

Short Communication

Per- and Polyfluoroalkyl Substances and Mercury in Arctic Alaska Coastal Fish of Subsistence Importance

Kevin M. Fraley,^{a,*} Carolyn R. Hamman,^b Trent M. Sutton,^b Martin D. Robards,^a Tahzay Jones,^c and Alex Whiting^d^aWildlife Conservation Society, Fairbanks, Alaska, USA^bCollege of Fisheries and Ocean Sciences, University of Alaska Fairbanks, Fairbanks, Alaska, USA^cU.S. National Park Service, Anchorage, Alaska, USA^dEnvironmental Program, Native Village of Kotzebue, Kotzebue, Alaska, USA

Abstract: Per- and polyfluoroalkyl substances (PFAS) and mercury (Hg) are harmful compounds that are widely present in the environment, partly due to spills and atmospheric pollution. The presence of PFAS and Hg in the tissues of animals that are harvested by rural and Indigenous Alaskans is of great concern, yet fish in Arctic Alaska have not previously been assessed for concentrations of PFAS. Fish species of subsistence and recreational importance were collected from nearshore Beaufort and Chukchi Sea, Alaska habitats and assessed for PFAS and total mercury concentrations [THg]. We found multiple PFAS compounds present at low levels (<3 µg/kg) in the muscle tissue of inconnu, broad whitefish, Dolly Varden char, Arctic flounder, saffron cod, humpback whitefish, and least cisco. In addition, [THg] levels in these fish were well below levels triggering local fish consumption guidelines (<170 µg/kg). These initial results indicate no evidence of the Alaska Arctic nearshore fish species examined as an avenue of PFAS or Hg exposure to people who harvest them. However, sources and trends of these contaminants in the Arctic require further investigation. *Environ Toxicol Chem* 2023;00:1–7. © 2023 SETAC

Keywords: Arctic; Coastal; Fish; Mercury; Per- and polyfluoroalkyl substances; Subsistence

INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals mainly produced for firefighting foam but also found in many everyday household and commercial products. These persistent organic pollutants are released into the environment primarily during firefighting training and response, through spills and releases from manufacturing facilities, and via atmospheric deposition from industrial emissions (Hu et al., 2016). Globally, PFAS have been released into soil, groundwater, and surface water, causing large-scale environmental contamination at thousands of locations (Kempisty et al., 2018). There is sufficient evidence that exposure to PFAS (usually through ingestion) results in decreased antibody count, dyslipidemia, decreased infant and fetal growth, and an increased risk of kidney cancer in humans (National Academies of Sciences, Engineering, and Medicine, 2022). In addition, PFAS exposure may increase the human risk of breast cancer, liver enzyme alterations, pregnancy-induced hypertension, testicular

cancer, thyroid disease, and ulcerative colitis; such exposure also has harmful effects on fish and animals (Fenton et al., 2021; Grandjean, 2018; Han & Fang, 2010; Sunderland et al., 2019). Some PFAS are bioaccumulative, particularly those with longer carbon chains, and can biomagnify in aquatic and terrestrial food webs (Conder et al., 2008; Goodrow et al., 2020; Taylor et al., 2018). To date, the efficacy of remediating PFAS contamination in the environment, particularly in water, has been poor (Darlington et al., 2018), and government regulation of the use and cleanup of PFAS is lacking (Grandjean, 2018; US Environmental Protection Agency [USEPA], 2019).

Mercury (Hg) has long been identified as a contaminant that can cause negative health effects if exposure occurs, and it is widely present in the environment. Unlike PFAS, it can be released or mobilized through both anthropogenic and natural pathways, for example, placer mining and thawing permafrost. Mercury, particularly monomethyl mercury (MeHg), can cause negative health effects in humans and animals (Sundseth et al., 2017), especially for sensitive populations such as pregnant women and children. Mercury bioaccumulates in wild-harvested foods such as fish (Jewett & Duffy, 2007), and biomagnifies in terrestrial and aquatic food chains. State and federal health agencies commonly issue Hg consumption advisories or guidelines for fish and wildlife to protect those who

This article includes online-only Supporting Information.

* Address correspondence to kfraley@wcs.org

Published online 21 July 2023 in Wiley Online Library

(wileyonlinelibrary.com).

DOI: 10.1002/etc.5717

might consume wild foods. For example, in Alaska, unrestricted fish consumption guidelines for pregnant women and children ($200\text{ }\mu\text{g/kg}$) can be exceeded in lingcod (*Ophiodon elongatus*) and lake trout (*Salvelinus namaycush*), which are long-lived predators that can bioaccumulate Hg at levels that could cause harmful dietary exposure (Alaska Department of Environmental Conservation, 2022).

Dietary exposure to PFAS, Hg, and other contaminants through harvest and consumption of wild foods is of concern (Denys et al., 2014; Fraley et al., 2020), and has led to federal and local testing of wildlife and fish tissues near contaminated areas (see Alaska Department of Environmental Conservation, 2019; New Jersey Department of Environmental Protection, 2018). The results from testing have led to issuance of consumption advisories, moratoriums on fish harvest, and hunting and fishing closures in some instances (Alaska Department of Fish and Game, 2019; Minnesota Department of Health, 2018). In Arctic Alaska, the potential presence of contaminants in wildlife and fish that rural and Indigenous residents rely on for food security and cultural identity is of special concern (Byrne et al., 2017). There are few point sources of PFAS and few anthropogenic Hg sources in the Arctic, but

long-range transport and atmospheric deposition have been linked to their presence in Arctic environments and biota (Dietz et al., 2013; Kahkashan et al., 2019). Limited PFAS assessments have been performed in the North American Arctic, and these chemicals have been found in marine mammals and fish of subsistence use, including beluga whales (*Delphinapterus leucas*) in Hudson Bay, Canada (Kelly et al., 2009) and Alaska blackfish (*Dallia pectoralis*) near St. Lawrence Island, Alaska (Byrne et al., 2017). Bioaccumulation of other contaminants such as Hg in harvested wild foods has been surveyed more broadly in the Arctic compared with PFAS (see Cyr et al., 2019). Of the conducted Arctic surveys, little research has been done on fish in the Beaufort Sea region of Alaska near the Prudhoe Bay oilfield industrial complex or in the Chukchi Sea near the Red Dog Mine port and road facilities (Figure 1 and Supporting Information, Figure S1), which are possible local sources of contamination.

