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Temporal trends, lake-to-lake variation, and climate effects on Arctic char (*Salvelinus alpinus*) mercury concentrations from six High Arctic lakes in Nunavut, Canada

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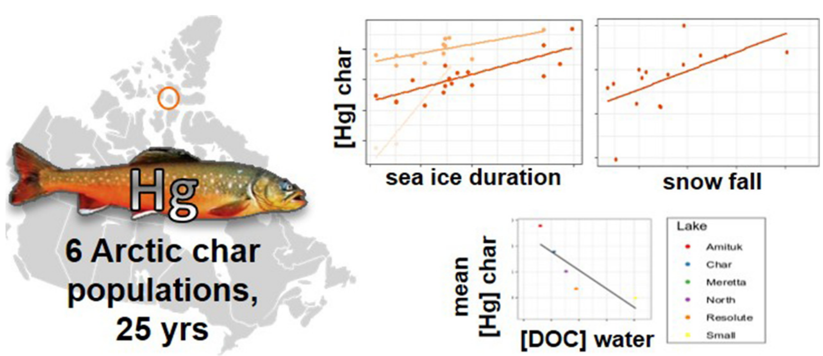
HIGHLIGHTS

- Mercury (Hg) and climate change are concurrently impacting Arctic ecosystems.
- We monitored Hg concentrations in landlocked Arctic char from 6 lakes (1989–2018).
- Organic carbon in water best explained differences between lakes.
- Sea ice and snow fall were strongly linked to Hg in char, but not for all lakes.
- Currently, lake-specific processes strongly temper climate effects on [Hg] in fish.

GRAPHICAL ABSTRACT



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ABSTRACT

Climate warming and mercury (Hg) are concurrently influencing Arctic ecosystems, altering their functioning and threatening food security. Non-anadromous Arctic char (*Salvelinus alpinus*) in small lakes were used to biomonitor these two anthropogenic stressors, because this iconic Arctic species is a long-lived top predator in relatively simple food webs, and yet population characteristics vary greatly, reflecting differences between lake systems. Mercury concentrations in six landlocked Arctic char populations on Cornwallis Island, Nunavut have been monitored as early as 1989, providing a novel dataset to examine differences in muscle [Hg] among char populations, temporal trends, and the relationship between climate patterns and Arctic char [Hg]. We found significant lake-to-lake differences in length-adjusted Arctic char muscle [Hg], which varied by up to 9-fold. Arctic char muscle [Hg] was significantly correlated to dissolved and particulate organic carbon concentrations in water; neither watershed area or vegetation cover explained differences. Three lakes exhibited significant temporal declines in length-adjusted [Hg] in Arctic char; the other three lakes had no significant trends. Though precipitation, temperature, wind speed, and sea ice duration were tested, no single climate variable was significantly

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correlated to length-adjusted [Hg] across populations. However, Arctic char Hg in Resolute Lake exhibited a significant correlation with sea ice duration, which is likely closely linked to lake ice duration, and which may impact Hg processing in lakes. Additionally, Arctic char [Hg] in Amituk Lake was significantly correlated to snow fall, which may be linked to Hg deposition. The lack of consistent temporal trends in neighboring char populations indicates that currently, within lake processes are the strongest drivers of [Hg] in char in the study lakes and potentially in other Arctic lakes, and that the influence of climate change will likely vary from lake to lake.

1. Introduction

The amount of mercury (Hg), a persistent and neurotoxic contaminant, circulating in the environment has increased 3–5 fold since the industrial revolution (Selin, 2009), concurrent with increases in global CO_2 emissions (Lamborg et al., 2002). Much of the Hg is released directly into the atmosphere (Streets et al., 2017), which, through natural circulation processes, acts as a conduit for contaminants to high latitude environments (Shindell et al., 2008). The reactivity of atmospheric Hg increases in high latitude environments due to low temperature halogen chemistry, promoting oxidation to water soluble and divalent species which readily deposit onto environmental surfaces (Angot et al., 2016). Consequently, the Arctic acts as a global sink for Hg, with an estimated 117 t deposited annually (Dastoor et al., 2015). This generates a disproportionately high risk of Hg exposure for Arctic indigenous people and wildlife (NCP, 2012; Dietz et al., 2009; AMAP, 2015), despite its great distance from the major Hg emission sources.

Once deposited, Hg toxicity is greatly increased by methylation, which occurs primarily in aquatic and semi-aquatic environments (Morel et al., 1998; Selin, 2009). Fish and top predators are especially prone to high levels of Hg due to bioaccumulation and biomagnification of methyl Hg (MeHg), and fish consumption is a major human exposure route (Selin, 2009). Mercury poses health risks to wildlife (Scheuhammer et al., 2015) and people (Ha et al., 2017), which led to the development of the *Minamata Convention on Mercury*, a voluntary global treaty to reduce Hg pollution (UNEP, 2013). The preamble to the Convention stresses the greater risk to Arctic Indigenous communities due to their reliance on wild foods.

At the same time, global climate change is also disproportionately impacting the Arctic. As climate change progresses, there is a need to better understand the dynamics of Hg in sensitive biota, as well as how climate change will affect the role of Arctic ecosystems as Hg sinks (Alava et al., 2017). For example, in two high Arctic sites, increased Hg deposition from the atmosphere occurred at higher temperatures (Cole and Steffen, 2010) and warming and decreased ice extent could promote the production of MeHg (AMAP, 2011). Field measurements of Hg in Arctic air exhibit recent declines (Steffen et al., 2015); with declines of 0.9%/year from 2000 to 2009 of gaseous elemental Hg in air at Alert (Cole et al., 2013). However, due to changes in snow pack and sea ice characteristics, the Global/Regional Atmospheric Heavy Metals model predicts that Hg deposition in the Arctic is increasing, and that the Arctic remains a net sink for Hg (Dastoor et al., 2015). More recently, the projected 30–99% decline in permafrost due to thawing was predicted to release a portion of the estimated 793 $\pm$ 461 (mean $\pm$ SD) gigagrams of Hg stored in northern hemisphere permafrost over the next century (Schuster et al., 2018).

Methyl Hg concentrations in Arctic animals vary greatly across species, time, and location (AMAP, 2011). The concentration of MeHg in fish in any given aquatic system is influenced by Hg inputs to the body of water and its watershed, water chemistry, methylation rates, and food web structure within the lake (Rypel, 2010). Across North America, trends in predator fish Hg concentrations over time vary by region (Zhou et al., 2017; Eagles-Smith et al., 2016; Gandhi et al., 2014; Gantner et al., 2009; Rennie et al., 2005) but do not necessarily reflect Hg deposition in many cases due to variation in Hg post-depositional processing. In the Arctic, post-depositional processing strongly influences population-level trends in biota [Hg]; although Hg deposition across the landscape is assumed to be similar (due to atmospheric transport), there are large differences in biota [Hg] at the population level (also see Pelletier et al., 2017).

