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# Elevated mercury and PCB concentrations in Dolly Varden (*Salvelinus malma*) collected near a formerly used defense site on Sivuqaq, Alaska

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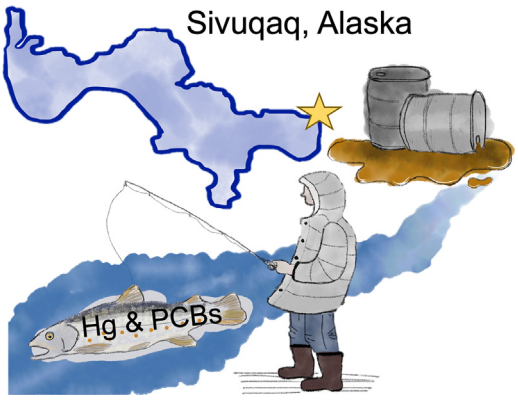
## HIGHLIGHTS

- Sivuqaq has 2 formerly used defense (FUD) sites, including at Northeast Cape (NEC).
- 89% of fish near the NEC FUD site exceeded the EPA's 0.049 μg/g Hg screening level.
- All fish near the NEC FUD site exceeded the EPA's 0.0015 μg/g PCB screening level.
- Relationships between $\delta^{13}C$, $\delta^{15}N$, [Hg], and [PCBs] were driven by the NEC FUD site.
- The NEC FUD site is a point source for Hg and PCB pollution of the local food web.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Environmental pollution causes adverse health effects in many organisms and contributes to health disparities for Arctic communities that depend on subsistence foods, including the Yupik residents of Sivuqaq (St. Lawrence Island), Alaska. Sivuqaq's proximity to Russia made it a strategic location for U.S. military defense sites during the Cold War. Two radar surveillance stations were installed on Sivuqaq, including at the Northeast Cape. High levels of persistent organic pollutants and toxic metals continue to leach from the Northeast Cape formerly used defense (FUD) site despite remediation efforts. We quantified total mercury (Hg) and polychlorinated biphenyl (PCB) concentrations, and carbon and nitrogen stable isotope signatures, in skin and muscle samples from Dolly Varden (*Salvelinus malma*), an important subsistence species. We found that Hg and PCB concentrations significantly differed across locations, with the highest concentrations found in fish collected near the FUD site. We found that 89% of fish collected from near the FUD site had Hg concentrations that exceeded the U.S. Environmental Protection Agency's (EPA) unlimited Hg-contaminated fish consumption screening level for subsistence fishers (0.049 μg/g). All fish sampled near the

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FUD site exceeded the EPA's PCB guidelines for cancer risk for unrestricted human consumption (0.0015 μg/g ww). Both Hg and PCB concentrations had a significant negative correlation with $\delta^{13}C$ when sites receiving input from the FUD site were included in the analysis, but these relationships were insignificant when input sites were excluded. $\delta^{15}N$ had a significant negative correlation with Hg concentration, but not with PCB concentration. These results suggest that the Northeast Cape FUD site remains a point source of Hg and PCB pollution and contributes to higher concentrations in resident fish, including subsistence species. Moreover, elevated Hg and PCB levels in fish near the FUD site may pose a health risk for Sivuqaq residents.

## 1. Introduction

Environmental pollution poses adverse health risks to people and may contribute to health disparities for communities that depend on subsistence foods, such as fish and marine mammals (Burger et al., 2007; Scrudato et al., 2012). Many environmental contaminants, including mercury (Hg) and polychlorinated biphenyls (PCBs), accumulate in arctic environments from both local point sources of pollution and long-range atmospheric transport from lower latitudes (AMAP, 2011; Scheringer et al., 2004; Wania and Mackay, 1993). Alaska contains over 600 formerly used defense (FUD) sites dating from World War II and the Cold War, with over 500 sites identified for environmental cleanup by the U.S. Army Corps of Engineers and the Alaska Department of Environmental Conservation (USDOI, 2016). Many of these contaminated sites are located near rural communities that rely on subsistence foods and these communities may be directly affected by environmental contamination through their diet (Welfinger-Smith et al., 2011; Williams and Cravez, 2018). As a result, communities located near FUD sites may experience elevated risk of adverse health effects (Carpenter et al., 2005; Miller et al., 2013; von Hippel et al., 2018).

Mercury is a toxic metal that enters the environment from natural (e.g., volcanic eruptions and weathering of Hg-containing deposits) and anthropogenic sources (e.g., coal burning, mining, and electronics, including those used by the military at FUD sites) (Wang et al., 2004). Once in the environment, inorganic Hg can be transformed by microbes into the organic form methylmercury (MeHg). MeHg is a potent toxicant that bioaccumulates in animal tissues and biomagnifies within food webs (Peng et al., 2016). Point sources of Hg pollution may have a greater effect on MeHg bioaccumulation patterns at higher latitudes due to increased trophic magnification slopes and slower excretion rates, leading to increased accumulation in biota (Lavoie et al., 2013; Trudel and Rasmussen, 1997). MeHg exposure disrupts a variety of health endpoints in vertebrates, including cognitive, thyroidal, adrenal, and gonadal functions (Wada et al., 2009). High levels of Hg in humans have been linked to low birth weights, cognitive deficiencies, and reproductive impairment (Clarkson et al., 2003; Guallar et al., 2002; Harada, 1995; Oken et al., 2005).

PCBs are highly stable synthetic chemicals that were used in plasticizers, capacitors, sealants, flame retardants, hydraulic lubricants, electrical transformers, and pesticides until bans on their production and use came into force beginning in the 1970s (Safe, 1994). Despite the inclusion of PCBs under provisions of the Stockholm Convention for global elimination, PCBs remain ubiquitous in the environment and are frequently detected in biotic and abiotic samples, including near Cold War era FUD sites (von Hippel et al., 2018). PCBs are lipophilic compounds that readily bioaccumulate and biomagnify within the food web (Aronson et al., 2000; Burreau et al., 2004; Dewailly et al., 1989). PCBs are known carcinogenic, immunotoxicant, and endocrine-disrupting compounds that negatively affect a wide range of organisms, including humans (Schell et al., 2014; Wassermann et al., 1979; Wurgler and Kramers, 1992).

Dietary exposure is the primary route of MeHg bioaccumulation in animals (Bloom, 1992; Hall et al., 1997; Klaverkamp et al., 1983), and fish tend to accumulate MeHg in muscle tissue (Bloom, 1992; Harley et al., 2015). Similarly, dietary exposure is the primary route of PCB accumulation in animals (Ampleman et al., 2015; Crinnion, 2011), but PCBs preferentially accumulate in lipids (Müllerová and Kopecký, 2007). Thus, exposure to MeHg and PCBs via consumption of fish poses a direct risk for higher trophic level, piscivorous organisms (Peng et al., 2016; Van Oostdam et al., 2005). People who consume large quantities of fish are at higher risk of toxicity due to chronic dietary exposure. Many Alaska Native communities rely on high trophic level, lipid-rich subsistence foods, including fish and marine mammals. For instance, rural communities in Western Alaska consume an average of 183 g fish per adult per day while the general U.S. population consumes only 22–24 g fish/adult/day (Polissar and Neradilek, 2019; USEPA, 2014). Food preparation methods do not eliminate MeHg (Morgan et al., 1997), which limits the ability of subsistence fishers to avoid contaminant exposure. Furthermore, these communities are often located near point sources of pollution, such as FUD sites (Scrudato et al., 2012; von Hippel et al., 2016).

Sivuqaq (St. Lawrence Island), Alaska is a remote island located in the northern Bering Sea, approximately 200 km off the west coast of mainland Alaska (Fig. 1). The island's proximity to Russia (formerly USSR) during the Cold War made it a strategic location for U.S. military defense sites. Two radar surveillance stations were installed on Sivuqaq as part of an early warning system to detect USSR aircraft, including a White Alice Communications Station on the Northeast Cape. The Northeast Cape FUD site was in operation from 1954 to 1972, until technological advances rendered it obsolete and it was abandoned (Carpenter et al., 2005). High levels of persistent organic pollutants and toxic metals continue to leach from the Northeast Cape FUD site despite large-scale remediation that occurred in the early 2000s (Scrudato et al., 2012; von Hippel et al., 2018; Welfinger-Smith et al., 2011). Scrudato et al. (2012) reported a six-fold increase in Hg concentrations in soils sampled near the Northeast Cape FUD site as compared to nearby background sites, as well as elevated concentrations of chlorinated contaminants such as PCBs, which suggests that fish and wildlife in this area may also be contaminated with these contaminants.