There is high harvest and human consumption of subsistence fish along the Chukchi and Beaufort Sea coastlines of Alaska. For example, annual harvests of up to 80 000 Arctic cisco (*Coregonus autumnalis*) occur in the Colville River delta and more than 25 000 inconnu (also known as sheefish;

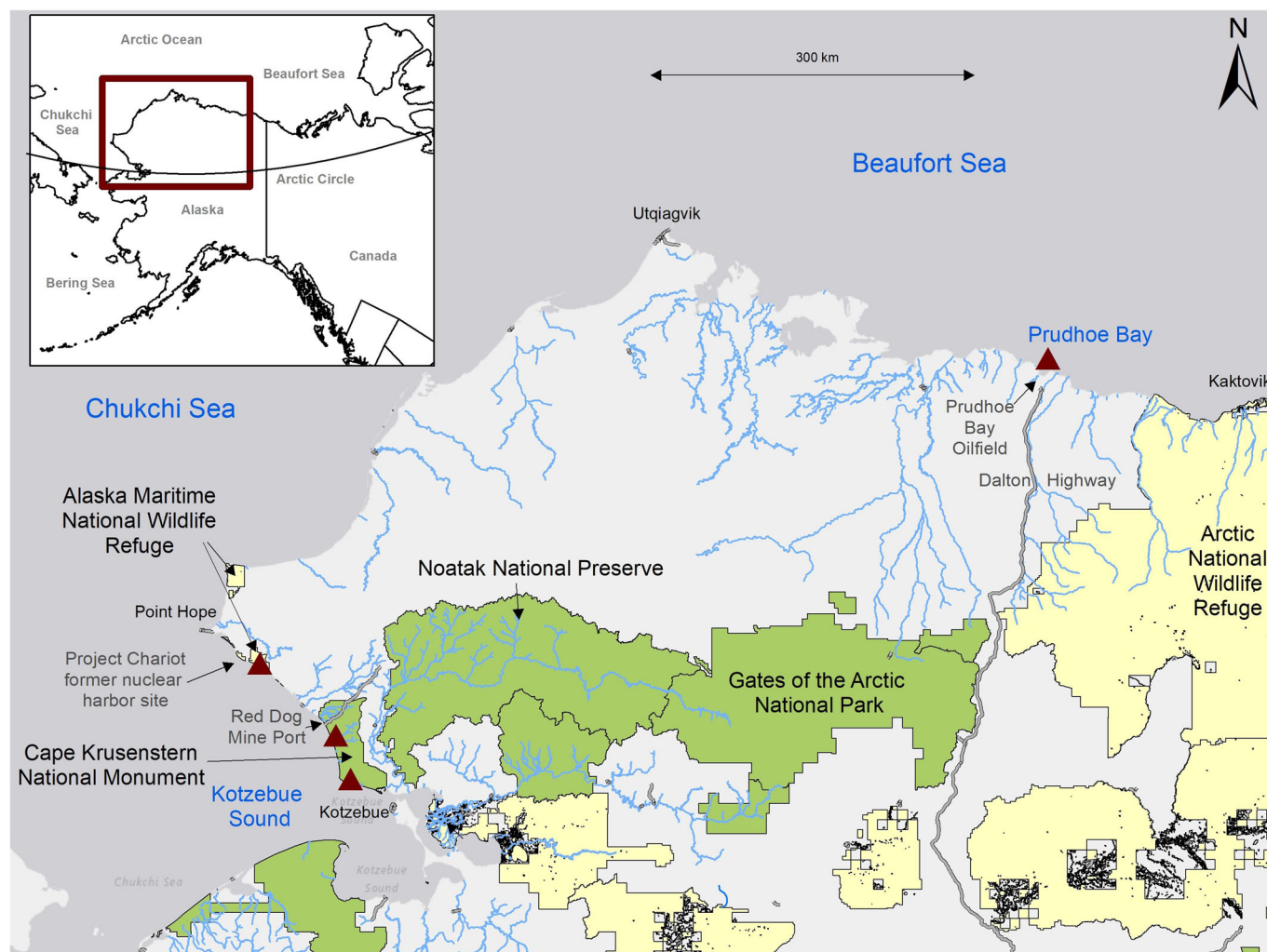


FIGURE 1: Map of the Beaufort and Chukchi Seas, Alaska. Fish capture locations are denoted by dark triangles.

Stenodus leucichthys) are harvested each year in Kotzebue Sound (Fechhelm et al., 2007; Whiting, 2006). Despite these harvests, minimal data exist regarding contaminant concentrations in these fish (but see Cyr et al., 2019). Therefore we undertook a preliminary assessment of PFAS and Hg concentrations in the muscle tissues of several species of diadromous and marine fish harvested by rural and Indigenous residents in the Alaskan Arctic. Our objectives were to: (1) provide initial PFAS and [THg] data for coastal subsistence fish in the region; and (2) ascertain whether any health concerns may exist for local residents harvesting and consuming these fish.