Non-migratory, or landlocked Arctic char (*Salvelinus alpinus*) are excellent biomonitors for freshwater ecosystems in the Arctic, because they are long-lived aquatic top predators, remain in a single ecosystem, are sensitive to environmental changes, and are a minor but dependable food source for some Arctic communities (Power et al., 2012; Barst et al., 2018). Mercury cycling is sensitive to meteorological drivers (Dastoor et al., 2015) which, in the Arctic, are undergoing rapid shifts due to climate change (Hinzman et al., 2005; Polyak et al., 2010). For example, warming may increase Hg methylation rate (Hammerschmidt and Fitzgerald, 2004, 2006; Lehnherr et al., 2012). Increased temperature has been shown to increase MeHg bioaccumulation in coastal marine killifish (*Fundulus heteroclitus*) attributed to increased metabolic rate (Dijkstra et al., 2013). Evans et al. (2013) reported higher [Hg] in lake trout (*Salvelinus namaycush*), northern pike (*Esox lucius*), and burbot (*Lota lota*) associated with cooler temperatures in Great Slave Lake (though they find no evidence of a direct effect of temperature), while Carrie et al. (2010) report increasing [Hg] in burbot in the neighboring Mackenzie River which the authors linked to warming-induced ice loss and increased primary productivity. In 90 large natural lakes of Québec, warmer temperatures were associated with higher growth rates resulting in lower [Hg] in walleye (*Sander vitreus*), while the warm temperatures were associated with increased [Hg] in northern pike from the same lakes (Lucotte et al., 2016). A study of landlocked Arctic char in a small lake in Greenland linked increasing [Hg] with increased temperatures, which was attributed to increases in primary production, similar to the logic of Carrie et al. (2010). In small southern Ontario lakes, a negative relationship was observed between temperature and [Hg] in largemouth and smallmouth bass (*Micropterus salmoides* and *M. dolomieu*, respectively) which was attributed to growth dilution (Chen et al., 2018). Ward et al. (2010) did not detect a temperature-dependent bioaccumulation effect in Atlantic salmon (*Salmo salar*), but rather, found that [Hg] in salmon was dependent on growth rate and prey [Hg], with fast-growing fish exhibiting growth dilution, and slow growing fish having higher [Hg]. The effect of temperature on Hg bioaccumulation in fish can therefore be said to be species- and habitat-specific, and to be modulated by atmospheric and biogeochemical processes prior to the biological transformations (methylation) preceding bioaccumulation in fish. Though Hg bioaccumulation is modulated by habitat-specific effects, biomagnification of Hg through aquatic food webs is a generally consistent process, exhibiting similar rates when diverse food webs are compared, but tending to be higher in oligotrophic, cold systems (Lavoie et al., 2013). As hypothesized by several of the researchers cited in the Hg bioaccumulation literature above, differences in aquatic predator [Hg] between populations is largely due to processes occurring at the base of the food web, which may or may not also influence fish growth rate (also see Pućko et al., 2014). Studies of Hg concentration in Arctic animals over time indicate that trends vary widely between and within taxa, with the majority reporting non-significant temporal trends (Rigét et al., 2011). The lack of consistency in trends underscores the importance of continued research of contaminants in Arctic animals, as it is thus far difficult to forecast changes in Hg dynamics and concentrations in organisms. In some locations of the Canadian Arctic, Hg concentrations are increasing in anadromous Arctic char, while other populations are declining or exhibiting no change (Evans et al., 2015; Brown et al., 2018). Likewise, the Hg trends in ringed seals (*Pusa hispida*) in the Eastern Canadian Arctic generally increased from the early 1970s to 2009 (Brown et al., 2018), but in polar bears (*Ursus maritimus*) current levels are not different from those in the 1980s (Brown et al., 2018). In five species of seabird eggs from the Canadian Arctic, Hg concentrations were increasing in the 1970s and 1980s before plateauing and then exhibiting declines from 1993 to 2013 (Braune et al., 2016). For example, a study of museum specimens of ivory gulls (*Pagophila eburnea*) reported a dramatic 45× increase in MeHg concentrations from 1877 to 2007 without any indication of a dietary shift (Bond et al., 2015).

Currently, our understanding of temporal trends and climate effects on [Hg] in animals is confounded by the complex biogeochemistry of Hg and differences in reported trends across and within geographic areas and over time. This study seeks to gain insight on how population-level (i.e., lake-to-lake) differences influence the current understanding of predator [Hg] temporal trends and the influence of climate by focusing on multiple populations of a single species in neighboring small lakes with similar food webs and a single species of fish. The objectives of this study were to describe temporal trends and lake-to-lake variation of [Hg] in landlocked Arctic char and determine if climate influenced the temporal trends. We analyzed [Hg] in char over a period of up to 29 years from six lakes on Cornwallis Island, Nunavut, Canada.

2. Materials and methods

2.1. Site description

Char, Resolute, North, Meretta, Small, and Amituk Lakes are located on Cornwallis Island (75.105° N, −94.828° W), Nunavut, Canada (Fig. 1), home to the second most northerly community in the world, Resolute Bay (Qausuittuq, ᖃᐅᓱᐃᑦᑐᖅ). The island lies within the polar desert ecozone of the high Arctic, where lakes make up <2% of the landscape (Walker et al., 2005). The lakes of Cornwallis are classified as ultra-oligotrophic, characterized by a short growing season (2––2.5 months of open water, occasionally retaining some ice cover throughout), limited nutrient availability (Schindler et al., 1974a), benthic based food webs (Welch and Kalff, 1974), low productivity (Rigler, 1978) and correspondingly low sedimentation rates (Whalen and Cornwell, 1985).

Arctic char are the only fish species in the study lakes. The Arctic char are landlocked due to the shallow depth of lake outflows resulting from isostatic rebound (Power et al., 2012). North and Small Lakes have adjoining watersheds, as do Resolute, Char, and Meretta Lakes. These five lakes range from 1 to 12 km from Resolute Bay on the southwestern coast of Cornwallis Island, while Amituk Lake is on the eastern edge of the island 48 km north of Resolute Bay. Ponds in the catchment of Meretta Lake received waste water draining from the Canadian Department of Transport Airport Base, the 'North Base', via a utilidor and a 1.6 km stream system from 1949 to 1998 (Schindler et al., 1974b; Douglas and Smol, 2000). The lake responded primarily by increasing benthic planktonic diversity and biomass, contributing to anoxia in the hypolimnion, with a very slight increase in phytoplankton production, and in recent years shows signs of recovery (Antoniades et al., 2011).

2.2. Arctic char sampling and Hg analysis

Arctic char collections began in 1989 and have been described in detail previously (Gantner et al., 2010a, 2010b; Lescord et al., 2015a; Muir et al., 2005). Briefly, annual char collections were carried out in late July or early August using gill nets (mesh size 36 and 42 mm). Conditions permitting, nets were set in the same area and orientation each year. Total and fork lengths (cm) and weight (g) were measured for each fish and a skinless dorsal muscle sample was collected and kept frozen prior to total Hg analysis. Fulton's condition factor (K) was calculated