Nearly all residents of Sivuqaq have familial ties to Siberia and identify themselves as Siberian Yupik or Sivuqaq Yupik. They have a subsistence culture that relies heavily on the harvest of marine mammals and fish (AKDFG, 2006; Welfinger-Smith et al., 2011). Savoonga is the closest extant village to the Northeast Cape FUD site. In a 2009 survey of subsistence harvests in Savoonga, 82% of households reported harvesting a variety of fish species, including fish caught in both marine and freshwater habitats (Tahbone and Trigg, 2010). Although the original residents of Northeast Cape were displaced and relocated to Savoonga when the military site was constructed, many residents of Savoonga still maintain hunting and fishing camps in proximity to the FUD site. Carpenter et al. (2005) found that residents with camps at Northeast Cape had higher levels of PCBs in their blood serum than did residents of Gambell, a village on the western side of the island. This finding validated concerns of the Northeast Cape families and suggests that people who continue to subsist at Northeast Cape continue to be exposed to higher levels of military-associated contaminants, including Hg and PCBs, through consumption of fish caught near the FUD site.

The Suqitughneq (Suqi) River runs north through the Northeast Cape FUD site from the Kinipaghulghat Mountains to the Bering Sea. Several native fishes inhabit the Suqi River, including the salmonid Dolly Varden (*Salvelinus malma*), which is a subsistence food source for the local people. Residents report that the Suqi River was once among the most productive salmonid streams on the island, but that fuel spills and other contaminants from military operations at Northeast Cape greatly diminished the fish populations and degraded their habitats (Miller et al., 2013). Residents report that these fish populations have not recovered.

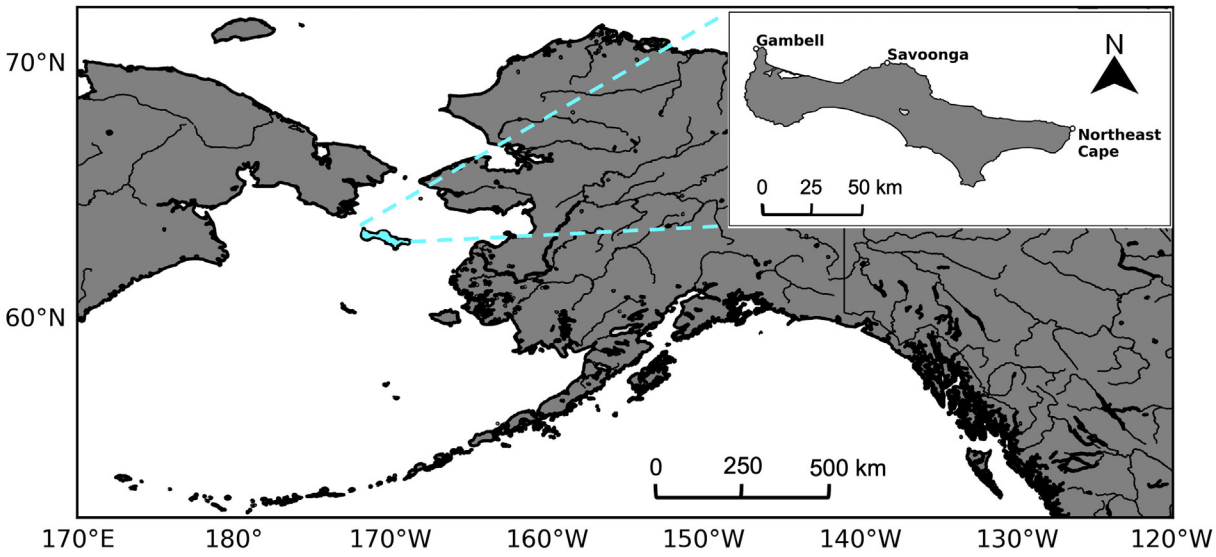


**Fig. 1.** Location of Sivuqaq (St. Lawrence Island), Alaska, and Northeast Cape.

von Hippel et al. (2018) showed that resident fish species collected downstream of the Northeast Cape FUD site in the Suqi River are negatively impacted by military contaminants, including PCBs. Effects on fish included endocrine disruption and altered gene expression. These results raise the question of whether local contamination also poses a health risk to residents who consume fish caught in the Suqi River. Given the findings of Scrudato et al. (2012) that Hg and PCBs are primary military contaminants of concern in the Suqi River watershed, and the findings of von Hippel et al. (2018) that highly chlorinated PCBs occur at high concentrations in small fish in the Suqi River (ninespine stickleback [*Pungitius pungitius*] and Alaska blackfish [*Dallia pectoralis*]), we decided to focus on Hg and PCBs in Dolly Varden as representative contaminants in subsistence fish.

We quantified total Hg and PCB concentrations in skin and muscle tissue of Dolly Varden collected from the Suqi River to determine if concentrations were elevated in fish living near the Northeast Cape FUD site and if those concentrations exceeded the U.S. Environmental Protection Agency (EPA) advisory limits for safe consumption. Because both MeHg and PCBs bioaccumulate and biomagnify, we also analyzed stable isotope ratios of carbon and nitrogen to examine associations between dietary carbon source (e.g., freshwater vs. marine), trophic level, site variability in isotope ratios, and contaminant concentration. We hypothesized that Dolly Varden collected from sites receiving inflow from the Northeast Cape FUD site would have significantly elevated total Hg and PCB concentrations and that the stable isotope ratios would account for some of the variation of both contaminants. Additionally, we sex-genotyped all fish to learn how variation in contamination relates to sex.

## 2. Methods

### 2.1. Fish collection

We collected Dolly Varden from the Suqi River in June–July of 2012, 2013, and 2015 using unbaited 0.32 and 0.64 cm wire-mesh minnow traps ($n = 41$ total fish). We collected fish from eight long-term monitoring sites (von Hippel et al., 2018) along the Suqi River (Fig. 2) and one reference site on the Tapisaggak (Tapi) River, located in an adjacent watershed 5 km to the east of the Northeast Cape FUD site. All fish were euthanized in the field with an overdose of pH-neutral fish anesthetic MS-222 (University of Alaska Anchorage IACUC #159870-20 and #439949-1). Samples were stored on ice in the field and transferred to −80 °C in the laboratory until analysis. We photographed each fish with a unique identifier and measured the standard length prior to dissecting a ~2 × 4 × 1 cm section of skin and muscle tissue, representing typical tissues consumed by residents, for the quantification of total Hg, PCBs, and stable isotope ratios. We also sampled a fin clip (2 × 2 mm) for sex genotyping. Dolly Varden were not captured at every site each year, which prevented us from comparing Hg and PCB concentrations across all sampling years.

### 2.2. Sex genotyping

Dolly Varden were sexed by multiplex PCR genotyping of the male-specific Y-chromosome gene *sdY* and the *18S* gene (present in both males and females) following the protocol described in Yano et al. (2013). We ran genomic DNA extractions on 15 mg of fin tissue for each fish using the Qiagen DNeasy Blood & Tissue kit. We quantified DNA concentrations using a ThermoFisher Nanodrop 1000 Spectrophotometer. PCR amplification of the *sdY* and *18S* genes was performed using 0.4 μM of each *sdY* primer, 0.2 μM of each *18s* primer, 50–100 ng of DNA, 1.5 units of TaKaRa Taq DNA polymerase (TaKara Bio), 1.5 mM $MgCl_2$, and 2.5 μl of 10× PCR buffer per 25 μl reaction. PCR products were electrophoresed on 2% agarose gels and imaged using an Alpha Innotech FluorChem SP imager.