METHODS

Fish were collected during June–August 2018–2021 in the estuarine band of the nearshore Beaufort Sea adjacent to the Prudhoe Bay oilfield industrial complex and in brackish coastal lagoons along the Chukchi Sea near the Red Dog Mine port facility and the former Project Chariot nuclear harbor site (Figure 1 and Supporting Information, Figure S1). Fish were captured via angling, fyke net, and tangle net by University of Alaska Fairbanks' Beaufort Sea Nearshore Long-Term Fisheries Monitoring Program and the Wildlife Conservation Society/National Park Service Chukchi Sea Coastal Lagoons Vital Signs Long-Term Ecological Monitoring Project. All species collected are commonly harvested for subsistence use by rural and indigenous Alaskan residents.

Fish were euthanized using an overdose of anesthetic (AQUI-S 20E), stored in clean bags, and placed in a cooler with ice until they could be transferred to a freezer. Given the chemical formula of AQUI-S 20E (10% eugenol solution), there is no reason to suspect any interference in the detection and quantification of PFAS compounds or Hg. Dissection and muscle tissue extraction were conducted at the University of Alaska Fairbanks (according to the University of Alaska Institutional Animal Care and Use Committee approval #1054743). Muscle tissue was selected for analysis because this is the most commonly consumed part of the fish by subsistence and recreational fishers, although liver and eggs are also occasionally eaten. Mercury and certain PFAS have been found to accumulate in muscle tissue, among other tissue types (Sun et al., 2022). Fish were removed from the freezer and thawed minimally to facilitate removal of skin and the extraction of approximately 10 g of dorsal muscle tissue from each fish. Dissection utensils were thoroughly rinsed with distilled water between each fish to ensure that cross-contamination of samples did not occur. For PFAS analysis of fish collected in 2020 and 2021, pieces of muscle tissue from fish were placed in jars provided by Eurofins TestAmerica Laboratories (Sacramento, CA, USA) to be homogenized and run as composite samples by species and by location (Chukchi or Beaufort Sea). Composite samples were employed due to the high cost of PFAS analyses. Samples were frozen and shipped to the Eurofins TestAmerica Laboratory for PFAS analysis by USEPA method 537M (Shoemaker et al., 2008). Total mercury analysis by USEPA method 7471B (2007) was conducted for 2020 Beaufort Sea

composite samples, but was assessed for individual fish via USEPA methods 7473 (2007) and 30B (2017) for those collected from the Chukchi Sea in 2021 at the University of Alaska Fairbanks Marine Ecotoxicology and Trophic Assessment Laboratory. These two methods are both considered acceptable and comparable for environmental samples (Rattonetti et al., 2013). For fish collected in 2018, muscle tissue was desiccated in coin envelopes placed in an airtight container with silica beads, and THg analyses were conducted in 2022 at the University of Alaska Fairbanks Marine Ecotoxicology and Trophic Assessment Laboratory by USEPA methods 7473 (2007) and 30B (2007). For the subset of desiccated samples, species-specific linear regressions were developed from data collected by Cyr et al. (2019) to calculate estimated wet weight [THg] for muscle tissue (see the Supporting Information for calculations). Total mercury analyses were selected because harmful MeHg concentrations in most fish tissues from the region have been found to consist of >80% THg (Cyr et al., 2019).

Otoliths were extracted from fish collected in 2022 and from some of the fish collected in previous years (see the Supporting Information, Tables S1 and S2) to estimate age of the fish assessed for contaminants. This was done to ascertain whether contaminant concentrations were associated with the age and size of the fish (i.e., bioaccumulation and trophic position). Available otoliths were ground and thin-sectioned in the US Fish and Wildlife Service laboratory in Fairbanks (AK) and aged under a microscope by Fish Biologist R. Brown (Supporting Information, Tables S1 and S2). For fish in which otoliths were unavailable, approximate age was estimated based on length-age information (when available).

RESULTS AND DISCUSSION

Least cisco (*Coregonus sardinella*; $n = 17$), humpback whitefish (*Coregonus pidschian*; $n = 15$), inconnu ($n = 9$), broad whitefish (*Coregonus nasus*; $n = 7$), Dolly Varden char (*Salvelinus malma*; $n = 7$), Arctic flounder (*Liopsetta glacialis*; $n = 7$), saffron cod (*Eleginus gracilis*; $n = 4$), Pacific herring (*Clupea pallasii*; $n = 3$), starry flounder (*Platichthys stellatus*; $n = 1$), and Bering cisco (*Coregonus laurettae*; $n = 1$) were collected from nearshore Beaufort and Chukchi Sea habitats for analyses (Supporting Information, Tables S1 and S2, and Figure S1). Results are reported in units of $\mu\text{g}/\text{kg}$ THg in wet muscle to provide a metric that is useful for people who harvest and consume these fish.