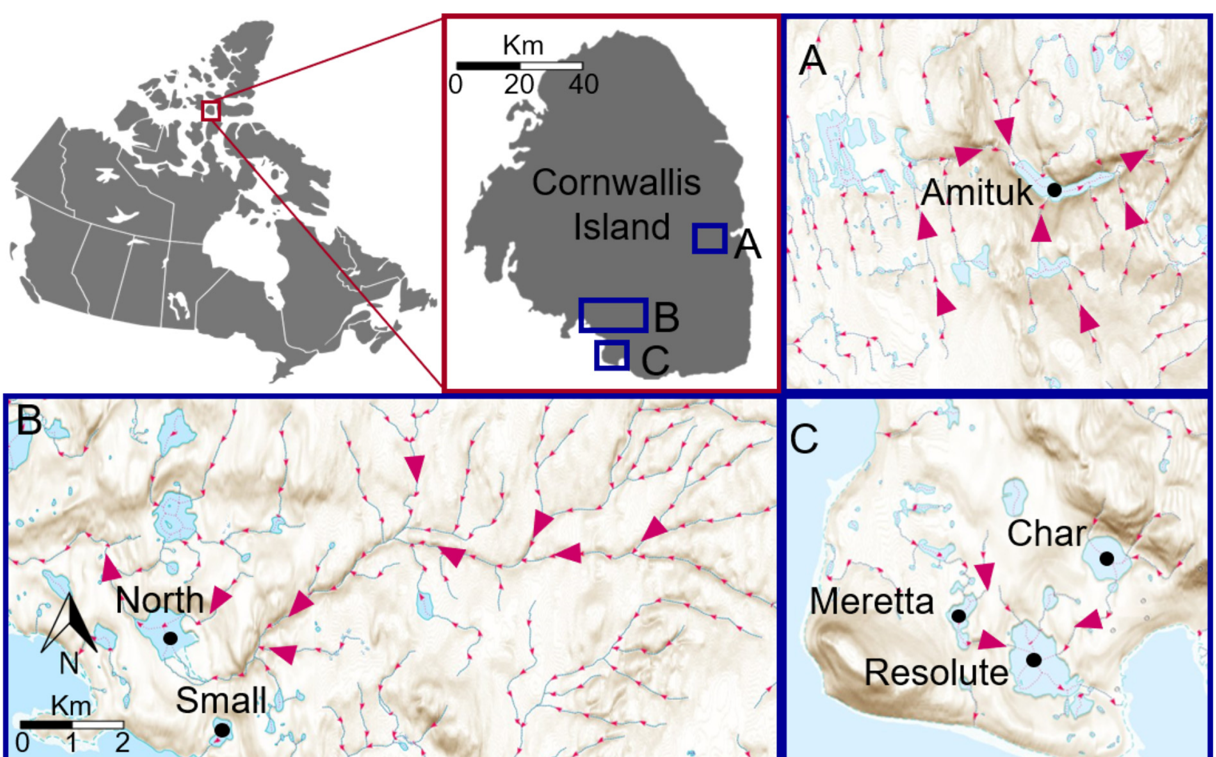

Fig. 1. Study location in six lakes on Cornwallis Island, NU, Canada where Hg in Arctic char muscle was monitored over time. Pink arrows indicate water flow direction. Scale bar on inset B also applies to insets A and C. Inset maps retrieved from GeoGratis Toporama (Natural Resources Canada, 2019).

as ($100 \times$ wt···fork length^{-3}) (Froese, 2006). Muscle samples were analyzed for total Hg by cold vapor atomic absorption spectrometry or by USEPA method 7473 (USEPA, 2007). DORM-3 (fish muscle protein) and DOLT-4 (dogfish liver) certified reference materials (National Research Council of Canada) were analyzed with samples to assess accuracy and precision. Recovery of the certified reference materials were 99.8% for DORM-3 ($n = 42$) and 97.5% for DOLT-4 ($n = 41$) for samples analyzed from 2011 to 2018. For samples analyzed prior to 2012, measurement accuracy was previously reported and achieved acceptable levels (see Gantner et al., 2010a; Lescord et al., 2015a). Blanks had non-detectable concentrations or levels <5% of measured values, and therefore no blank correction was used. The analytical laboratories (National Laboratory for Environmental Testing (NLET) and the Muir analytical group, Canada Centre for Inland Waters, Environment and Climate Change Canada, Burlington, ON) have successfully participated in the annual Northern Contaminants Program Quality Assurance program (NCP, 2012). The annual means for these measurements are provided in Tables SI-1–6.

2.3. Statistical analyses

Alpha was set at 0.05 for statistical analyses, which were performed with R version 3.5.1 software (Copyright 2018, The R Foundation for Statistical Computing).

2.3.1. Fish population characteristics

Fish total length (cm), weight (g), and age (years) data for the six char populations were normally distributed and their means were compared by analysis of variance (ANOVA) and Tukey's post-hoc tests. Linear regression was then used to determine if these characteristics changed over time within each population.

2.3.2. Arctic char Hg concentrations–temporal trends and lake-to-lake variation

Arctic char muscle total [Hg] were log transformed to achieve normality for each lake for each year, based on Shapiro-Wilk's tests. Because fish size varied significantly within lakes between years, and across lakes, a single length-adjusted annual log [Hg] was generated. This was achieved using a regression between log [Hg] and fish total length at the mean total length for all the fish (40.1 cm, Fig. SI-1) for each year and lake, a method of length adjustment used in previous studies (Evans et al., 2005; Miller et al., 2013). Our a priori requirements for an adjustment variable were that the variable exhibit a consistent relationship to log [Hg] among populations, and that the same adjustment method be applied to all lakes and all years. For comparison, log [Hg] by age relationship for each lake is presented in Fig. SI-2 but was not found to be better than length. Another approach to size adjustment is to use an analysis of covariance approach (AMAP, 2011, Section 5.3.3.4), however, this approach was not feasible for this data set due to the significant change over time exhibited by the biological variables (Table SI-7). Length was selected as the adjustment variable over other characteristics, such as age, $\delta^{15}N$, and weight, because for these populations log [Hg] it exhibited the most consistent relationship (both among and within-lakes). Though there are other significant differences in the other biological characteristics between populations (Table 1), once the adjustment was made for fish length, the significance of other biological variables for predicting log [Hg] greatly decreased, indicating that much of the variability in log [Hg] both within and among lakes was explained by fish length. The annual length-adjusted log [Hg] was used for the temporal and climate analyses; while the mean of the annual length-adjusted log [Hg] was used for the lake-to-lake comparisons (Table SI-8).

To assess the temporal trends in [Hg] in the Arctic char, linear regressions were run for each lake with length-adjusted log [Hg] versus year. There was an eight-year gap between the earliest samples and more recent samples collected from Amituk Lake, which could lead to errors in interpretation of the temporal trend. Therefore, separate regressions were run with all the data (1989–2018, 18 years) and with only recent data (2001–2018, 16 sampling years).

To compare [Hg] in Arctic char between lakes, we first used length-adjusted log transformed [Hg] and year as the experimental unit to compare lakes by an ANOVA and Tukey's post-hoc test. Because there were significant temporal trends in [Hg] in three of the lakes according to the linear regressions described above, the differences detected by this approach may not be persistent over time. To assess the current state of Hg in Arctic char in these lakes, we used Hg data from all fish collected in 2017 and 2018. The [Hg] were log transformed, and we used an ANCOVA with fish length as a co-variate to remove the effect of size, and Tukey's post-hoc tests to compare between lakes. The results of the ANCOVA confirmed that the lake-to-lake differences from the initial ANOVA approach were persistent for the 2016–2018 Arctic char.

2.3.3. Influence of watershed, lake and climate characteristics on Hg temporal trends

The three types of variables known to influence [Hg] in fish (geographic i.e., watershed; lake, i.e., depth and water chemistry; and climate) were tested against the length-adjusted [Hg] of Arctic char using Spearman correlations. The watershed and water chemistry analyses used Spearman's rank comparisons because the mean length-adjusted [Hg] was compared to single means for these measures, making lake the experimental unit. For the climate analysis, multiple measurements per lake and per climate variable over time were tested,

Table 1
Selected descriptors (mean ± 1 SD, unless otherwise stated) of landlocked Arctic char from the six study lakes on Cornwallis Island, Nunavut, Canada. Letters indicate significant differences in the annual means among lakes for each descriptor (ANOVA with Tukey's test, $\alpha = 0.05$). See Tables SI 1–6 for annual means. See Table 2 for length-adjusted means over time. Lower panel, selected descriptors of each lake.