### 2.3. Mercury analysis

We quantified total Hg concentrations in samples containing both skin and muscle tissue from 41 Dolly Varden samples. We extracted and freeze-dried each section (~2 g wet weight; ww) of skin and muscle tissue from frozen whole-body fish. We used a subsample of 0.25–0.50 g dried tissue for each homogenate sample and digested them using an open acid digestion protocol outlined by Mohammed et al. (2017). We added 1.5 ml hydrochloric acid (HCl; Fisher Chemical), 1 ml of 5% potassium permanganate in 0.1% HCl ($KMnO_4$; Fisher Chemical), 500 μl hydrogen peroxide ($H_2O_2$; Medivators Inc.), and 7 ml deionized (DI) water to each sample and let them sit overnight at room temperature. After approximately 12 h, we capped samples and placed them in a hot water bath at 85 °C for 2 h. Following digestion, we filtered samples using a vacuum filtration apparatus to remove residual tissue debris from each sample. We then added hydroxylamine hydrochloride ($NH_2OH{\cdot}HCl$; Medivators Inc.) to each sample to neutralize the $KMnO_4$ added during the digestion step and diluted each sample to 15 ml with DI water.

MeHg typically accounts for >90% of bioaccumulated Hg in fish, with the highest MeHg concentrations found in muscle tissue (Bloom, 1992). Previous work on Dolly Varden collected from the Yukon Territory, Canada showed that MeHg accounted for over 91% of the total bioaccumulated Hg (Tran et al., 2016). Because of this, we measured total Hg concentration as a proxy for MeHg concentration. We analyzed samples

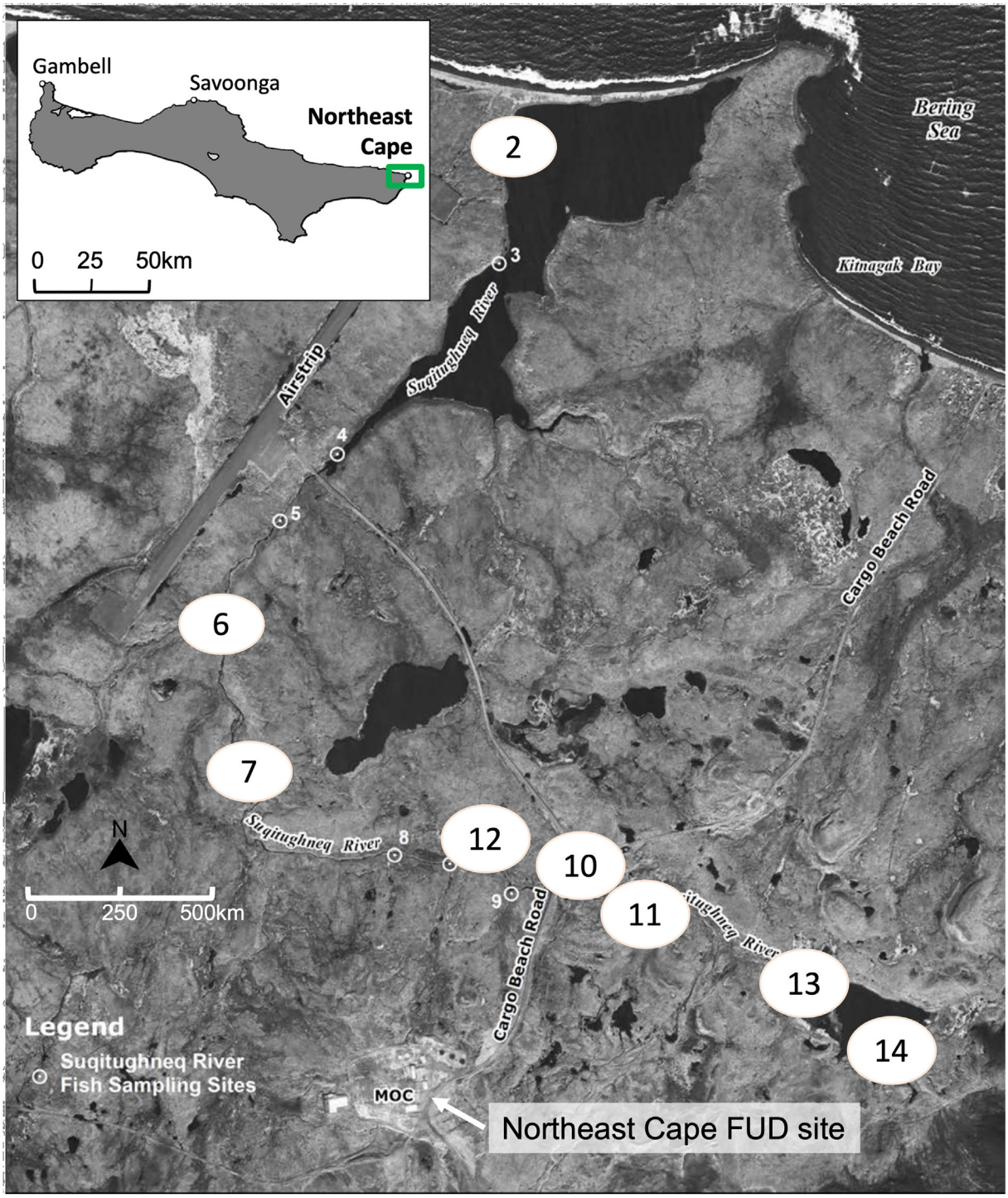


**Fig. 2.** Sampling site locations along the Suqitughneq (Suqi) River on the Northeast Cape of Sivuqaq (St. Lawrence Island), Alaska. Site numbers are long-term monitoring sites. Sites 2, 6, and 7 are designated as downstream sites because they are located downstream of the formerly used defense (FUD) site. Sites 10, 11, and 12 are designated as input sites because they receive direct input from the FUD site. Sites 13 and 14 are designated as upstream sites due to their location upstream of the FUD site. Fish were also collected from the Tapisaggak (Tapi) River in an adjacent watershed (not pictured), which serves as a reference site.

for total Hg concentrations on a Perkin Elmer FIMS-100 cold-vapor atomic absorption analyzer with an attached auto-sampling unit using argon gas. Tin (II) chloride dihydrate ($SnCl_2$; Fisher Chemical) was used as the reducing agent and 3% (*v*/v) HCl as the carrier solution. For quality assurance and quality control, each analysis contained a 10 ppb certified tuna fish flesh homogenate reference sample (International Atomic Energy Agency [IAEA] 436), a 1 ppb spiked Dolly Varden sample, an extraction blank, and an analysis blank that we prepared along with target samples. Analysis blanks and standards contained all digestion reagents to control for the sample matrix. Additionally, we re-analyzed a random subset of five samples to determine reproducibility. Calibration standards for total Hg analysis were made from 1000 μg/ml Hg standard stock (PerkinElmer, catalog #N9300174) and serially diluted to 10 ppb, 5 ppb, and 0.5 ppb concentrations for instrument calibration. A certified standard (10 ppm Hg; Inorganic Ventures, catalog #MSHG-10PPM-125ML) was diluted to 1 ppb and run as an unknown sample for quality control. Analytical extraction efficiencies

averaged 98 ± 0.04% ($n$ = 4) for certified reference material and 96 ± 0.07% (n = 4) for spiked recoveries.

We compared total Hg concentrations to the EPA's unlimited consumption screening level for subsistence fishers consuming Hg-contaminated fish (0.049 μg/g in fish; USEPA, 2000). The EPA recommends using this threshold in either dry or wet weight depending on which measurement more accurately reflects the method of fish preparation. Sivuqaq residents prepare fish in a variety of ways, including dried and half dried (fish are frozen and later eaten with seal oil), boiled, fried, and baked. For this reason, we report the EPA unlimited consumption guideline in both dry weight (dw; 0.049 μg/g) and the wet weight equivalent (0.049 μg/g ww = 0.2 μg/g dw). We considered the EPA's unlimited screening level to be more appropriate for Sivuqaq than the less conservative EPA Fish Tissue Residue Criterion for MeHg (0.3 Hg μg/g ww; USEPA, 2001) because Dolly Varden are a subsistence food for residents and the consumption rate for subsistence fishers more accurately reflects the fish consumption rate of rural Alaskan communities.