Chemical analyses indicated nondetect (ND) or very low results ($<3 \mu\text{g}/\text{kg}$) for all PFAS in the analytical suite tested (24 compounds) and ND to moderate [THg] in all fish species at all locations (Tables 1 and 2). Due to laboratory blank contamination for the Beaufort Sea sample run, it was unclear whether perfluorooctanesulfonic acid (PFOS; one of the most prominent PFAS) and perfluorobutanoic acid (PFBA) were actually present in fish caught in the Beaufort Sea, because reanalysis of the samples with blank contamination subsequently resulted in ND results or indicated further blank contamination. Regardless of whether the detections were due to laboratory

TABLE 1: Per- and polyfluoroalkyl substances muscle tissue composite sample results from fish collected in the Beaufort Sea near Prudhoe Bay in 2020 and Chukchi Sea coastal lagoons in 2021

Location/study	Fish species	No.	PFOS (μg/kg)	PFBA (μg/kg)	PFNA (μg/kg)	Other (μg/kg)
Beaufort Sea, Alaska (present study)	Broad whitefish	5	2.8 ^a	1.1 ^b	ND	ND
	Dolly Varden	5	ND	0.42 ^b	ND	ND
	Arctic flounder	5	1.5 ^a	0.39 ^b	0.25	ND
	Saffron cod	2	1.1 ^a	0.62 ^b	0.22	ND
Chukchi Sea, Alaska (present study)	Broad whitefish	2	ND	ND	0.38	PFOA: 0.29 PFHpA: 0.52
	Dolly Varden	2	ND	ND	ND	ND
	Arctic flounder	3	ND	ND	ND	PFDA: 0.12 ^b
	Saffron cod	3	ND	ND	0.21	ND
	Inconnu	3	ND	ND	0.20	ND
	Humpback whitefish	3	ND	ND	0.56	PFOA: 0.45 PFHpA: 0.69
	Least cisco	3	ND	ND	0.18	ND
	Alaska blackfish	—	ND	ND	ND	PFBS: <LOD–59.2
St. Lawrence Island (Byrne et al., 2017)	Ninespine stickleback	—	<LOD–16.1	ND	<LOD–4.13	PFOA: <LOD–1.51
Canadian and European Arctic, Greenland (Muir et al., 2019)	Various freshwater fish	—	ND–31.75	0.1–0.14	ND–1.3	PFOA: ND–2.1
	Various marine and diadromous fish	—	ND–4.27	ND	ND–3.9	PFOA: ND–0.36
	Various marine mammals	—	0.06–3270	<LOD–<0.07	<LOD–497	PFOA: <LOD–39.2

^aResults revealed laboratory blank contamination, and samples were reanalyzed for PFOS, with either ND or additional blank contamination found.

^bResults revealed laboratory blank contamination, but reanalysis was not conducted for this analyte.

See the Supporting Information for full analytical results. Results reported from other studies are ranges of average contaminant values across species, locations, and tissue types.

<LOD = the value was less than the analytical method's level of detection; No. = number of individual fish that comprised the composite sample; ND = no detection of the analyte; PFBA = perfluorobutanoic acid; PFDA = perfluorodecanoic acid; PFHpA = perfluoroheptanoic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFBS = perfluorobutane sulfonate.

contamination, the concentrations of PFOS and PFBA were at such low levels (<3 μg/kg) that the potential contamination is of no consequence in the context of North American human consumption advisory levels.

Perfluorononanoic acid (PFNA) was detected at very low concentrations in two Beaufort Sea marine species (saffron cod and Arctic flounder [0.22–0.25 μg/kg]), and five Chukchi Sea diadromous and marine species (inconnu, humpback whitefish,

TABLE 2: Total mercury (THg) muscle tissue composite sample results from fish collected in the Beaufort Sea near Prudhoe Bay in 2020 and average [THg] results by species from Chukchi Sea coastal lagoons in 2018 and 2021

Location/study	Fish species	No., comp	No.	[THg] (μg/kg)	MeHg (μg/kg)
Beaufort Sea, Alaska (present study)	Broad whitefish	5	N/A	24	—
	Dolly Varden	5	N/A	ND	—
	Arctic flounder	5	N/A	27	—
	Saffron cod	2	N/A	30	—
Chukchi Sea, Alaska (present study)	Broad whitefish	N/A	2	3.62	—
	Dolly Varden	N/A	2	2.94	—
	Arctic flounder	N/A	2	67.68	—
	Saffron cod	N/A	2	48.45	—
	Humpback whitefish	N/A	15	24.74	—
	Inconnu	N/A	9	88.41	—
	Least cisco	N/A	17	83.50	—
	Bering cisco	N/A	1	40	—
	Starry flounder	N/A	1	77	—
	Pacific herring	N/A	3	21.46	—
Chukchi Sea, Alaska (Cyr et al., 2019)	Various marine and diadromous fish	—	—	4.7–81.7	3.3–74.5
Canadian and European Arctic, Greenland (Dietz et al., 2013)	Various freshwater and marine fish	—	—	10–500	—
Canadian Arctic (Tran et al., 2015)	Whale, seal, and walrus liver	—	—	20–75 540	—
	Dolly Varden	—	—	15–254	—

Testing was conducted by TestAmerica Laboratories using USEPA method 7471B (2007) or by the University of Alaska Fairbanks Marine Ecotoxicology and Trophic Assessment Laboratory (DMA-80 Direct Mercury Analyzer; Milestone). See the Supporting Information for full analytical results. Results reported from other studies are ranges of average contaminant values across species, locations, and tissue types.

N/A = not available; ND = no detection of the analyte; No., comp = number of individual fish that comprised a composite sample, if applicable; No. = number of fish of that species that were analyzed and values averaged.

least cisco, broad whitefish, and saffron cod [0.18–0.56 $\mu\text{g}/\text{kg}$]). In addition, perfluorooctanoic acid (PFOA) and perfluoroheptanoic acid were detected in broad and humpback whitefish from Chukchi Sea lagoons (<1 $\mu\text{g}/\text{kg}$). Anadromous Dolly Varden appeared to carry the lowest levels of PFAS, with the sample from the Chukchi Sea showing no detections and the sample from the Beaufort Sea showing only a detection of PFBA (but with blank contamination). However, we note that for most samples, the chemical results were less than the reporting limit but greater than or equal to the method detection limit, and thus the concentrations are approximate values.