	Amituk	Char	Meretta	North	Resolute	Small
No. of years	18	13	8	14	22	14
Catch/year	10 ± 6	7 ± 2	14 ± 4	13 ± 5	14 ± 4	16 ± 6
No. of fish	184	85	111	181	299	220
Total length (cm)	44.3 ± 5^{a}	39.4 ± 5.2^{ab}	43 ± 5^{ab}	39.3 ± 5^{ab}	40.7 ± 5^{ab}	37.1 ± 5^{b}
Weight (g)	701 ± 273^{a}	756 ± 284^{a}	721 ± 273^{a}	494 ± 273^{ab}	537 ± 286^{ab}	357 ± 273^{b}
Fulton's condition factor	0.89 ± 0.12^{ab}	0.94 ± 0.15^{a}	1.01 ± 0.09^{a}	0.91 ± 0.06^{a}	0.94 ± 0.07^{a}	0.81 ± 0.07^{b}
Age (y)	18 ± 3^{ab}	17 ± 3^{bcd}	7 ± 3^{d}	16 ± 2^{bc}	19 ± 2^{a}	14 ± 2^{c}
$\delta^{13}C$ (‰)	-22.3 ± 0.9^{a}	-23.8 ± 0.9^{b}	-22 ± 0.8^{a}	-23.7 ± 0.8^{b}	-22.3 ± 0.8^{a}	-23.4 ± 0.8^{b}
$\delta^{15}N$ (‰)	11.9 ± 0.9^{a}	11.4 ± 0.9^{a}	11.7 ± 0.9^{a}	11.7 ± 0.9^{a}	11.7 ± 0.9^{a}	9.4 ± 0.9^{b}
Hg (μg/g, wet wt.), all years	1.28 ± 0.23^{a}	0.43 ± 0.23^{b}	0.14 ± 0.23^{bc}	0.28 ± 0.23^{bc}	0.18 ± 0.23^{c}	0.12 ± 0.23^{c}
Hg (μg/g, wet wt.), 2016 and - 2018 only	1.09 ± 0.11^{a}	$0.71–0.66^{b}$	0.08 ± 0.11^{c}	0.33 ± 0.11^{c}	0.17 ± 0.11^{c}	0.11 ± 0.11^{c}
Hg (μg/g wet wt.) for a 40.1 cm char	0.90 ± 0.28^{a}	0.47 ± 0.13^{b}	0.12 ± 0.07^{e}	0.26 ± 0.05^{c}	0.17 ± 0.06^{d}	$0.13 \pm 0.03^{d,e}$
Lake surface area (km^2)	0.378	0.526	0.262	0.957	1.27	0.140
le	43	27	10	14	22.5	8.2
Watershed area (km^2)	31.1	3.32	6.15	97.0	11.6	0.878
Watershed:lake area ratio	0.012	0.16	0.043	0.010	0.109	0.159

making year the experimental unit. Because multiple comparisons were made, *p* values were corrected to prevent test-wise type 1 error inflation using the false discovery rate method (Benjamini and Hochberg, 1995). Details on the variable used for each Spearman correlation analysis are provided below.

2.3.3.1. Watersheds. Fourteen watershed variables, several of which have been previously shown to influence Hg in lakes or in fish, were tested against Hg concentrations in Arctic char. A land cover shapefile derived from Landsat 5 and 7 ortho-images (4) describing vegetation cover types (Land Cover Circa 2000), the Canadian Digital Elevation Model raster and National Hydro Network polyline and polygon shapefiles were all used to generate maps of each of the lake watersheds; all data were retrieved from the NRCan OpenData portal (http://geogratis.gc.ca). Geographic analyses were performed in ArcMap 10.3.1 (Esri, Redlands, CA, USA). Watershed delineation using the elevation and stream network data was carried out for each of the study lakes by using a digital elevation model to locate pour points and flow direction of streams within the watersheds using the Hydrology toolbox in the Spatial Analyst extension. The land cover polygon shapefile describing vegetation cover types was clipped to each delineated watershed in order to calculate the area of each vegetation type in each watershed (Figs. SI-3–5, Table SI-9). The surface area of each lake and the maximum depth (z_{max}) were also included.

2.3.3.2. Water chemistry. Six water chemistry variables were tested against [Hg] in Arctic char. Water sampling for this dataset has been described elsewhere (Cabrerizo et al., 2018; Lescord et al., 2015a). Briefly, water samples were collected annually concurrent with fish sampling (but less frequently), from mid-lake or as near to mid-lake as ice cover would permit. When ice cover prevented boat access to the deepest area of the lake, only surface samples were collected. Using a pre-cleaned 2 L Niskin sampler, water was collected from 0.5–1 m in depth, and from 1 m above the deepest depth for each lake (see Kirk and St. Louis, 2009). Samples were stored in pre-cleaned brown glass bottles at 4 °C, and filtered using GF/C (47 mm, pore size 1.2 μm for chlorophyll *a* (Chl *a*) and particulate organic carbon (POC); 25 mm, pore size 0.7 μm for dissolved organic carbon (DOC)) within 24 h. The filters were wrapped in aluminum foil and frozen until analysis. Unfiltered subsamples for analysis of total dissolved nitrogen (TDN), nitrite/nitrate (NO_3/NO_2), total dissolved phosphorus (TDP), were stored at 4 °C until analysis. Water samples and filters were analyzed at NLET within 28 days of collection. Chlorophyll *a* was extracted with acetone and measured using spectrophotometry at wavelengths 663, 645, and 630 nm (NLET method 01-1100, Environment Canada, 2010). Means for this dataset are presented in Table SI-10.

2.3.3.3. Weather and climate data. Climate variables were tested against [Hg] in Arctic char. Daily and hourly weather data (temperature (°C), snow fall (cm), rain fall (mm), accumulated snow (cm), and wind speed (km/h) were collected by Environment and Climate Change Canada weather stations near Resolute Bay. The Resolute CARS weather station provided data from 1988 to 2013 and the Resolute CS weather station provided data from 2013 to 2015. Differences in temperature and snow depth between weather stations were not significant for overlapping years (*t*-test), so no distinction was made when combining station data. Sea ice thickness (cm) and duration (days) provided by the Canadian Ice Service was also included because sea ice is both sensitive to climate and weather and plays a large role in determination of climate and weather through feedback mechanisms (Curry et al., 1995). Sea ice measurements were made through the ice in Resolute Bay about 1 km from Resolute Lake. Repeating patterns in sea surface temperature or height (which is related to pressure and temperature of the water mass) that influence global weather patterns are known as climate oscillations. Weather in the Canadian Arctic is influenced by several of these oscillations (AMAP, 2011), and trends in oscillation values have been associated with contaminant transfer to the Arctic (Eckhardt et al., 2003; Octaviani et al., 2015). The indices of the Pacific North American Pattern (PNAP), the Arctic Oscillation (AO), the Pacific Decadal Oscillation (PDO) and the North Atlantic Oscillation (NAO) provided by the National Weather Service Climate Prediction Centre of the National Oceanic and Atmospheric Administration) were included in the analysis. Air temperatures, climate oscillation indices, and ice data were compiled into annual measures. For temperature, wind speed, oscillations, and sea ice thickness, annual means were calculated. For the precipitation measures (rain fall, snow fall, and snow accumulation depth) annual totals were calculated. When >10% of any measurements were missing for the year, the annual summaries were not calculated. For this analysis, both the corrected *p* values and the uncorrected p values are presented in the results because, while we acknowledge the possibility that some of the significant correlations may be due to type I error, the cost of type I error in an exploratory statistical study is sufficiently low to balance the risk of type II error in favor of detecting a biologically significant result (see Cabin and Mitchell, 2000). For additional interpretation of these tests we provide plots of each significant relationship in Fig. SI-6.

2.3.4. Modeling length-adjusted Hg concentration using climate

After the analyses of temporal trends in [Hg], lakes were divided by trend type (i.e., decreasing (3 lakes) or no change (3 lakes)). The length-adjusted log [Hg] in Arctic char of the three declining lakes (46 total sampling events) were subjected to a mixed model and stepwise selection procedure: a mixed model was constructed with all of the 11 annual climate variables and included lake as a random variable using the lme4 package (Bates et al., 2015) before applying a stepwise backward selection process on the fixed effects, using the cut-off for significance (α) of 0.05 and estimating degrees of freedom using the Kenward-Roger estimation method available in the MuMin package (Bartoń, 2018). This procedure selects variable sets which do not exhibit correlation among variables. We performed the same analysis for the length-adjusted log [Hg] in Arctic char from the three lakes which did not exhibit a linear trend ($n = 40$ observations). Then, to better understand the importance of each lake's record in the selection process and the relationship of the selected climate variables on each of the study populations [Hg], this model was applied to each of the lakes separately.