### 2.4. PCB analysis

We analyzed total PCB concentrations in freeze-dried skin and muscle tissue from the same 41 individual Dolly Varden analyzed for Hg using methods described in Zheng et al. (2020). Approximately 2 g of fresh tissue was freeze dried, pre-treated with diatomaceous earth, and spiked with surrogate standards (PCB-14, -65, and -166) before extraction using a Dionex Accelerated Solvent Extractor 350 with hexane and acetone (1:1, $v$/v) at 90 °C and 1500 psi over three static cycles. We separated 10% of the extract to determine lipid content by gravimetric analysis. The remaining 90% of extract was reduced to 1 ml by rapid evaporation using nitrogen gas and cleaned on a multilayer silica column containing glass wool, 2 cm of neutral silica, 5 cm of acid silica, 2 cm of neutral silica, and 1 cm of sodium sulfate. We eluted PCBs with 40 ml hexane and dichloromethane (1:1, $v$/v) and then concentrated, solvent exchanged to hexane, reduced again to 1 ml with nitrogen gas, and spiked with internal standards (PCB-30 and -204). We analyzed samples using an Agilent 7890 gas chromatograph (GC) equipped with an electron capture detector and a DB-5MS fused silica capillary column (60 m × 0.32 mm × 0.25 μm). We used helium as the carrier gas and held the flow rate constant at 4.3 ml/min. The GC thermocycler was set to the following sequence: 100 °C for 1 min, 1 °C/min to 240 °C, 10 °C/min to 280 °C, and held for 20 min. We set the injector (splitless mode) and the electron capture detector temperatures to 250 °C and 350 °C, respectively. Six procedural blank and six matrix spike recovery samples were included in the extraction of all fish samples. All PCB concentrations were blank corrected and method detection limits (MDLs) were set at three times the standard deviation of the target analyte levels detected in the procedural blanks. For the compounds not detected in the blanks, MDLs were based on a signal-to-noise ratio of three. The average absolute recovery for the spiked samples (mean ± standard error) was 85 ± 1.7%. The mean (with standard errors) recoveries of the surrogate standards were 84 ± 3.8%, 84 ± 3.1% and 86 ± 4.1% for PCB-14, -65 and -166, respectively. We assumed equal contributions for co-eluted PCB congeners and assigned equal parts of the total PCB concentration for each congener (e.g., PCB 4 and PCB 10 each received half of the reported PCB concentration).

### 2.5. Stable isotope analysis

We analyzed stable isotopes to determine if total Hg and PCB concentrations were associated with either nitrogen isotope ratios (reflecting trophic level) or carbon isotope ratios (reflecting dietary source of carbon). We collected approximately 1 mg of freeze-dried skin and muscle tissue from each fish analyzed for contaminants and homogenized tissue in a tissue mill with 3.2 mm steel beads. We sealed each sample into a tin capsule and analyzed for $\delta^{13}C$, $\delta^{15}N$, %C, %N, and C/N at the Colorado Plateau Stable Isotope Laboratory. We ran samples on a ThermoScientific Delta V Advantage Isotope Ratio Mass Spectrometer (Thermo-Electron Corp., Bremen, Germany) configured through a Finnigan CONFLO III coupled to a Carlo Erba NC2100 elemental analyzer (CE Instruments, Milan, Italy). All instrumentation was calibrated using a suite of standards from the IAEA, including drift and linearity standards (peach leaves; SRM 1547), isotope standards (IAEA CH6 & IAEA CH7 for $\delta^{13}C$ and IAEA N1 & IAEA N2 for $\delta^{15}N$), elemental standards (including acetanilide, cystine, and methionine), and secondary check standards (including NIST pine needles and NIST apple leaves). Nine replicates of the internal laboratory working standard (peach leaves; SRM 1547) were interspersed every 11 fish samples to ensure measurement reproducibility ($\delta^{13}C = -26.15 \pm 0.04‰$, $\delta^{15}N = 1.88 \pm 0.04‰$). External isotope reference standard reproducibility for carbon and nitrogen was below ±0.2‰.

### 2.6. Statistical analysis

We conducted all statistical analyses using R statistical computing software, R version 4.1.0 (2009–2021 RStudio, Inc.). We used an alpha level of 0.05 to determine significance for all statistical tests. Due to small sample sizes, we grouped collection sites according to their location relative to the FUD site. Sites 2, 6, and 7 are located downstream of the Northeast Cape FUD site and were pooled as downstream sites ($n$ = 6 fish; Fig. 2). Sites 13 and 14 are located upstream of the FUD site and were pooled as upstream sites ($n$ = 10 fish; Fig. 2). Sites 10, 11, and 12 receive direct input from the Northeast Cape FUD site and were pooled as input sites ($n$ = 21 fish; Fig. 2). Samples collected from the Tapi River were designated as the reference site ($n$ = 4 fish). We ran a linear mixed model (results not shown) to account for the potential random effect of pooled collection sites at each location; however, we found no differences by site and removed site as a variable from our model. Furthermore, the mixed effects model and an ANOVA model produced similar findings for modeling Hg and PCB concentrations.

We established whether sampling location was a significant factor in predicting total Hg (per dw) and PCB concentrations (per ww) using ANOVA models. Total Hg and total PCB concentrations were natural log transformed for ANOVAs and Pearson product-moment correlations to correct for abnormal data distributions and heteroskedasticity. We confirmed ANOVA results with the non-parametric Kruskal-Wallis and Dunn's multiple comparisons tests to account for small sample sizes and non-normal distributions of our data. We did not use a mixed effect model because the random effects of fish sex and sampling location were not significant (results not shown). We employed a one-way ANOVA to model total Hg concentration because sampling location was the only significant variable for the model. We employed a two-way ANOVA to model total PCB concentrations because both sampling location and sex were significant predictors. We did not length standardize total Hg concentrations because all fish were of the same species and the Pearson product-moment correlation between total Hg and standard length was not significant ($r = 0.03, n = 41, p = 0.843$). We corrected for pairwise comparisons of total Hg and PCB concentrations for both year and location using the Tukey method. We analyzed sex differences using Mann-Whitney U non-parametric tests to account for non-normality and small sample sizes. We were unable to compare year-to-year differences in Hg and PCB concentrations because we did not capture fish at each site for all sampling years. Only input sites had fish sampled from all three years. We used raw data for non-parametric analyses. We analyzed $\delta^{15}N$ and $\delta^{13}C$ isotope ratios separately against both total Hg and PCB concentration and standard length to determine relationships using Pearson product-moment correlations. To compare our results to those in the literature, we converted Hg dry weight concentrations to wet weight using the equation from Canham et al. (2021) and assuming a mean fish moisture content of 76% as reported by Eagles-Smith and Ackerman (2014) (see Table S1, which provides both ww and dw values for Hg and both ww and lipid weight (lw) values for PCBs).

## 3. Results

### 3.1. Fish samples and model selection

The concentrations of Hg and PCBs in Dolly Varden suggest that the Northeast Cape FUD site remained a major source of contamination during the study period. Of the 41 Dolly Varden caught, 23 were male and 18 were female. Mean total Hg concentration (± standard error) in females (0.217 ± 0.033 μg/g dw) was significantly higher than in males (0.133 ± 0.022 μg/g dw) across all samples (Mann-Whitney $U$ test, $p = 0.030$); however, this is likely an artifact of the disproportionate number of female fish caught at input sites (15 females versus 6 males). PCB concentration did not differ by sex. We found the highest mean total Hg concentrations in fish collected at input sites receiving water flow directly from the Northeast Cape FUD site (0.224 ± 0.03 μg/g dw; Table 1 and Fig. 3). Fish collected at sites upstream of the FUD site had the highest mean total PCB concentrations (31.7 ± 5.33 ng/g ww; Table 1 and Fig. 4). Congener-specific concentrations of PCBs are presented in Table S2. Dolly Varden samples had a mean lipid content of 2.16%.

We found that standard length was not a significant predictor of total Hg or PCB concentration. Dolly Varden standard length ranged between 81.5 and 178.0 mm, with a mean of 120.5 ± 3.5 mm, and was comparable among sampling locations and years. Because standard length did not correlate with either total Hg concentration (Pearson correlation, $r = 0.03$, $n = 41$, $p = 0.843$) or total PCB concentration (Pearson correlation, $r = -0.03$, n = 41, $p = 0.876$), we excluded standard length from the models (Suppl. Fig. 1). Similarly, likelihood ratio tests indicated that the interactions between standard length, location, and year were not significant.

### 3.2. Total Hg concentration

Model selection tests indicated that sampling location was the only significant covariate in modeling Hg concentration. Total Hg concentrations in Dolly Varden differed significantly across sampling locations (ANOVA, $F_{3,37} = 5.56$, $p = 0.003$; Kruskal-Wallis $\chi^2 = 12.34$, $p = 0.006$). Total Hg concentrations in fish collected at input sites were 4.3-fold higher than in fish from reference sites (ANOVA, $p = 0.015$; Dunn's test, $p = 0.018$) and 1.7-fold higher than in fish from upstream sites (ANOVA, $p = 0.014$; Dunn's test, $p = 0.040$; Fig. 3).