Compared with our results, the other nearby PFAS investigation of Alaska blackfish and ninespine stickleback (*Pungitius pungitius*) in freshwater near subarctic St. Lawrence Island, Alaska exhibited much higher maximum concentrations (up to 16.1 $\mu\text{g}/\text{kg}$ PFOS and 59.2 $\mu\text{g}/\text{kg}$ perfluorobutane sulfonate; Byrne et al., 2017; Table 1). This is likely a result of the proximity of that collection site to formerly used military defense sites and landfills that may have been point sources for PFAS contamination. Contamination of fish by PFAS is typically tied more closely to point source releases of firefighting foam or biosolids rather than long-range pollution.

In addition, most freshwater, marine, and diadromous fish in the Canadian and European Arctic contained higher average concentrations of all analytes compared with our results (up to 31.75 $\mu\text{g}/\text{kg}$ PFOS; Muir et al., 2019; Table 1). Notably, several of the fish we examined have not been assessed for PFAS concentration in the published literature (Muir et al., 2019). There are few set fish consumption PFAS advisory limits, but most states or institutions that issue advisories use a cutoff between 9 and 47 $\mu\text{g}/\text{kg}$; however, the analytes of interest can be individual PFAS compounds or multiple in combination (Alaska Department of Fish and Game, 2019; Fliedner et al., 2016; Minnesota Department of Health, 2018; New Jersey Department of Environmental Protection, 2018). For example, The European Food Safety Authority (EFSA) has set a tolerable weekly intake of fish meat at 0.0044 $\mu\text{g}/\text{kg}$ for PFOA, PFNA, perfluorohexane sulfonic acid, and PFOS in combination (EFSA Panel on Contaminants in the Food Chain, 2020). Based on our results, there is no evidence of a PFAS-related health concern through consumption of the fish species we tested in this jurisdiction of the Arctic. However, current North American guidelines may not be adequate to protect human health given the higher volume of consumption of wild foods by Alaska Native subsistence users (Byrne et al., 2017) and when considering more conservative advisories such as the one issued by the EFSA.

Mercury concentrations in Beaufort Sea fish tissue ranged from ND in Dolly Varden char to 30 $\mu\text{g}/\text{kg}$ in saffron cod (Table 2). In the Chukchi Sea, concentrations ranged from 2.3 $\mu\text{g}/\text{kg}$ in a Dolly Varden char to 162.4 $\mu\text{g}/\text{kg}$ in a least cisco. The fish tissue [THg] concentrations from the present study (ND–162.4 $\mu\text{g}/\text{kg}$) were similar to the range of those found by Cyr et al. (2019) for diadromous and marine fish in the nearby Chukchi Sea (average values by species ranged from 4.7 to 81.7 $\mu\text{g}/\text{kg}$; Table 2) and closer to the lower range of the values reported by Dietz et al. (2013) for freshwater and marine

species in the Canadian and European Arctic (average [THg] 10–500 $\mu\text{g}/\text{kg}$ by species/location; Table 2). Our Chukchi and Beaufort Sea results for anadromous Dolly Varden (ND–3.6 $\mu\text{g}/\text{kg}$) were notably lower than values for the same species found by Tran et al. (2015) in the Canadian Arctic (average 15–254 $\mu\text{g}/\text{kg}$ by location), perhaps due to differences in life histories between the populations studied. The low values detected for Dolly Varden and other species in our study are surprising given that there are natural (i.e., permafrost melting/freshwaters; Leitch et al., 2007) as well as potential anthropogenic (Prudhoe Bay oilfield, Red Dog Mine Port) sources of aquatic Hg deposition near the study area. The differences in values between the studies highlight the importance of local environmental conditions that influence methylation of Hg, and the importance of food web dynamics, which drive how much Hg is made bioavailable. Sources of mercury are simply some of the numerous factors that can influence mercury concentrations in the food web.

In the United States (including Alaska), fish consumption guidelines for Hg are triggered when concentrations reach between 200 and 1000 $\mu\text{g}/\text{kg}$ MeHg (MeHg comprises a portion of THg; Jewett & Duffy, 2007; Verbrugge, 2007), but consumption guidelines and advisories differ between states and nations. Only one fish sampled (a least cisco that registered 162.4 $\mu\text{g}/\text{kg}$ [THg] in muscle tissue) approached an advisory level. Because harmful MeHg makes up approximately 80% of THg, this fish was well below the 200 $\mu\text{g}/\text{kg}$ State of Alaska fish consumption guideline for MeHg (Hamade, 2014). Thus the [THg] of the fish analyzed for the present study provided no evidence of a health concern for people who harvest and consume them.

The low or nonexistent concentrations of PFAS and Hg in the fish we examined are particularly notable because these species represent a wide suite of life-history types (see the Supporting Information, Tables S1 and S2, for detailed information). For example, Dolly Varden char travel long distances through freshwater rivers, the nearshore estuarine band, and the open Beaufort and Chukchi Seas (Courtney et al., 2018), increasing the likelihood of consuming prey in any number of habitats that contain PFAS or Hg, thus potentially accumulating significant amounts. However, Dolly Varden exhibited lower [THg] and PFAS concentrations than other species. In addition, the diets of the fish examined vary from benthic invertivorous to pelagic piscivorous, covering multiple facets of the nearshore Beaufort and Chukchi Sea food webs. This suggests that freshwater, estuarine, and marine environments in Alaska's Arctic are fairly pristine in comparison with urban and industrial settings (at least for the contaminants we assessed), despite the presence of oilfields and busy port sites within a few kilometers of some sampling locations. Conceivably, a moderate amount of contaminants would be released to the terrestrial, freshwater, and marine environments surrounding oilfields and ports through deposition from vehicle and machinery exhaust, spills, and fugitive dust blowing from roads. However, even with fish up to age 13+ included in our analyses (Supporting Information, Table S1), which would be prone to accumulate higher

contaminant loads over their longer lifetimes, low amounts of PFAS and Hg were detected in their tissues.