3. Results

3.1. Arctic char samples

Arctic char sampling occurred most often in Resolute Lake (22 sample years and 299 fish) and least in Meretta Lake (8 years of sampling and 111 fish) (Table 1). The goal of each sampling was to catch 10 or more adult (>200 g) fish, which was achieved in all but Char Lake, where yields were usually below 10 (see Tables SI 1–6 for annual means of the char population characteristics).

3.2. Change over time in population characteristics

The regressions of the annual means of the char population characteristics revealed that, for at least one population, each of the annual mean population characteristics changed over time (Table SI-7). In North, and Resolute Lakes, annual mean fish total length significantly increased with time, while in Small Lake, length significantly decreased. Similarly, the Resolute char increased in mean weight over time while weights in Small Lake decreased. For the Amituk, Meretta, and Small Lake char populations, annual mean K significantly declined over time.

3.3. Size – Hg relationships of Arctic char populations

The majority (51 of 87) of the annual log Hg-fish length regressions had significant *p*-values (Fig. SI-1); in general, only years which had very low or negative slopes did not have significant relationships. Regardless of significance, all regressions were retained for subsequent analyses. The mean slope of the log Hg and length regressions for each lake (mean ± standard deviation) ranged from 0.016 ± 0.018 in Char Lake to 0.039 ± 0.012 in Small Lake.

3.4. Lake-to-lake variation in Arctic char Hg

Using length-adjusted of [Hg] over the entire collection period to compare lakes, concentrations in the Small Lake char were the lowest but not significantly different than Meretta and Resolute Lakes (Table 1). Amituk Lake char had significantly higher [Hg] than the other lakes; the next highest lake-mean [Hg] were those in Char Lake which were nearly three times lower (Table 1). When the length-adjusted mean [Hg] were compared (Tables 1 and SI-8), the grouping assignments were similar to the groupings for the non-length-adjusted.

[Hg], with Amituk, Char, and North populations significantly higher than the others.

3.5. Temporal trends in Arctic char Hg by lake

Based on linear regressions, length-adjusted log [Hg] in char did not significantly change in North, Small, or Char Lakes ($p = .73$, 0.58, and 0.07, respectively), and significantly decreased in Resolute (slope = −0.005, $R^2 = 0.33$, $p = 4.9\text{e-}3$) and Meretta (slope = −0.015, $R^2 = 0.88$, $p = 6.0\text{e-}4$), over time (Fig. 2). For the Amituk Lake char, length-adjusted log [Hg] decreased over the period 2001 to 2018 (slope = −0.016, $R^2 = 0.35$, $N = 14$, $p = .016$), but was not significant (slope = −0.005, $R^2 = 0.081$, $N = 16$, $p = .252$) when the two early sampling years were included. When the slope values are back-transformed from the log, the change over time for the significantly decreasing lakes are −1.94% for Amituk, −3.5% for Meretta, and −1.1% ng/g wet wt. for Resolute.

3.6. Influence of water chemistry, lake and climate characteristics on Hg trends

None of the watershed characteristics were significantly correlated to the lake-to-lake variation in length-adjusted log [Hg] in Arctic char (Table 2). For water chemistry data, DOC in both surface and profundal waters, and POC in profundal waters were significantly correlated with Arctic char [Hg] (adj. $p = .019$ for each measure, Table 2, Fig. SI-6), although the lakes are well mixed and so differences in water chemistry over depth may be biologically insignificant. Among the climate variables, sea ice duration in nearby Resolute Bay was significantly positively correlated to the [Hg] over time for Resolute Lake (adj. $p = .041$, Table 3, Fig. 3) and snow fall was significantly positively correlated to Amituk Lake char [Hg] (adj. $p = .024$).

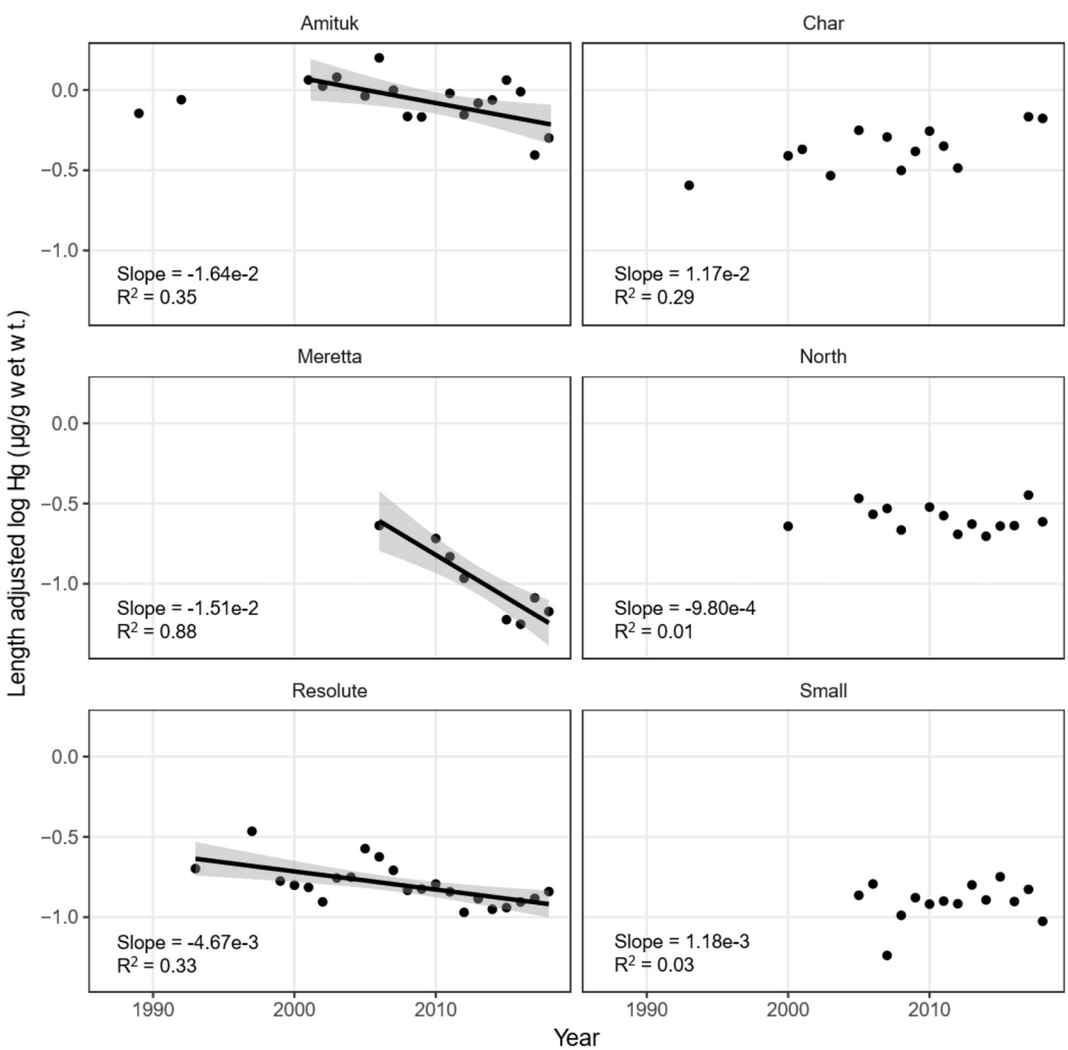

Fig. 2. Linear trends over time in the length-adjusted log Hg concentrations in Arctic char over time for each lake from Cornwallis Island, NU, Canada. Shading represents 95% confidence level for regression line. For Amituk Lake, the regression line presented is for 2001–2018; when earlier points are included, regression is not significant.