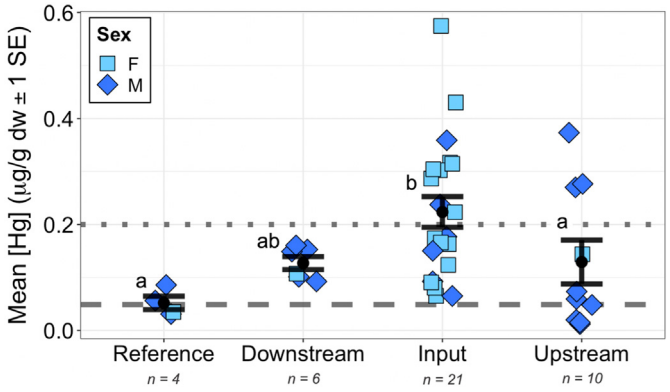


**Fig. 3.** Total mercury (Hg) concentration in Dolly Varden collected from four locations over three years at the Northeast Cape on Sivuqaq (St. Lawrence Island), Alaska. These include sites downstream of the formerly used defense (FUD) site, input sites adjacent to the FUD site, sites upstream of the FUD site, and a reference site in an adjacent watershed. Locations with different letters significantly differ in total Hg concentration ($p < 0.05$). A one-way ANOVA suggests that fish collected at input sites had significantly higher total Hg concentration than did fish collected at reference and upstream sites ($p = 0.015$ and $p = 0.014$, respectively). The dashed horizontal line indicates the U.S. EPA screening level of 0.049 μg/g dw for unlimited fish consumption for subsistence fishers using a drying cooking method. The dotted horizontal line indicates the U.S. EPA screening level of 0.2 μg/g dw for unlimited fish consumption for subsistence fishers using a non-drying cooking method.

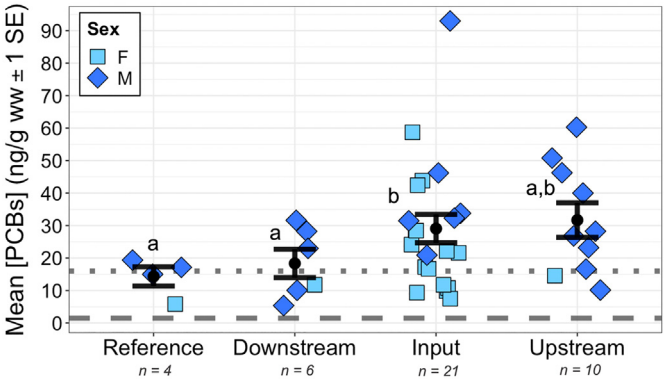


**Fig. 4.** Total PCB concentration in Dolly Varden collected from four locations over three years at the Northeast Cape on Sivuqaq (St. Lawrence Island), Alaska. Locations with different letters significantly differ in total PCB concentration ($p < 0.05$). A two-way ANOVA with location and sex as fixed factors suggests that fish collected at input sites had significantly higher total PCB concentration than did fish collected at reference and downstream sites ($p = 0.043$ for both comparisons). The dashed horizontal line indicates the U.S. EPA screening level for unlimited fish consumption for PCBs (cancer risk for human consumption; 0.0015 μg/g ww). The dotted horizontal line indicates the U.S. EPA screening level for three servings (8 oz.) of fish per month (cancer risk for human consumption; 0.016 μg/g ww).

### 3.3. Total PCB concentration

Model selection tests indicated that sampling location and sex were the only significant covariates in modeling PCB concentration. A two-way ANOVA model with location and sex as fixed factors best explained the variance in PCB concentration across samples. Total PCB concentrations in Dolly Varden differed significantly among sampling locations and sex (ANOVA $F_{4,36} = 4.192$, $p = 0.007$; Fig. 4). Fish collected at input sites had significantly higher total PCB concentrations than did reference fish (2.0-fold higher; ANOVA, $p = 0.043$) and downstream fish (1.6-fold higher; ANOVA, p = 0.043), but did not differ from upstream fish. Total PCB concentration did not significantly correlate with total Hg concentration (Pearson, $r = -0.21$, $n = 41$, $p = 0.178$). Dolly Varden collected at Suqi River sites tended to have heavier PCBs than those collected at the Tapi River reference site (Fig. 5). *Hepta-* and *octa*-chlorinated congeners were significantly higher in Suqi River Dolly Varden than in Tapi River Dolly Varden (Kruskal-Wallis, $\chi^2 = 7.48$, $p = 0.006$ and $\chi^2 = 5.78$, $p = 0.016$, respectively).

### 3.4. Stable isotope signatures

Given that both Hg and PCBs bioaccumulate and biomagnify, we hypothesized that nitrogen and carbon stable isotope ratios would correlate with total Hg and PCB concentrations, but the relationships were not as

**Table 1**
Mean total mercury (Hg) and mean total polychlorinated biphenyl (PCB) concentrations in Dolly Varden (±1 standard error) collected from four locations over three years at the Northeast Cape on Sivuqaq (St. Lawrence Island), Alaska. These include sites downstream of the formerly used defense (FUD) site, input sites adjacent to the FUD site, sites upstream of the FUD site, and a reference site in an adjacent watershed.

| Location | Mean [total Hg] μg/g dw | Mean [total PCBs] μg/g ww |
|---|---|---|
| Reference | 0.052 ± 0.012 | 0.014 ± 0.003 |
| Downstream | 0.127 ± 0.012 | 0.018 ± 0.004 |
| Input | 0.224 ± 0.029 | 0.029 ± 0.004 |
| Upstream | 0.129 ± 0.041 | 0.032 ± 0.005 |

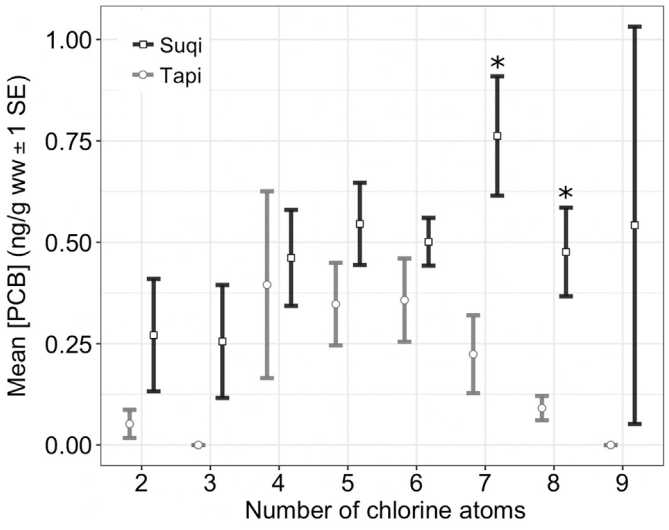


**Fig. 5.** Differences in PCB concentration, based on the number of chlorine atoms present on the biphenyl ring, in Dolly Varden collected from two rivers on the Northeast Cape of Sivuqaq (St. Lawrence Island), Alaska. The Suqitughneq (Suqi) River (black square; $n = 37$ fish) receives inflow from the Northeast Cape formerly used defense (FUD) site. The Tapisaggak (Tapi) River (gray circle; $n = 4$) is located in an adjacent watershed and serves as a reference site. Lightly- to intermediately-chlorinated PCBs (those with 2–6 chlorine atoms) did not differ significantly between Suqi River and Tapi River Dolly Varden, though the means were always lower in the Tapi River. *Hepta-* and *octa*-chlorinated congeners were significantly higher in Suqi River Dolly Varden than in Tapi River Dolly Varden (Kruskal-Wallis $\chi^2 = 7.48, p = 0.006$ and $\chi^2 = 5.78, p = 0.016$, respectively), suggesting that the Northeast Cape FUD site is a point source of PCBs.

expected. Nitrogen stable isotope ratios ranged from 5.5‰ to 11.4‰, spanning nearly two trophic levels (based on a trophic fractionation of 3.4‰; Post, 2002), with a mean of 8.66 ± 0.18‰. $\delta^{15}N$ did not vary by site and did not correlate with either total PCB concentration or standard length. We found a significant negative relationship between total Hg concentration and $\delta^{15}N$ when input sites were included (Pearson correlation, $r = -0.31, n = 41, p = 0.049$). This relationship was slightly stronger when input sites were excluded from the analysis (Pearson correlation, $r = -0.36, n = 20, p = 0.116$; Fig. 6); however, it was no longer significant, likely due to low statistical power. The power to detect a significant effect across all samples and at a linear correlation coefficient of 0.3 was 49%. The power fell to 26% when input sites were removed from the analysis.