CONCLUSIONS

Based on the results of the present study of PFAS and Hg concentrations in fish of varying life histories in the Beaufort Sea near the Prudhoe Bay oilfields and the Chukchi Sea near the Red Dog Mine port site, fish harvested by rural and Indigenous Alaskans for subsistence, some of which are also caught by recreational anglers, had PFAS and [THg] levels below established health thresholds and measurably lower than in other fish studies. Due to the small sample size for PFAS (functionally $n = 1$ sample/species/location due to the composite sample methodology) and patchy geographic area our samples were collected from (Figure 1), it should be noted that a wider ranging, more robust contaminant survey could be conducted to confirm these results. However, our data represent valuable baseline findings for the fish species examined in this region. Other species in the region, including freshwater, marine, and diadromous taxa of subsistence importance that we did not examine (e.g., Pacific salmon and Arctic cisco), could also be assessed. In addition, other potential contaminants of concern could be examined, such as polycyclic aromatic hydrocarbons, pesticides, or flame retardants.

Despite our findings, there is a well-documented presence of PFAS, Hg, and other contaminants at levels of concern in marine mammals (and some fish) in the Arctic (Byrne et al., 2022; Dietz et al., 2013; Table 2), and further investigation is warranted to determine how these contaminants enter and magnify through the food web. In addition, potential point sources of PFAS, Hg, and other contaminants in the Arctic should be prioritized for assessment, especially in the context of subsistence-harvested fish. For example, there is a need to investigate potential PFAS contamination of fish from a known source area along the Beaufort Sea coast at the Naval Arctic Research Laboratory contaminated site in Utqiagvik, Alaska, where combined PFOS and PFOA concentrations in the water have been measured up to 200.9 ng/L (US Navy, 2017). Finally, additional work is needed toward regulating the manufacture and use of PFAS and devising strategies for remediation of PFAS environmental contamination (Darlington et al., 2018; Grandjean, 2018), to minimize future point source and long-range contamination impacts in the Arctic and on a global basis.

Supporting Information—The Supporting Information is available on the Wiley Online Library at <https://doi.org/10.1002/etc.5717>.

Acknowledgments—We thank A. Cyr, Alaska Department of Health, for providing advice on analysis methods and interpretation of data. Thanks to M. Castellini, University of Alaska Fairbanks (UAF) Marine Ecotoxicology and Trophic Assessment Laboratory, for giving advice and conducting Hg analyses. K. Gatt graciously offered data and input on Beaufort Sea fish ages. We are also grateful to R. Brown of the US Fish and

Wildlife Service for aging fish otoliths. Field logistics, fish capture, sample storage, and laboratory space through the Beaufort Sea Nearshore Long-Term Fisheries Monitoring Program were facilitated by Hilcorp Alaska. Additional thanks to B. Sharp (Hilcorp Alaska), J. Dushane (Hilcorp Alaska), A. Erickson (Weston Solutions), and M. Larson (Weston Solutions), who provided assistance before, throughout, and after the field season with logistics, billeting, and overall project management. M. Hill, D. Graves, B. Henderson, H. Henson, and C. Jones (all from Hilcorp Alaska) provided additional assistance with in-season changes in field logistics, continual updates on sampling conditions, and details of COVID-19 safety protocols. S. Kramer (Hilcorp Alaska) provided security detail while we were sampling. Technicians/interns M. Lunde (Wildlife Conservation Society [WCS]), A. Page (UAF), F. Elmore (UAF), and K. Witte (WCS) participated in field sampling and/or laboratory preparation. Research was conducted under permits issued by the Alaska Department of Fish and Game (SF2018-061, CF-20-021, and SF2021-088), University of Alaska IACUC (1054743), the North Slope Borough (NSB 19-011), and the National Park Service. Funding for chemical analyses was provided by the National Fish and Wildlife Foundation (grant: 8006.18.059259), the National Park Service (grant: P19AC00638), and the Western Division of the American Fisheries Society Small Project Grants program.

Author Contributions Statement—**Kevin M. Fraley**: Conceptualization; Methodology; Investigation; Resources; Writing—original draft; Project administration. **Carolyn R. Hamman**: Investigation; Supervision; Writing—review & editing. **Trent M. Sutton**: Resources; Writing—review & editing. **Martin D. Robards**: Funding acquisition; Writing—review & editing. **Tahzay Jones**: Funding acquisition; Writing—review & editing. **Alex Whiting**: Visualization; Writing—review & editing.

Data Availability Statement—All data are included in the manuscript Supporting Information files.