Table 2
Spearman's rho rank correlations for length-adjusted Hg concentrations in Arctic char and each watershed, lake, or water chemistry feature for the 6 lakes from Cornwallis Island, Nunavut. Ties in rank (due to measures of 0 area for the given cover type in multiple watersheds) are indicated in the last column. P values are corrected for multiple comparisons using the Benjamini-Hochberg false discovery rate procedure (1995); significant *P* values are indicated with *. See Tables SI-9 for watershed feature data and Table SI-10 for water chemistry data.

Feature	Rho	P	Ties
% Water	−0.314	0.957	
% Barren	−0.086	0.957	
% Till/colluvium	−0.086	0.957	
% Gramminoid shrub tundra	−0.030	0.957	Tie
% Snow/ice	0.638	0.749	Tie
% Bare soil with cryptogram crust	0.029	0.957	
% Dwarf shrub	0.131	0.957	Tie
% Tussock tundra	0.034	0.957	Tie
% Wet sedge	−0.270	0.957	Tie
% Wetland	0.655	0.749	Tie
% No data	0.655	0.749	Tie
Lake surface area, km^2	0.371	0.957	
Max. depth Z_{max}, m	0.886	0.339	
Watershed area, km^2	0.429	0.957	
Surface water [Chl *a*]	−0.600	0.499	–
Surface water [DOC]	−0.886	0.055	–
Surface water [POC]	−0.943	0.019*	–
Surface water [NO_2]	0.314	0.816	–
Surface water [SRP]	−0.087	0.873	–
Surface water [NO_3/NO_2]	−0.087	0.873	–
Profundal water [Chl *a*]	0.371	0.803	–
Profundal water [DOC]	−0.943	0.019*	–
Profundal water [POC]	−0.943	0.019*	–
Profundal water [NO_2]	−0.100	0.873	–
Profundal water [SRP]	−0.371	0.803	–
Profundal water [NO_3/NO_2]	−0.200	0.873	–

3.7. Models of length-adjusted log Hg over time vs. climate

The stepwise selection procedure generated the following model equation for the declining [Hg]lakes:

Length-adjusted log [Hg] = (Intercept) + 0.170 Snow fall – 0.242 Rain fall - 0.553 NAO, for which, the marginal (fixed effect) R^2 was 0.035 and the conditional (fixed and random effects) R^2 was 0.942. As seen in Table 4, the 2.5% confidence intervals for the variables did not overlap 0, therefore the effect is significant, which is confirmed by the *p* values for the model variables (Table 4). The very small marginal R^2 (especially in comparison to the larger conditional R^2) indicates the poor fit of the model when all lakes are included, and the small contribution of the fixed climate factors to overall fit relative to the contribution of "population". For the lakes which did not exhibit significant temporal trends for Hg, the selection process yielded an intercept-only model as no climate variables were significant predictors of length-adjusted log Hg in char for these lakes (data not shown). When the three selected variables were used to model each lake separately, the model exhibited a significant fit with Resolute Lake, but not with any of the other lakes (Table SI-11); significance of each of the climate variables for each of the lakes are presented in Table SI-12. The p values indicate that the selected climate variables generally fit better with the declining Hg populations, but that Resolute and Amituk Lakes appear to have driven the selection process for these variables (Table SI-12). The fit of the model is very poor for the no-trend populations.

Table 3
Spearman correlation coefficients (rho) of climate variables and oscillations with the length-adjusted log Hg concentration in Arctic char over time for each of the study lakes. Significant correlation coefficients after application of the Benjamini-Hochberg false discovery rate p value correction are indicated with *, those that were significant before *p* value correction are italicized. See Fig. SI-5 for plots of significant relationships. AO = Arctic Oscillation, PNAP = Pacific North American Pattern, NAO = North Atlantic Oscillation, and PDO = Pacific Decadal Oscillation.

Climate parameter	Amituk	Char	Meretta	North	Resolute	Small
Snow fall	*0.718**	−0.573	0	−0.082	0.397	0.198
Wind speed	−0.088	0.145	*−0.786*	−0.064	*−0.478*	0.336
Rain fall	−0.382	−0.027	−0.286	−0.088	*−0.480*	−0.176
Snow accum. depth	−0.038	0.091	−0.200	0.236	0.212	−0.227
Temp.	−0.121	0.245	*0.821*	0.368	−0.290	−0.011
Sea ice thickness	−0.027	−0.300	−0.429	−0.294	−0.358	−0.063
Sea ice duration	−0.200	0.075	*0.829*	*0.591*	*0.727**	−0.236
AO	0.218	−0.064	−0.286	0.033	−0.071	0.187
PNAP	0.075	0.600	−0.321	0.434	0.309	−0.302
NAO	0.175	0.227	−0.500	0.055	−0.090	0.495
PDO	0.343	0.390	−0.643	0.181	0.103	0.462

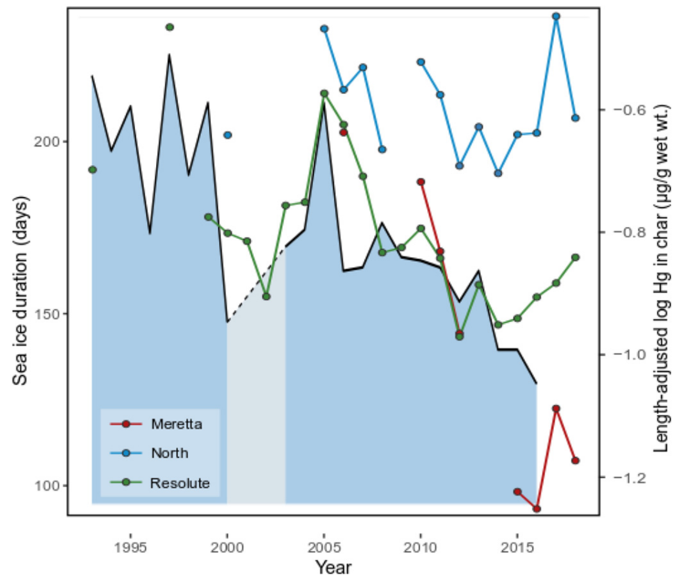

Fig. 3. Temporal relationship between sea ice duration in Resolute Bay (left axis) and length-adjusted Hg concentration in Arctic char from Meretta, North, and Resolute Lakes (right axis) on Cornwallis Island, NU, Canada. Where sea ice duration data was missing, a dotted line was drawn in to complete the shape.

4. Discussion

Temporal trends in Arctic char muscle length-adjusted [Hg] in six small high Arctic lakes either decreased or did not significantly change. Mercury concentrations in char differed widely between the lakes despite their geographic proximity and similar food webs, and the relative differences have persisted throughout the 25 years of sampling (1993–2018). The best explanatory variables for lake-to-lake differences were POC and DOC concentrations in profundal waters near the site of methylation in the sediments, but watershed variables were not significant. Although there was no single climate variable that was related to char [Hg] across all populations, sea ice duration was significantly positively related to [Hg] in three populations (two populations were not significant after p value correction), two of which are likely also impacted by recovery from historical waste water pollution. The mixed model analysis generated a similar result: NAO and rain fall were significant predictors in Resolute but not strong predictors in

Table 4
Variables selected by the backwards stepwise selection procedure on the three Arctic char populations which exhibited significant declining temporal trends in length-adjusted log [Hg]. Coefficients for the model equations and their associated confidence intervals, degrees of freedom (DF), t values and p values are presented.

