAS than liver tissue or blood (Greaves et al. 2012, Van de Vijver et al. 2005). However, research on caribou and wolves suggests that muscle may contain up to 90% of the total body burden of PFAS (Muller et al. 2011), and serum or liver samples may not accurately represent the relative concentrations of PFAS in muscle. In circumstances where biomonitoring of biota has direct implications for human health, tissue-specific concentrations of POPs should be determined with local consumption patterns in mind.

The current study shows that recent and current use POPs such as PBDEs and PFAS are also present in marine mammal tissues. Because PBDEs and PFAS likely contribute a small proportion of total bioaccumulative toxicant concentrations in marine biota, we have not provided risk-based consumption guidelines for the traditional foods in this study. For example, previous research on traditional foods on Sivuqaq showed that marine mammals, specifically bowhead whales and seals, contain high body burdens of legacy POPs such as dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB), PCBs, and chlordanes in addition to mercury (Welfinger-Smith et al. 2011). In some species, PCBs and mercury are present at concentrations that would trigger reduced consumption advisories (≥ 0.029 μg/g for Mercury and ≥ 1.5 ng/g for PCBs), based on the EPA guidance for fish consumption for the general population (US EPA 2000). Specifically, blubber or rendered oil of marine mammals, including bowhead whales, bearded seals, and spotted seals, often contain total PCB concentrations of several hundred ppb; the EPA guidelines recommend no consumption for fish tissues above 94 ppb (US EPA 2000). Similarly, mercury concentrations in muscle tissues of spotted seal, bearded seal, and

bowhead whale are often above the 0.029 µg/g guideline for unlimited consumption in fish tissues (US EPA 2000; Welfinger-Smith et al. 2011). Potential risk from a traditional diet should be assessed holistically, considering both total bioaccumulative toxicant exposure as well as cultural, spiritual, and economic benefits of continued consumption. Beyond the risk to human health, PFAS and PBDE exposure also negatively impacts the health of Arctic biota (AMAP 2018).

Limitations

The small sample size for each tissue type and species limits our ability to estimate the distribution of concentrations present in each species. Because of the low sample size per species, we reported data on seals collectively. However, different seal species are likely to have different exposure patterns based on diet and other aspects of their natural histories. Specifically, bearded seals are primarily benthic feeders and invertebrates make up a larger proportion of their diet compared to seals that feed primarily in the water column (Crawford et al. 2015). Bearded seal samples did not have uniformly lower concentrations of contaminants. Larger sample sizes may reveal differences between species with different life histories. The age and sex of harvested animals were not recorded. Age and sex are known to influence POP concentrations in marine mammals and other species. Specifically, reproductive females tend to have lower concentrations than males because females offload POPs to their young both during gestation and lactation (Aguilar et al. 1999). We do not have information on the frequency of consumption of individual traditional foods, and therefore we did not calculate estimated daily intakes for PBDEs or PFAS.

This study utilized tissue samples from animals harvested by local hunters and processed by traditional methods. Because PFAS tend to be present at relatively high concentrations in blood serum, the circumstances of harvest and butchering of food animals may influence PFAS concentrations in tissues. Specifically, thorough bleeding may alter PFAS concentrations in peripheral tissues, and this may be a source of variation in reported concentrations. Contamination of samples is a possibility. In particular, deca-BDE is present at high concentrations in many indoor environments, including those on Sivuqaq, and there is a risk of contamination by contact with house dust (Byrne et al. 2017). However, resection of samples to eliminate external contamination limits the potential for contamination. Resection also reduces the potential impact of sample dehydration during storage, which may impact ww concentrations. Replicate analysis of samples was not conducted.

Conclusions

Both PFAS and PBDEs are present in the traditional subsistence foods of Sivuqaq. Relative concentrations of PFAS present in muscle and adipose tissue of traditional foods were dissimilar to those present in human serum from the same region. Relative concentrations of PFAS in spotted seal organs were similar in composition to those of human sera. Tissue partitioning likely plays an important role in determining the relative concentration of PFAS. Consumption of organs could be an important source of PFAS exposure. PBDE concentrations were dominated by BDEs 47, 99, and 209. Tissues with high lipid concentrations had both a greater total concentration and greater diversity of PBDEs. Both PBDEs and PFAS tended to be present at higher concentrations in seals than in bowhead whales or reindeer. Traditional food consumption on Sivuqaq contributes to PFAS and PBDE exposure.

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Data availability Data may be made available upon reasonable request and with permission of the Native Village of Gambell and the Native Village of Savoonga.

Declarations

Ethics approval This research includes samples of foods derived from animals that were traditionally harvested by the Indigenous Peoples of Sivuqaq, Alaska. No live animals were used in the course of this research.

Informed consent Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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