REFERENCES

- Alaska Department of Environmental Conservation. (2019). Per- and polyfluoroalkyl substances (PFAS) in Alaska's fish, fish samples collected: 2009–2010. Office of the State Veterinarian, Fish Monitoring Program. https://dec.alaska.gov/media/14391/pfas_results-2009-2010.pdf
- Alaska Department of Environmental Conservation. (2022). Total mercury in fish and shellfish caught in alaskan waters: Fish samples collected: 2001–2021. <https://dec.alaska.gov/media/22378/mercury-in-alaska-fish.pdf>
- Alaska Department of Fish and Game. (2019). Sport fishing closure of Kimberly and Polaris Lakes in the North Pole and Eielson Areas. Emergency Order No. 3-R-U-01-19. April 3, 2019. https://www.adfg.alaska.gov/Static-sf/EONR/PDFs/2019/R3/EO%203-R-U-01-19%20Stocked%20Waters%20Closure_FINAL.pdf
- Byrne, S., Seguinot-Medina, S., Miller, P., Waghiyi, V., von Hippel, F. A., Buck, C. L., & Carpenter, D. O. (2017). Exposure to polybrominated diphenyl ethers and perfluoroalkyl substances in a remote population of Alaska Natives. *Environmental Pollution*, 231, 387–395.
- Byrne, S., Seguinot-Medina, S., Waghiyi, V., Apatiki, E., Immingan, T., Miller, P., & Carpenter, D. O. (2022). PFAS and PBDEs in traditional subsistence foods from Sivuqaq, Alaska. *Environmental Science and Pollution Research*, 29(51), 77145–77156.