Variable	Coefficient ± SE	2.5% CI	97.5% CI	DF	t-Value	p-Value
Intercept	0.053 ± 0.82	−1.83	1.93	2	0.064	0.950
Snow fall	0.17 ± 0.074	0.026	0.31	36	2.29	0.028
Rain fall	−0.24 ± 0.069	−0.374	−0.11	36	−3.53	0.0012
NAO index	−0.55 ± 0.16	−0.86	−0.24	36	−3.46	0.0014

other lakes, while snow fall was significant for Amituk Lake in the correlation and modeling analyses. The Resolute Lake population with the most complete sampling history ($n = 22$), exhibited significant results in both the correlation and the mixed model analyses, highlighting the importance of high-resolution biological data for monitoring and modeling studies. However, spurious results are also possible as more data is acquired. Our results confirm that [Hg] in Arctic char can vary widely between lakes, even over a relatively small geographic area that likely receives similar atmospheric deposition of Hg, and the influence of a changing Arctic climate has not created clear trends but appears to manifest in a system-specific manner. Therefore, it is recommended that larger-scale temporal trend and climate analyses consider populations as distinct experimental units within a geographic area.

4.1. Mercury in Arctic char: comparison between lakes and influence of water chemistry and watersheds

Arctic char muscle [Hg] increased with body length, which is consistent with a recent study documenting a positive relationship between length and [Hg] for 59 populations of non-anadromous Arctic char (Barst et al., 2018). The relative ranking by lake of Hg concentrations did not vary when concentrations were corrected for length, which reflects the effort of this monitoring program to collect similar size fish from all lakes, reducing the impact of size differences from year to year.

Mercury concentrations in the Arctic char varied widely between the six lakes sampled in this study, despite their geographical proximity and the similarity of their predominantly benthic food webs (Lescord et al., 2015a, 2015b; Gantner et al., 2010a). These concentrations were in range of other reports for landlocked Arctic char from the eastern Canadian Arctic (Gantner et al., 2009; Van der Velden et al., 2015; Barst et al., 2018), although [Hg] in Amituk Lake char exceeded most other reports. These concentrations essentially mirrored those reported in Gantner et al. (2010b) for the same populations from 1999 to 2007, consistent with temporal trends observed in this study. The char in Amituk Lake had almost three times higher [Hg] than fish from the other study lakes, perhaps because it was the only lake that contained a wetland within the watershed. Wetlands are productive features in terms of vegetation, and wetland soils, including those in the Arctic, and are known to be important sources of MeHg (Loseto et al., 2004a; St. Louis et al., 1994). The small wetland area, or the reported potential geological Hg in Amituk Lake's watershed (Semkin et al., 2005), are potential reasons why this lake is higher in [Hg] in char than the other Cornwallis Lakes; however; our results below indicate that water chemistry differences are a more likely explanation.

Char, Resolute, and Meretta Lakes are intermittently hydrologically linked and have adjoining watersheds, and yet trends in the Arctic char [Hg] differ between the lakes. Meretta Lake is upstream of Resolute Lake, and there is some evidence that Resolute Lake was also affected by the waste water inputs based on elevated perfluorooctane sulfonate (PFOS) concentrations in water (Lescord et al., 2015b) and in catchment soils (Cabrerizo-Pastor et al., 2018). The history of contamination of the Meretta Lake catchment by waste waters may be a factor in the observed decline of [Hg] in char over time although [Hg] in the char were similar to North and Resolute lakes when sampling began in 2006. Meretta is in the process of returning to the pre-waste water reference state (Antoniades et al., 2011) and its current water chemistry is similar to that of North and Resolute Lakes (Table SI-10; Lescord et al., 2015a). The nutrient and contaminant signature of sewage or waste water can have long lasting effects in Arctic lakes due to slow turnover (Hermanson, 1998). Waste water or the nearby airport may have been sources of Hg, PFOS, and other contaminants. Regarding Hg, it is likely that the nutrients in the waste water stimulated methylating bacteria in the lake sediments, increasing the bioavailable Hg in Meretta, especially under the reported anoxic conditions (Antoniades et al., 2011). As the lake returns to oligotrophic status, MeHg production in the sediments could be rapidly declining, and the [Hg] in the char may reflect this decline.

Previous research demonstrates that lake and watershed factors play a large role in determining the condition of individuals (Munkittrick and Dixon, 1989), as well as the mercury burden of the fish (Munthe et al., 2007; Chen et al., 2005; Johnston et al., 1991). Previous studies which included the current study lakes found that the watershed area: lake area ratio was correlated to [Hg] in Arctic char (Gantner et al., 2010b) and watershed area was important for Hg in char (Lescord et al., 2015a); however watershed size was not a significant factor in this study. We found that lake-to-lake variation in char [Hg] is better explained by water chemistry features than watershed features or the lake morphology attributes, although lake morphology can influence water chemistry and aspects of the char populations therein (Rigét et al., 2000). A study of land-locked monomorphic Arctic char morphological variants in 35 Icelandic lakes found that the underlying geology, water source (spring-fed or runoff), and fish community structure were more influential than other physical factors (lake area, depth, or volume) but that these physical factors were nonetheless correlated to char morphological characteristics (Kristjánsson et al., 2011). In the absence of geological, community, or water source differences, it is likely that lake dimensions play a large role in shaping the benthic community of the lakes, which influences both the water chemistry properties and the char populations. A survey of land-locked char populations in 27 Greenland lakes ranging from >200 to 3.3 m in depth found that lake volume and depth were significantly positively correlated with the maximum size of char (Rigét et al., 2000). Further, Rigét et al. found that decreased water transparency due to glacial runoff led to fewer char compared to a similar sized, but not glacially-influenced lake in the same watershed. The Rigét et al. study demonstrates that both lake size and water transparency shape char populations.

Inputs of terrestrial organic matter to polar desert lakes are minimal, and food webs are supported largely by benthic primary production (Chételat et al., 2010), which could contribute to the relative importance of in-lake processes rather than watershed processes. Our water chemistry values were consistent with the findings of Michelutti et al. (2003) and Lescord et al. (2015a) for the study lakes and typical for ice-dominated oligotrophic Arctic lakes (Chételat et al., 2015). The relationship between DOC and Hg or MeHg in fish across lakes has been well studied (Driscoll et al., 1995). Typically, as lake DOC concentration increases, so do Hg or MeHg concentration in water (Watras et al., 1998; MacMillan et al., 2015) leading to greater Hg bioaccumulation (O'Driscoll et al., 2005), although the inverse relationship has also been reported (Kamman et al., 2005). This relationship holds up to a point: a study in tundra lakes the Mackenzie River Delta (Northwest Territories, Canada) region demonstrated that there is a concentration threshold of 8.5 mg C/L, and higher concentrations beyond this inhibit Hg bioaccumulation in amphipods (French et al., 2014). The oligotrophic study lakes are well below this threshold, and yet, our findings demonstrate the opposite relationship expected for DOC concentrations in water and [Hg] in biota. Recent work which included four of the lakes in this study demonstrated that MeHg bioaccumulation in lake food webs is strongly influenced by the MeHg:DOC ratio, with primary producers in low DOC lakes exhibiting greater uptake of MeHg along a DOC gradient (Chételat et al., 2018). Lakes which had low concentrations of DOC and MeHg in the waters were found to be more sensitive to MeHg contamination, resulting in increased [MeHg] in biota when DOC was lowest. This was attributed to more efficient uptake of MeHg from water and organic matter in oligotrophic systems where organic matter concentrations are low. Our findings support this: DOC and POC were inversely related to [Hg] in Arctic char. Overall, we found only slightly tighter relationship between profundal water chemistry and char [Hg] than surface water chemistry; probably in part due to the well-mixed nature of the lakes. This finding indicates that Hg monitoring programs would be better served by monitoring profundal water chemistry than surface water chemistry, particularly in systems where the food web is benthically based or in stratified systems. Additionally, in areas where the DOC concentrations are below the threshold of 8.5 mg C/L, it may be advisable to avoid consuming fish from the lakes with the clearest water. This "rule of thumb" warrants further investigation and if it holds true, would be a simple way to assess the potential Hg levels in char providing a guide for fish consumption and inform monitoring programs.