Carbon stable isotope ratios ranged from −34.0‰ to −22.1‰, with a mean of −29.3 ± 0.4‰. We found a significant positive relationship between $\delta^{13}C$ and $\delta^{15}N$ (Pearson correlation, $r = 0.51, n = 41, p < 0.001$; Fig. S2). $\delta^{13}C$ values differed significantly by site (ANOVA, $F_{3,37} = 8.18$, $p < 0.001$; Kruskal-Wallis, $\chi^2 = 14.87, p = 0.002$). Fish collected at input sites had significantly lower $\delta^{13}C$ values than did fish collected at reference or upstream sites (ANOVA, $p < 0.001$ and $p = 0.047$; Dunn's test, $p = 0.005$ and $p = 0.034$, respectively). We found significant negative relationships between $\delta^{13}C$ and both total Hg (Pearson correlation, $r = -0.35$, n = 41, $p = 0.026$) and total PCB concentration (Pearson correlation, $r = -0.36$, n = 41, $p = 0.021$) when input sites were included in the analysis (Fig. 7). However, these trends did not hold when fish collected at input sites were removed from the analysis (Pearson correlation, $r = -0.11$, $n = 20, p = 0.634$ and $r = -0.32$, n = 20, $p = 0.162$, respectively).

### 3.5. Contaminant values in relation to EPA health screening levels

We found that 89% of Dolly Varden collected from the Suqi River had total Hg concentrations in muscle and skin that exceeded the EPA screening level of 0.049 μg/g dw. This screening level applies to unlimited consumption of Hg-contaminated fish by subsistence fishers using a drying preparation method such as smoked, dried or half-dried fish (USEPA, 2000). Of these fish with high Hg concentrations, 64% were collected from input sites receiving water flow directly from the Northeast Cape FUD site (Fig. 3). All fish collected at input sites exceeded the 0.049 μg/g dw EPA threshold. For fish prepared without drying (e.g., raw, baked, fried, or boiled), the EPA recommends using the threshold as a wet weight value rather than a dry weight value (0.049 μg/g ww = 0.2 μg/g dw). At this threshold, 35% of Dolly Varden collected from the Suqi River exceeded the EPA screening level for safe consumption of Hg-contaminated fish, of which 77% were collected from input sites (Fig. 3). Nearly half (48%) of Dolly Varden from input sites exceeded this threshold, while none of the fish collected from the Tapi River reference site exceeded this threshold. If the less conservative EPA Fish Tissue Residue Criterion for MeHg

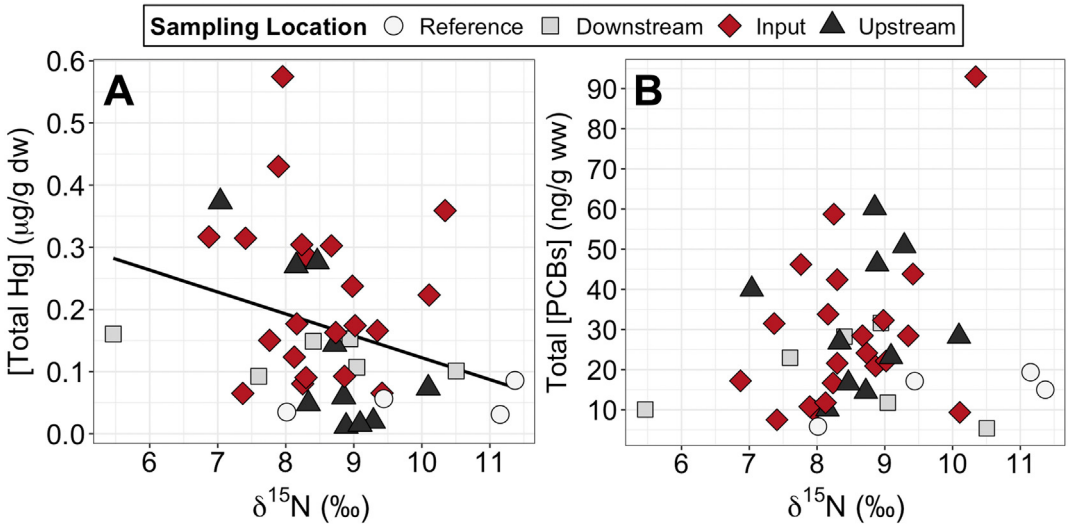


**Fig. 6.** $\delta^{15}N$ stable isotope signature plotted against A) total Hg concentration and B) total PCB concentration in Dolly Varden collected from the Northeast Cape on Sivuqaq (St. Lawrence Island), Alaska. Color and shape indicate sampling location: sites downstream of the formerly used defense (FUD) site (medium gray square), input sites adjacent to the FUD site (red diamond), upstream sites (black triangle), and reference sites (light gray circle). We found a significant negative relationship between $\delta^{15}N$ and total Hg concentration when input sites were included (Pearson correlation; $r = -0.31, n = 41, p = 0.049$). This relationship was stronger when input sites were removed from the analysis, but no longer significant (Pearson correlation; $r = -0.36, n = 20, p = 0.116$), likely due to the loss of statistical power. We did not find a significant correlation between $\delta^{15}N$ and total PCB concentration.

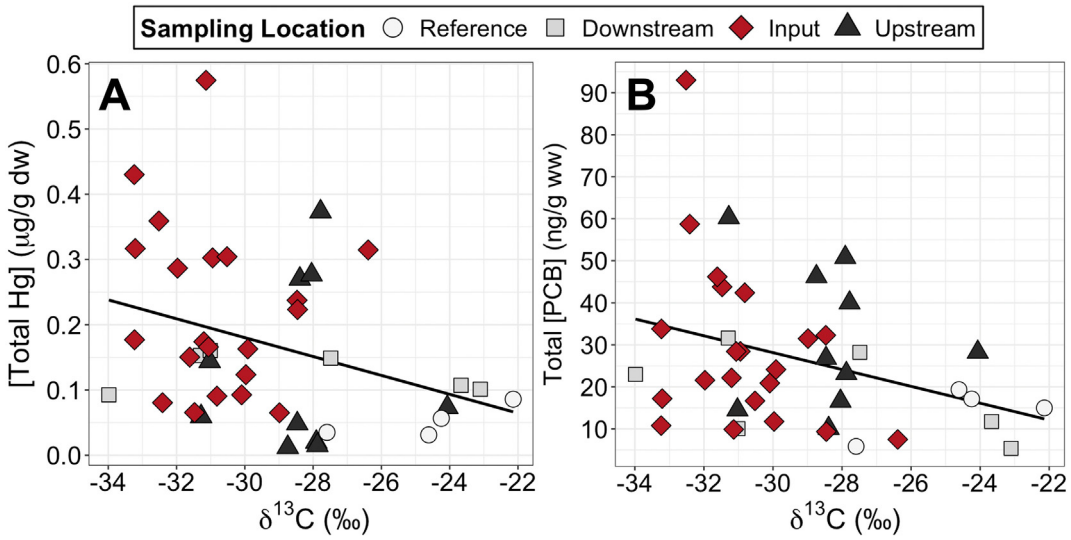


**Fig. 7.** $\delta^{13}C$ stable isotope signature plotted against A) total Hg concentration and B) total PCB concentration in Dolly Varden collected from the Northeast Cape on Sivuqaq (St. Lawrence Island), Alaska. Color and shape indicate sampling location: sites downstream of the formerly used defense (FUD) site (medium gray square), input sites adjacent to the FUD site (red diamond), upstream sites (black triangle), and reference sites (light gray circle). We found significant negative relationships between $\delta^{13}C$ and both total Hg concentration (Pearson correlation, $r = -0.35$, $n = 41$, $p = 0.026$) and total PCB concentration (r = −0.36, n = 41, $p = 0.021$) when input sites were included. However, this trend did not hold when input sites were removed from the analysis (Hg: $r = -0.11$, $n = 20$, $p = 0.634$; PCB: Pearson $r = -0.32$, n = 20, $p = 0.162$).

