Science of the Total Environment 509–510 (2015) 195–205

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Factors affecting biotic mercury concentrations and biomagnification through lake food webs in the Canadian high Arctic

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## HIGHLIGHTS

- Mercury (Hg) in Arctic char and invertebrates from 6 Arctic lakes were compared
- Food web biomagnification of Hg was variable across lakes
- Aqueous ions were negative predictors of benthic invertebrate [MeHg]
- Catchment size and nitrate were negative predictors of [THg] in young char
- [Hg] in these biota were affected by physical-chemical characteristics of lakes

## ARTICLE INFO

*Article history:*
Received 2 October 2013
Received in revised form 21 April 2014
Accepted 28 April 2014
Available online 6 June 2014

*Keywords:*
Mercury
Biomagnification
Food web
Arctic char
Stable isotopes
Principal component analysis

## ABSTRACT

In temperate regions of Canada, mercury (Hg) concentrations in biota and the magnitude of Hg biomagnification through food webs vary between neighboring lakes and are related to water chemistry variables and physical lake features. However, few studies have examined factors affecting the variable Hg concentrations in landlocked Arctic char (*Salvelinus alpinus*) or the biomagnification of Hg through their food webs. We estimated the food web structure of six high Arctic lakes near Resolute Bay, Nunavut, Canada, using stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopes and measured Hg (total Hg (THg) in char, the only fish species, and methylmercury (MeHg) in chironomids and zooplankton) concentrations in biota collected in 2010 and 2011. Across lakes, $\delta^{13}C$ showed that benthic carbon (chironomids) was the dominant food source for char. Regression models of log Hg versus $\delta^{15}N$ (of char and benthic invertebrates) showed positive and significant slopes, indicting Hg biomagnification in all lakes, and higher slopes in some lakes than others. However, no principal components (PC) generated using all water chemistry data and physical characteristics of the lakes predicted the different slopes. The PC dominated by aqueous ions was a negative predictor of MeHg concentrations in chironomids, suggesting that water chemistry affects Hg bioavailability and MeHg concentrations in these lower-trophic-level organisms. Furthermore, regression intercepts were predicted by the PCs dominated by catchment area, aqueous ions, and MeHg. Weaker relationships were also found between THg in small char or MeHg in pelagic invertebrates and the PCs dominated by catchment area, and aqueous nitrate and MeHg. Results from these high Arctic lakes suggest that Hg biomagnification differs between systems and that their physical and chemical characteristics affect Hg concentrations in lower-trophic-level biota.



## 1. Introduction

Mercury (Hg) is a global pollutant because it undergoes long range transport to remote areas like the Canadian high Arctic. During the polar spring, gaseous elemental Hg (GEM) in the atmosphere photochemically reacts with halogen radicals to form particulate Hg(II), a process known as atmospheric Hg depletion events (AMDEs; Steffen et al., 2008). Hg(II) is subsequently deposited in wet and dry deposition around the high Arctic (Steffen et al., 2008; Loseto et al., 2004). In the summer, snow and ice melt water carry Hg(II) into lakes, where it can be methylated by sulfate reducing bacteria into its more toxic form, methylmercury (MeHg; Schaefer and Morel, 2009; Lehnherr et al., 2012a). In addition to within-lake methylation, snow melt also provides an important external source of MeHg to these lakes (Loseto et al., 2004; Semkin et al., 2005). Due to its rapid accumulation in protein-rich tissues and slow excretion, MeHg is the form of Hg that is bioaccumulated in organisms and biomagnified through food webs (Kidd et al., 2012;

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http://dx.doi.org/10.1016/j.scitotenv.2014.04.133
0048-9697/

Kidd and Batchelar, 2012). Across Canada, Alaska and Greenland, char are often reported with Hg concentrations exceeding governmental consumption guidelines (0.5 ppm, wet wt.; Douglas et al., 2012; Health Canada, 2007).