- Conder, J. M., Hoke, R. A., Wolf, W. D., Russell, M. H., & Buck, R. C. (2008). Are PFCA's bioaccumulative? A critical review and comparison with regulatory criteria and persistent lipophilic compounds. *Environmental Science & Technology*, 42(4), 995–1003.
- Courtney, M. B., Scanlon, B., Brown, R. J., Rikardsen, A. H., Gallagher, C. P., & Seitz, A. C. (2018). Offshore ocean dispersal of adult Dolly Varden *Salvelinus malma* in the Beaufort Sea. *Polar Biology*, 41(4), 817–825.
- Cyr, A. P., López, J. A., Wooller, M. J., Whiting, A., Gerlach, R., & O'Hara, T. (2019). Ecological drivers of mercury concentrations in fish species in subsistence harvests from Kotzebue Sound, Alaska. *Environmental Research*, 177, 108622.
- Darlington, R., Barth, E., & McKernan, J. (2018). The challenges of PFAS remediation. *The Military Engineer*, 110, 58–60.
- Denys, S., Fraize-Frontier, S., Moussa, O., Le Bizet, B., Veyrand, B., & Volatier, J. L. (2014). Is the fresh water fish consumption a significant determinant of the internal exposure to perfluoroalkylated substances (PFAS)? *Toxicology Letters*, 231, 233–238.
- Dietz, R., Sonne, C., Basu, N., Braune, B., O'Hara, T., Letcher, R. J., & Asmund, G. (2013). What are the toxicological effects of mercury in Arctic biota? *Science of the Total Environment*, 443, 775–790.
- European Food Safety Authority Panel on Contaminants in the Food Chain. (2020). Risk to human health related to the presence of perfluoroalkyl substances in food. *EFSA Journal*, 18(9), e06223.
- Fechhelm, R. G., Streever, B., & Gallaway, B. J. (2007). The Arctic cisco (*Coregonus autumnalis*) subsistence and commercial fisheries, Colville River, Alaska: A conceptual model. *Arctic*, 60, 421–429.
- Fenton, S. E., Ducatman, A., Boobis, A., DeWitt, J. C., Lau, C., Ng, C., & Roberts, S. M. (2021). Per- and polyfluoroalkyl substance toxicity and human health review: Current state of knowledge and strategies for informing future research. *Environmental Toxicology and Chemistry*, 40, 606–630.
- Fliedner, A., Lohmann, N., Rüdell, H., Teubner, D., Wellnitz, J., & Koschorreck, J. (2016). Current levels and trends of selected EU Water Framework Directive priority substances in freshwater fish from the German environmental specimen bank. *Environmental Pollution*, 216, 866–876.
- Fraleigh, K. M., Fraley, H. N., Arthur, D., & Walther, E. J. (2020). Per- and polyfluoroalkyl substances (PFAS): Anglers may be exposed to harmful chemicals in their catch. *Fisheries*, 45(3), 138–144.
- Goodrow, S. M., Ruppel, B., Lippincott, R. L., Post, G. B., & Procopio, N. A. (2020). Investigation of levels of perfluoroalkyl substances in surface water, sediment and fish tissue in New Jersey, USA. *Science of the Total Environment*, 729, 138839.
- Grandjean, P. (2018). Delayed discovery, dissemination, and decisions on intervention in environmental health: A case study on immunotoxicity of perfluorinated alkylate substances. *Environmental Health*, 17, 62.
- Hamade, A. K. (2014). Fish consumption advice for Alaskans: A risk management strategy to optimize the public's health. Alaska Scientific Advisory Committee for Fish Consumption, State of Alaska.
- Han, J., & Fang, Z. (2010). Estrogenic effects, reproductive impairment and developmental toxicity in oviparous swordtail fish (*Xiphophorus helleri*) exposed to perfluorooctane sulfonate (PFOS). *Aquatic Toxicology*, 99, 281–290.
- Hu, X. C., Andrews, D. Q., Lindstrom, A. B., Bruton, T. A., Schaidler, L. A., Grandjean, P., & Higgins, C. P. (2016). Detection of poly- and perfluoroalkyl substances (PFASs) in US drinking water linked to industrial sites, military fire training areas, and wastewater treatment plants. *Environmental Science & Technology Letters*, 3, 344–350.
- Jewett, S. C., & Duffy, L. K. (2007). Mercury in fishes of Alaska, with emphasis on subsistence species. *Science of the Total Environment*, 387(1–3), 3–27.
- Kakhkashan, S., Wang, X., Chen, J., Bai, Y., Ya, M., Wu, Y., & Inam, A. (2019). Concentration, distribution and sources of perfluoroalkyl substances and organochlorine pesticides in surface sediments of the northern Bering Sea, Chukchi Sea and adjacent Arctic Ocean. *Chemosphere*, 235, 959–968.
- Kelly, B. C., Ikonomou, M. J., Blair, J. D., Surridge, B., Hoover, D., Grace, R., & Gobas, F. A. (2009). Perfluoroalkyl contaminants in an Arctic marine food web: Trophic magnification and wildlife exposure. *Environmental Science & Technology*, 43, 4037–4043.
- Kempisty, D. M., Xing, Y., & Racz, L. (2018). *Perfluoroalkyl substances in the environment: Theory, practice, and innovation*. CRC Press.
- Leitch, D. R., Carrie, J., Lean, D., Macdonald, R. W., Stern, G. A., & Wang, F. (2007). The delivery of mercury to the Beaufort Sea of the Arctic Ocean by the Mackenzie River. *Science of the Total Environment*, 373(1), 178–195.
- Minnesota Department of Health. (2018). Fish Consumption Advisory Program meal advice tables. April 2018. <https://www.health.state.mn.us/communities/environment/fish/docs/eating/mealadviceables.pdf>
- Muir, D., Bossi, R., Carlsson, P., Evans, M., De Silva, A., Halsall, C., & Roos, A. (2019). Levels and trends of poly- and perfluoroalkyl substances in the Arctic environment—An update. *Emerging Contaminants*, 5, 240–271.
- National Academies of Sciences, Engineering, and Medicine. (2022). *Guidance on PFAS exposure, testing, and clinical follow-up*. The National Academies Press. <https://doi.org/10.17226/26156>
- New Jersey Department of Environmental Protection. (2018). Investigation of levels of perfluorinated compounds in New Jersey fish, surface water, and sediment. SR15-010. June 18, 2018. <https://www.nj.gov/dep/dsr/publications/Investigation%20of%20Levels%20of%20Perfluorinated%20Compounds%20in%20New%20Jersey%20Fish,%20Surface%20Water,%20and%20Sediment.pdf>
- Rattonetti, A., Salvato, M., Mays, G., Nguyen, M., & Wen, S. Z. (2013). Determination of mercury in environmental samples. *Proceedings of the 23rd Annual International Conference on Soil, Water, Energy, and Air (Volume 1)*, pp. 194–209.
- Shoemaker, J. A., Grimmett, P., & Boutin, B. (2008). Method 537 Modified: Determination of selected perfluorinated alkyl acids in drinking water by solid phase extraction and liquid chromatography/tandem mass spectrometry (LC/MS/MS). US Environmental Protection Agency.
- Sun, J. M., Kelly, B. C., Gobas, F. A., & Sunderland, E. M. (2022). A food web bioaccumulation model for the accumulation of per- and polyfluoroalkyl substances (PFAS) in fish: How important is renal elimination? *Environmental Science: Processes & Impacts*, 24(8), 1152–1164.
- Sunderland, E. M., Hu, X. C., Dassuncao, C., Tokranov, A. K., Wagner, C. C., & Allen, J. G. (2019). A review of the pathways of human exposure to poly- and perfluoroalkyl substances (PFASs) and present understanding of health effects. *Journal of Exposure Science & Environmental Epidemiology*, 29(2), 131–147.
- Sundseth, K., Pacyna, J. M., Pacyna, E. G., Pirrone, N., & Thorne, R. J. (2017). Global sources and pathways of mercury in the context of human health. *International Journal of Environmental Research and Public Health*, 14(1), 105.
- Taylor, M. D., Beyer-Robson, J., Johnson, D. D., Knott, N. A., & Bowles, K. C. (2018). Bioaccumulation of perfluoroalkyl substances in exploited fish and crustaceans: Spatial trends across two estuarine systems. *Marine Pollution Bulletin*, 131, 303–313.
- Tran, L., Reist, J. D., & Power, M. (2015). Total mercury concentrations in anadromous Northern Dolly Varden from the northwestern Canadian Arctic: A historical baseline study. *Science of the Total Environment*, 509, 154–164.
- US Environmental Protection Agency. (2007). Method 7471B: Mercury in solid or semisolid waste (manual cold-vapor technique).
- US Environmental Protection Agency. (2007). Method 7473: Mercury in solids and solutions by thermal decomposition, amalgamation, and atomic absorption spectrophotometry (SW 846).
- US Environmental Protection Agency. (2017). Method 30B: Mercury sorbent trap procedure.
- US Environmental Protection Agency. (2019). EPA's per- and polyfluoroalkyl substances (PFAS) action plan. EPA 823R18004. February 2019. https://www.epa.gov/sites/production/files/2019-02/documents/pfas_action_plan_021319_508compliant_1.pdf
- US Navy. (2017). Naval Arctic Research Laboratory—Imikpuk Lake drinking water investigation fact sheet. https://dec.alaska.gov/Applications/SPAR/PublicMVC/CSP/Download?documentID=33461&fileName=26855_Final%20Off-base%20sampling%20Fact%20Sheet_Barrow_08292017_lores.pdf
- Verbrugge, L. A. (2007). Fish consumption advice for Alaskans: A risk management strategy to optimize the public's health. *State of Alaska Epidemiology Bulletin*, 11(4), 1–39.
- Whiting, A. (2006). 2002–2004 Harvest Survey Program Report. Native Village of Kotzebue.