(0.3 Hg µg/g ww; USEPA, 2001) is employed, then none of the fish in the Suqi River exceeded the threshold. However, the 0.3 Hg µg/g ww criterion is based on a consumption rate of 17.5 g of fish per day, whereas rural communities in Western Alaska consume an average of 183 g (6.5 oz.) of fish per day, about 10 times as much (Polissar and Neradilek, 2019). Therefore, the more conservative unlimited consumption screening level is more appropriate for the Sivuqaq community.

All the Dolly Varden sampled in this study exceeded the EPA's guideline for unrestricted (128 oz./month) consumption of PCB-contaminated fish (cancer risk for human consumption of fish; 0.0015 µg/g [ppm] ww; Fig. 4; USEPA, 2000). Of the fish caught from the Suqi River, 73% exceeded the EPA safe consumption limit for 24 oz./month (cancer risk for human consumption of fish; 0.016 µg/g ww; Fig. 4) and 11% of fish surpassed the EPA safe consumption limit for 8 oz./month (cancer risk for human consumption of fish; 0.047 µg/g ww).

## 4. Discussion

### 4.1. FUD sites as a source of pollution

Despite large-scale remediation at the Northeast Cape FUD site completed in 2014 (ADEC, 2019), contamination at the site may continue to pose a health risk to Sivuqaq residents who conduct subsistence activities there. Mean total PCB concentrations presented in this study were substantially higher than previously reported by Welfinger-Smith et al. (2011) in Dolly Varden collected by subsistence hunters on Sivuqaq (0.00255 ± 0.00199 µg/g ww) and by Zheng et al. (2020) in ninespine stickleback collected from Troutman Lake on the opposite side of the island (0.01 µg/g ww), indicating that Dolly Varden have higher PCB concentrations at Northeast Cape than elsewhere on the island. Our results are consistent with those reported by von Hippel et al. (2018) for ninespine stickleback and Alaska blackfish collected at input and downstream sites in the Suqi River. Dolly Varden in the current study had higher PCB concentrations at upstream sites than did stickleback or blackfish, likely because Dolly Varden are stronger swimmers that are better able to swim between input and upstream sites.

PCB congener profiles can distinguish between point sources of pollution and accumulation via global distillation (Hong et al., 2012; Muir et al., 2000; von Hippel et al., 2018). Heavier PCB classes are less volatile and tend to remain relatively close to the pollution source whereas lighter weight PCBs undergo long-range transport and accumulate far from their sources of emission (Wania and Mackay, 1993). As a result, Arctic environments without point sources of pollution have a higher abundance of volatile PCBs (*tri-*, *tetra-*, and *penta*-chlorinated congeners) and relatively low levels of heavier PCBs. We found that fish collected at Suqi River sites had heavier PCBs than did fish collected at the Tapi River reference site, especially for classes with six or more chlorine atoms (Fig. 5). These results are consistent with von Hippel et al. (2018), who found that both stickleback and blackfish collected at sites downstream of the FUD site had significantly higher concentrations of heavier PCBs than did fish collected from upstream sites. These contaminant signatures indicate that the FUD site remains a point source of PCB pollution. These results imply that the consumption of fish caught at Northeast Cape, particularly near the FUD site, may contribute to disproportionately high exposure to PCBs for the Sivuqaq community.

Collectively, our data suggest that the Northeast Cape FUD site is a point source for both Hg and PCB contamination and that contaminants originating from the FUD site are being incorporated into the local food web. Our Hg findings are consistent with Scrudato et al. (2012), who analyzed sediment cores and found that the FUD site at Northeast Cape is the primary source of Hg in the watershed. PCB congener profiles in this study and in von Hippel et al. (2018) indicate that the Northeast Cape FUD site is a point source of PCB contamination. Furthermore, the FUD site appears to contribute to elevated serum levels of PCBs (Carpenter et al., 2005) and hexachlorobenzene (Byrne et al., 2015) in Savoonga residents who hunt and fish at Northeast Cape. Regardless of which conservative screening level is used, the FUD site continues to pose a risk for consumption of Suqi River fish due to exposure to Hg and PCBs.

More generally, these results point to the need to assess local sources of pollution in the Arctic, and not just atmospheric deposition due to global distillation, and to adequately remediate contaminated sites. FUD sites were often abandoned by the military without adequate containment of contaminants, leading to levels of local pollution that far exceed the background levels due to atmospheric deposition. Furthermore, FUD sites, including those on Sivuqaq (Zheng et al., 2020), are often a source of numerous other pollutants, including emerging contaminants. In that light, the Hg and PCBs should be viewed as indicator pollutants of a broader problem. Additionally, the Arctic is warming at approximately twice the

global average (Richter-Menge et al., 2019), which exacerbates the problem as POPs formerly sequestered in ice and permafrost re-enter food webs and the human diet.

### 4.2. Risks to wildlife

In addition to the risk posed to residents by elevated concentrations of Hg and PCBs, the FUD site also presents a risk to piscivorous wildlife. Dietary exposure to MeHg has been linked to reduced survival, growth, gonadal development, and spawning success in fish (Drevnick and Sandheinrich, 2003; Friedmann et al., 1996; Hammerschmidt et al., 2002). We found that 30% and 49% of Dolly Varden from the Suqi River exceeded MeHg dietary thresholds associated with biochemical (0.06 μg/g ww) and reproductive (0.04 μg/g ww) impairment in predatory fishes, respectively (Depew et al., 2012). None of the Dolly Varden exceeded MeHg thresholds for altered behavior (0.5 μg/g ww) or growth (1.44 μg/g ww) in fishes, or reproductive impairment for avian piscivores (0.16 μg/g ww; Fuchsman et al., 2017). For PCBs, we found that 89% of Dolly Varden from the Suqi River exceeded the lower limit of dietary threshold values associated with adverse effects in marine mammals (0.01 μg/g ww) (Kannan et al., 2000). However, PCB concentrations in Dolly Varden did not exceed Environment Canada's prey tissue residue guideline of 0.05 μg/g ww for wildlife consumers of fish (Alava et al., 2012). PCB levels were below thresholds for fish survival, growth, and reproduction (Berninger and Tillitt, 2019).

### 4.3. Life history variation and contaminant modeling

A growing body of research highlights the need to incorporate life history variables to accurately assess contaminant distributions and dynamics in food webs (Burke et al., 2020; Swanson and Kidd, 2010; Thomas et al., 2016). The focus of this study was to evaluate total Hg and PCB concentrations relevant to human consumption; however, investigation of life history differences of Dolly Varden may provide insights into both the movement of contaminants in this system and consumption risks. Specifically, differences in total Hg and PCB concentrations may be further explained by age, growth rates, and life history differences between anadromous and resident freshwater ecotypes (Howland et al., 2001). We would expect older Dolly Varden to have higher Hg and PCB values due to bioaccumulation. Because the FUD site is a point source of Hg and PCB pollution, we would expect resident freshwater fish to have higher concentrations than do anadromous fish, which complete most of their growth at sea (Rikardsen et al., 2006; Rikardsen et al., 2000). Anadromous fish are often more enriched in $\delta^{13}C$ than freshwater fish, so $\delta^{13}C$ signatures can be used to differentiate between freshwater and anadromous ecotypes (Guiry et al., 2020; Robson et al., 2016; Ruokonen et al., 2019). Although we did not differentiate between Dolly Varden ecotypes in this study, our $\delta^{13}C$ values are consistent with isotopic signatures of resident freshwater Dolly Varden (Hart et al., 2015; Tran et al., 2016). Tran et al. (2016) found that resident freshwater Dolly Varden had a mean muscle $\delta^{13}C$ value of −31.8‰, whereas anadromous Dolly Varden had a mean $\delta^{13}C$ value of −22.7‰. Studies on other northern fishes report similar findings. For example, Ruokonen et al. (2019) showed that $\delta^{13}C$ signatures in scales allowed the differentiation between resident freshwater and anadromous brown trout (means, −23‰ and −18‰, respectively). Guiry et al. (2020) found that $\delta^{13}C$ signatures in scale collagen could differentiate between resident freshwater *Oncorhynchus nerka* (kokanee; mean, −24‰) and anadromous *O. nerka* (sockeye salmon; mean, −16‰).