Arctic char, the top predator in high Arctic lakes, has mean total Hg (THg; 95% of which is MeHg; Swanson and Kidd, 2010) concentrations that vary by up to 4.2-fold between neighboring (<25 km) systems (means differ by up to 0.35 μg/g, wet wt; $p < 0.05$; Gantner et al., 2010b). Part of this variability can be explained by differences in the physical and biological characteristics of the fish themselves; generally, older, longer, heavier, and higher-trophic-level fishes have higher Hg concentrations (Muir et al., 2005; Gantner et al., 2009; Swanson and Kidd, 2010). Physical and chemical characteristics of lakes can also affect Hg concentrations and biomagnification through food webs (Kidd et al., 2012; Clayden et al., 2013). More specifically, fish and other biota from larger and deeper lakes and ones with higher catchment area: lake area ($A_C$:$A_L$) ratios have higher Hg concentrations (Evans et al., 2005; Clayden et al., 2013). Although water chemistry variables, such as dissolved organic carbon (DOC), have complex relationships with Hg, lakes with higher nutrients (indicative of productivity) tend to have biota with lower Hg concentrations (Ulrich et al., 2001) likely due to a dilution of Hg over a larger total biomass, an effect known as biodilution (Chen et al., 2004). Compared to southern environments, Arctic lakes can be very unproductive, with highly variable climatic and physiographic characteristics that are known to affect Hg transport and cycling (Douglas et al., 2012). However, little is known about how the size, nutrient concentrations, catchment area, and depth of high Arctic lakes affect biotic concentrations of Hg or its biomagnification through the food webs. Additionally, many water chemistry variables are inherently related, making statistical analyses of their effects on Hg concentrations in biota challenging.

To quantify the biomagnification of Hg through aquatic food webs, its concentrations in biota are regressed against their $\delta^{15}N$ values (representative of trophic position; Jardine et al., 2006). This approach has been used extensively in systems from the tropics (Chasar et al., 2009; Chumchal et al., 2011; Kwon et al., 2012) to the Arctic (Atwell et al., 1998; Campbell et al., 2004; Gantner et al., 2012) and Hg concentrations in biota are consistently, positively related to their $\delta^{15}N$ values in aquatic food webs. The slopes resulting from these regressions represent the magnitude of and average Hg transfer through food webs (Wyn et al., 2009; Lavoie et al., 2013), which vary between lakes for reasons that are mechanistically not understood (Kidd et al., 2012).

Our study compared the Hg concentrations of invertebrates and char, as well as the Hg biomagnification slopes, across six high Arctic lakes near Resolute Bay, Nunavut (NU), Canada. Given the well established relationships between biological characteristics (e.g. size, age, trophic level) and Hg concentrations in fish, we focussed this study on abiotic factors and examined whether the physical and chemical features of the lakes, which could affect the bioavailability of MeHg, would explain among-lake variability in Hg biomagnification through food webs and Hg concentrations in fish and invertebrates.

## 2. Methods

### 2.1. Study site

Six ultra-oligotrophic lakes located in the central Canadian Arctic Archipelago were chosen for our study. These lakes (Meretta, Resolute, Char, Small, North, and 9-Mile) are within a 20 km radius of one another and are located in the southwest corner of Cornwallis Island, NU, Canada (75°08′N 95°00′W; Fig. 1). Cornwallis Island is a polar desert, with a mean annual temperature and precipitation of - 16.4 °C and 150 mm, respectively (Antoniades et al., 2011). To the best of our knowledge, the six lakes are landlocked (fish have no marine access) and char are the only fish species present. Char in this study had low $\delta^{34}S$ values (ranging from - 10.06 to - 3.04‰; see supporting information), further supporting that these fish populations do not access marine environments (compared to anadromous char, which have higher $\delta^{34}S$ values (e.g. 18.02‰); Swanson et al., 2011).

Many of the lakes are used for recreational fishing by the local Inuit community of Resolute Bay (Gantner et al., 2010b) and have been studied since the 1970s (e.g. Schindler et al., 1974). Lake areas, depths, and catchment areas used herein were obtained from Muir et al. (2009) and Gantner et al. (2010b); water depths were confirmed with a depth sounder at the time of sampling. All field work was conducted from the Polar Continental Shelf Program Base (PCSP, Natural Resources Canada).

### 2.2. Sample collection

Lakes were sampled for biota and water chemistry from early July to mid-August in 2010 and 2011. Compared to southern systems, these high Arctic lakes are unproductive and have low biomasses of invertebrates.