4.2. Temporal trends in Arctic char Hg concentrations and the influence of climate

There were two types in temporal trends in the total [Hg] in Arctic char from the six study lakes: three of the lakes significantly decreased, while three others did not exhibit a significant linear trend. In a small lake in southwest Greenland, [Hg] in landlocked Arctic char increased from 1994 to 2008 (Rigét et al., 2010). For both anadromous and non-anadromous Arctic char in Labrador, researchers report significant increases, decreases, and non-significant trends for 1977–78 to 2007–09, but this was based on only two time points, and in general, concentrations were largely unchanged over time (Van der Velden et al., 2015). The mismatch of trend directions for [Hg] in Arctic char, amid dramatic climate changes and decreased concentrations of gaseous elemental Hg in the Arctic atmosphere demonstrates the complexity of the Hg biogeochemical cycle and its impact on fish [Hg].

Mercury concentrations in Arctic char from Resolute Lake were positively correlated to (after corrections for multiple testing) sea ice duration in Resolute Bay. When p values were not adjusted, sea ice duration was positively correlated to Meretta and North Lake char [Hg] as well (Fig. 3). During the period of this study, a rapid decline in sea ice duration in Resolute Bay was observed (-3.1 days/year, $p = .00001$, cumulative decrease of 62 days). In the Alaskan Arctic, declines in lake ice have been linked to concurrent sea ice declines due to more open ocean area and warmer temperatures (Arp et al., 2015), and lake ice extent decline has been documented in other Arctic lakes over a wide geographical area (Šmejkalová et al., 2016; Smol et al., 2005). Lake ice can greatly influence productivity in Arctic lakes (Smol et al., 2005; Williamson et al., 2009) and may also influence Hg chemistry by reducing photoreduction of Hg^{+2} and MeHg, reducing evasion of gaseous Hg (Soerensen et al., 2016), and stabilizing the water column and reduced mixing, promoting anoxic (i.e., methylating) conditions near the sediment (Eckley and Hintelmann, 2006). The impact of declining lake ice may not influence char in a uniform manner across lakes because lake ice extent varies widely due to factors such as lake depth and wind fetch.

Snow fall was positively correlated to Amituk Lake char [Hg] (before and after p value adjustment). Similarly, in the modeling analysis, snow fall significantly predicted char [Hg] for Amituk. This relationship is supported by experimental evidence which demonstrated that snowmelt and the spring freshet are the primary source of total Hg (Semkin et al., 2005) and MeHg (Loseto et al., 2004b) to Amituk Lake. Topography, prevailing wind direction, and the course of snowmelt reaching the lakes play important roles in snow accumulation patterns in the polar desert and may be of interest for further investigation which make Amituk Lake unique among the study lakes. Amituk Lake is of interest due to the exceptionally high concentration of Hg (and other metals) in Arctic char from this lake (Muir et al., 2005; Barst et al., 2016). In this study, we found that the water of this lake has low concentrations of dissolved and particulate organic carbon (even when compared to the other oligotrophic lakes), which likely increases the susceptibility of biota to Hg contamination (Chételat et al., 2018). This sensitivity, when combined with the demonstrated importance of snow melt as a source of Hg and MeHg to this lake (Semkin et al., 2005; Loseto et al., 2004b) are likely factors which set this Hg "hot spot" apart from the other lakes in the study and warrant further investigation.

The mixed modeling analysis indicated that for the Resolute Lake char, the NAO index was significantly negatively related to [Hg], The NAO was weakly positively correlated to POPs in char from three of the study lakes in a previous analysis (Cabrerizo et al., 2018). The statistically weak findings in both studies could indicate that NAO is important for these char populations, but differences in contaminant pharmacokinetics may drive the inverse relationship indicated by the coefficients between the two studies. In its positive phase, the NAO is important for movement of air masses from lower latitudes poleward; it can therefore be associated with warmer temperatures in the Arctic and cooler temperatures at lower latitudes, as air masses overlying these two regions mix (Ambaum et al., 2001). The NAO exerts strong control over the transport of atmospheric contaminants poleward (Eckhardt et al., 2003; Octaviani et al., 2015), consistent with previously reported positive relationships to POPs in Arctic char. It is noteworthy that NAO was not a significant explanatory variable with any char population Hg concentration in the correlation analysis. In the modeling analysis, its significance is reliant on the presence of the variable rain fall, without which it is no longer a significant predictor for Resolute Arctic char Hg (though rain and NAO are not correlated), further demonstrating that the relationship is statistically weak.

Environmental conditions have been shown to influence temporal trends in predatory fish growth and [Hg] in small lakes, both winter temperature ($-$) and mean annual temperature ($+$) were implicated as drivers (Lucotte et al., 2016). Evans et al. (2013) found that temperature was the most consistent explanatory variable of [Hg] in predatory fish in Great Slave Lake although inclusion of the PNAP improved explanatory power for trends of Hg in lake trout. Here we did not find a consistent significant effect of temperature on [Hg] in the study populations, despite steadily increasing mean annual temperatures during most of the study period. In contrast to previous studies for persistent organic pollutants on the same char populations (Cabrerizo et al., 2018), precipitation and sea ice were stronger predictors of char Hg than climate oscillations, but the same oscillation was weakly implicated for the Cornwallis Island char populations in both studies.

The correlations of climate variables between Resolute Lake and Amituk Lakes, which have the longest sampling history, indicates that additional sampling years may reveal additional climate effects using statistical analyses. This exploratory analysis has demonstrated strong statistical links between two of the six monitored populations and two rapidly changing climate variables but did not detect any variables which were significant across all the lakes. Further work is warranted to investigate the effects of ice extent on contaminant burdens in Arctic char in additional populations and continued study of the populations described here.

4.3. Summary

This statistical analysis found that in six neighboring high Arctic lakes with significantly different mean [Hg] in char, trends in [Hg] in landlocked Arctic char over time either decreased, or exhibited no clear linear trends amid concurrent declines in atmospheric gaseous [Hg], (Steffen et al., 2015). Lake-to-lake differences are likely due to organic carbon content and Hg processing of each of the lakes; watershed characteristics were not correlated to [Hg] in fish. No single climate factor influenced all populations consistently, but local sea ice duration was significantly positively correlated to [Hg] for the Resolute population, and snow fall was positively related to [Hg] in the Amituk population. The study lakes with the most complete sampling history, Resolute and Amituk Lakes, had the strongest relationships with climate variables. This highlights the need for continued monitoring and high-resolution sampling to better understand the roles of multiple stressors on biomonitoring targets while demonstrating that "hot spots" of Hg contamination may respond differently to the changing climate. Overall, we demonstrate that population-level differences in animal [Hg] temporal trends and climate influences can occur even at very fine geographic scales, and that oversimplification of these trends should be avoided for larger-scale studies.

Conflict of interest

The authors declare no conflict of interest.

Associated content

Open data portal links:
Contaminant and biological data on landlocked Arctic char
https://open.canada.ca/data/en/dataset/44319e2b-135c-4585-a6ab-87cb29ee2acf
Mercury in landlocked Arctic char
https://open.canada.ca/data/en/dataset/90e55c10-6fef-4387-a03f-6cd1a644e8b3

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.04.453.

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