Stable isotope signatures of nitrogen also provide valuable ecological information relevant to contaminant modeling. For example, resident freshwater fish often have lower $\delta^{15}N$ isotopic signatures than do anadromous fish due to the fact that marine foods are enriched in heavier nitrogen isotopes (France, 1995), although these trends are not as strong as with $\delta^{13}C$ (McCarthy and Waldron, 2000; Ruokonen et al., 2019). Our $\delta^{15}N$ values are also consistent with isotopic signatures of resident freshwater Dolly Varden (Tran et al., 2016). Tran et al. (2016) reported that resident freshwater Dolly Varden had a significantly lower $\delta^{15}N$ signature than did anadromous Dolly Varden (means, 9.2‰ and 15.0‰, respectively). Future studies at Northeast Cape should distinguish between anadromous and resident freshwater Dolly Varden. This can be achieved in several ways, such as via analysis of strontium in otoliths (Campana, 1999; Hart et al., 2015; Kennedy et al., 2002). Such analyses may show that anadromous Dolly Varden harvested from the Suqi River are safe to eat, in which case the focus of public outreach could include education on the identification of anadromous Dolly Varden or mapping of areas of the Suqi River with the greatest proportion of anadromous Dolly Varden.

We observed a significant negative slope between $\delta^{13}C$ and both total Hg concentration and total PCB concentration, but those effects disappeared when fish collected at input sites were excluded from the analyses. Several possibilities warrant further investigation. These patterns may be an artifact of elevated total Hg and PCBs at the input sites and not due to established bioaccumulation and biomagnification patterns (Kidd et al., 2011; Lavoie et al., 2013), even though the $\delta^{15}N$ values spanned nearly two trophic levels (using a trophic fractionation value of 3.4‰; Post, 2002). Alternatively, differences in $\delta^{13}C$ values may be driven by life history. Individual fish on the right side of the graphs on Fig. 6 may be anadromous whereas those on the left side may be resident freshwater. We found that the correlation between $\delta^{15}N$ and total Hg increased when fish collected at input sites were excluded, suggesting that the lack of significance when these fish were excluded is likely due to a reduction in power to detect the correlation. In contrast, input sites appeared to drive the negative relationship between total Hg and $\delta^{13}C$ values.

### 4.4. Health disparities

The traditional knowledge of Sivuqaq residents speaks to a history of health disparities associated with the Northeast Cape FUD site. Elders have observed that cancers, reproductive disorders, and thyroid disease are more prevalent among people who engage in subsistence activities at Northeast Cape (Carpenter et al., 2005; Miller et al., 2013). In 2011, the tribal government of Savoonga requested a Public Health Assessment and Health Consultation by the Agency for Toxic Substances and Disease Registry (ATSDR) to assess health disparities associated with the Northeast Cape FUD site. The ATSDR assessment (USATSDR, 2017), released in 2017, concluded that contaminant levels in fish from the Suqi River do not pose a health risk to local residents. This conclusion was based on a 2001 sampling project conducted by the U.S. Army Corps of Engineers in which eight Dolly Varden collected from the estuary of the Suqi River were analyzed for PCBs (USATSDR, 2006). Given the high variability in PCB concentrations among fish in the Suqi River found in this study and in von Hippel et al. (2018), a sample size of eight individuals is insufficient for a reliable assessment. Furthermore, the Army Corps sampling occurred before the major remediation efforts at Northeast Cape. Remediation activities often release contaminants during the process, which can lead to an increase in contaminant concentration for some time after remediation (Scrudato et al., 2012; Voie et al., 2002). Dolly Varden used in the ATSDR health assessment were collected from the estuary (about 2.4 km downstream of the FUD site) instead of at input sites. Estuarine environments receive tidal influx that disperses and dilutes contaminants and are thus not comparable to upstream contaminated sites. To our knowledge, the ATSDR did not examine PCB profiles to differentiate potential sources of PCBs in Dolly Varden. Additionally, the study on which the ATSDR based their assessment did not differentiate between anadromous and resident freshwater Dolly Varden. Resident freshwater fish accumulate contaminants throughout their lifespan in the local habitat, whereas anadromous fish complete most growth in the ocean and are thus unlikely to reflect local freshwater pollution sources.

Collectively, our results contradict the findings reported in the ATSDR assessment. The ATSDR reported mean concentrations of Aroclor 1254 of 0.014 μg/g and Aroclor 1260 of 0.00096 μg/g, which is less than half of the mean total PCBs (0.029 μg/g) that we measured in Dolly Varden collected at input sites. PCB profiles in this study and in von Hippel et al. (2018) show that fish collected near the Northeast Cape FUD site have

significantly heavier PCBs than do fish from reference sites across three fish species. Because heavier PCB classes are less volatile and do not reach higher latitudes through global distillation, heavily chlorinated PCBs in Suqi River fish are likely a result of FUD site contamination. In summary, the ATSDR concluded that the FUD site does not pose a health risk to Sivuqaq residents (USATSDR, 2017). However, our results do not support this conclusion and highlight the need for robust health assessments based on relevant data to accurately assess risk. In particular, an assessment of human health consequences for consumption of Suqi River fish should include: 1) a large sample size of fish collected from sites spanning from the headwaters to the estuary across multiple years, 2) an assessment of life history for individual fish to differentiate between resident freshwater and anadromous, and 3) analysis of all pollutants known to be associated with the Northeast Cape FUD site and consideration of additive and/or synergistic effects.

### 4.5. Limitations

Small sample sizes limited our statistical power to detect significant differences. We found a significant negative correlation between $\delta^{15}N$ and total Hg when input sites were included; however, it was no longer significant when input sites were removed from the analysis, despite a stronger correlation coefficient. Similarly, the relationship between total PCB concentration and $\delta^{13}C$ was no longer significant when input sites were excluded from analysis. Future research should increase the sample size for more robust statistical analyses. However, we trapped only a small number of Dolly Varden despite considerable effort, which likely reflects a population that has not yet recovered from the effects of FUD site pollution.

As mobile animals, Dolly Varden may swim between input sites and either upstream or downstream sites, and we are not able to determine what fraction of their time they developed at different sites. Nevertheless, we found elevated Hg and PCB concentrations in Dolly Varden collected at input sites, suggesting that the signal of contamination originating at the FUD site is sufficiently strong to be apparent despite fish movement. Future research could overcome this limitation by caging fish in different locations such that all exposure would have occurred at that location.

We measured total Hg rather than MeHg in fish muscle tissue, and MeHg is the form of primary health concern. However, MeHg typically accounts for over 90% of measured total Hg in fish (Bloom, 1992), including in Dolly Varden (Tran et al., 2015, 2016). Additionally, the EPA recommends measuring total Hg as a proxy for MeHg when comparing fish Hg concentrations to their unlimited consumption screening levels (USEPA, 2000).

## 5. Conclusion

Dolly Varden are an important subsistence food source for Sivuqaq residents and fish caught in the Suqi River may pose health risks due to contaminants originating at the Northeast Cape FUD site. Northeast Cape was a vital community and gathering place for traditional foods prior to the construction of the military site during the Cold War. Residents of Sivuqaq want to safely re-establish the community at Northeast Cape, which necessitates long-term robust health assessments, as well as suitable remediation and monitoring of remaining contamination originating from the FUD site. Furthermore, additional remediation of the Suqi River would facilitate the re-establishment of healthy populations of other subsistence fish species, such as salmon, which will further enhance community rebuilding. A vigorous process of assessment, remediation, and monitoring should be accomplished under Tribal supervision and agreement to ensure sovereignty of the data and relevance of the process to Tribal priorities, such as decisions on siting of the restored community.

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## Credit authorship contribution statement

**Conceptualization**: FAvH, PM, DC, CLB, RJW
**Funding Acquisition**: FAvH, PM, DC, CLB, RJW
**Study Design**: FAvH, PM, DC, CLB, RJW
**Field Work**: FAvH, DD, JG, TI, JHP, CLB
**Laboratory Analysis**: RJW, ED, GZ, AS
**Statistical Analysis**: RJW, FAvH, SB
**Manuscript Writing**: RJW and FAvH, with contributions from all authors

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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