#### 2.2.1. Invertebrates

Larval and emerging adult chironomids (Order Diptera) were collected using kick sweeps and aspirators, respectively (1–3 samples/lake/year for both life stages). After the spring melt, additional emerging chironomids were collected by dragging dip nets along the water surface. In 2011, lake ice melted earlier than expected (~July 9th) and adult invertebrates were aspirated from rocks along lake shores. To collect larval chironomids, the littoral zone of each lake was sampled two to four times per year and individuals were picked from bulk sweeps and frozen in clean glass or polypropylene vials. All chironomids were identified and separated based on trophic ecology; predatory chironomids in the subfamily Tanypodinae and herbivorous chironomids from the subfamilies Orthocladiinae or Chironomidae were separated for all analyses. These lakes have a relatively low diversity of chironomids (Scott et al., 2011) which allows for this broader separation of taxa, as in previous studies (Chételat et al., 2008; Gantner et al., 2010a).

Pelagic zooplankton were collected once the ice had receded using a Wisconsin® net (20 μm mesh). During each sampling period (1–4 times/lake/year), nine samples were collected: three bulk samples were separated into three size classes (<250 μm, 250–500 μm, and > 500 μm), similar to Gantner et al. (2010a). High Arctic zooplankton are comprised mainly of copepods (e.g. *Limnocalanus macrurus*) and, once sorted by size, samples were analyzed directly without further identification as per Chételat et al. (2012).

#### 2.2.2. Arctic char

At the end of the field season, large (fork length >18 cm; $n = 10–20$ fish/lake/year) and small ($n = 10–20$ fish/lake in 2010; 5–7/lake in 2011; fork length <18 cm) char were collected using gill nets and a Smith-Root® electrofisher, respectively. Total sample sizes were comparable between five of the six lakes ($n = 30 \pm 4$); the exception was Char Lake ($n = 13$) where prolonged fishing was less successful. At the field station, all fish were weighed and measured (fork length, FL; and total length, TL) and large fish were dissected for dorsal muscle and otoliths. Small char were frozen whole for later tissue sampling. Dorsal muscle was removed from all small char (mean FL = 9.6 ± 2.9 cm) except those with FL < 4.8 cm; these fish were analyzed whole. The percentage of small fish analyzed as whole body homogenates were 15 (Char Lake), 19 (Small Lake), 31 (Meretta Lake), 63 (North Lake), 67 (Resolute Lake) and 93% (9-Mile Lake; see SI for further details); for statistical tests, muscle Hg concentrations were estimated using Peterson et al. (2005; see SI for details).

#### 2.2.3. Water and periphyton

Since baseline $\delta^{15}N$ values can differ between lakes (Chételat et al., 2010), periphyton samples were scraped from littoral rocks at three sites on each lake in both 2010 and 2011. THg and MeHg water samples were obtained weekly in 2010 and biweekly in 2011 at the surface and

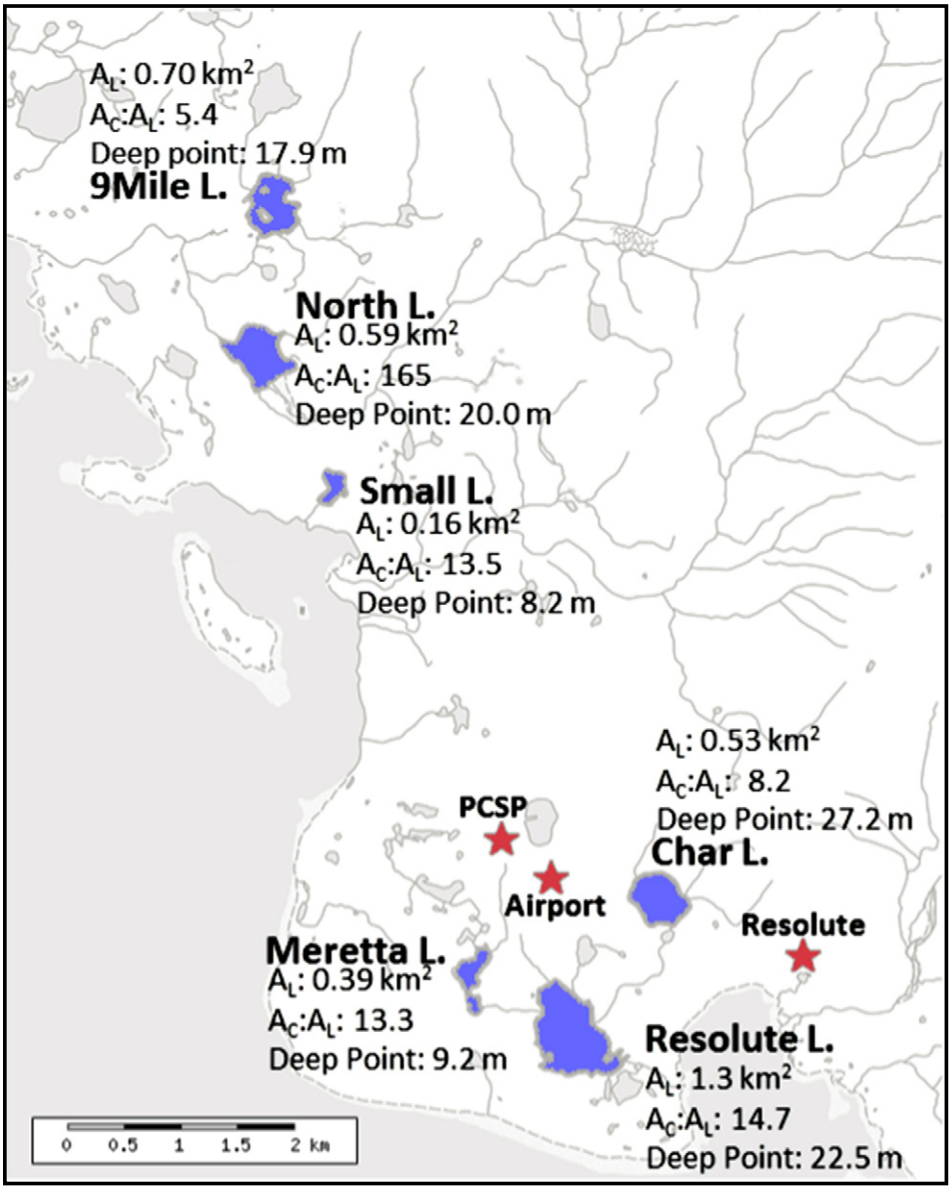


**Fig. 1.** Southwest corner of Cornwallis Island, Canada, showing the six lakes and their corresponding physical characteristics (Muir et al., 2005, 2009; Gantner et al., 2010b). $A_L$: Lake area; $A_C$:$A_L$; Catchment area to lake area ratio (NRC, 2012).

deep point of each lake. Water was collected using clean methods (Kirk and St Louis, 2009) and a pre-cleaned, 2 L Niskin water sampler, then stored in pre-cleaned 250 mL amber glass bottles. Unfiltered water samples were preserved with trace-grade hydrochloric acid (HCl) equivalent to 0.2% by volume and stored in a dark cold room (~4–5 °C) until analyzed. Bulk water samples (1 L) for analysis of water chemistry variables were also collected at the same time and at the same depths as the Hg water samples. Subsamples were filtered immediately after collection for DOC, dissolved inorganic carbon (DIC), particulate organic carbon (POC), particulate organic nitrogen (PON), and chlorophyll *a* (chl *a*). GF/C filter paper was used (47 mm, pore size 1.2 μm for chl *a*/POC/PON; 25 mm, pore size 0.45 μm for DOC/DIC). Unfiltered subsamples were collected directly from bulk samples for analysis of major ions, nitrite and nitrate ($NO_3^-$/$NO_2^-$), total phosphorus (TP), total nitrogen (TN), pH, alkalinity, and conductivity (cond). All standard water chemistry samples were analyzed at the *National Laboratory for Environmental Testing* at the Canada Centre for Inland Waters (CCIW) in Burlington, Ontario.

### 2.3. Laboratory analyses

#### 2.3.1. Stable isotope analysis

All biotic samples were dried using a LabconcoFreeZone© freeze-drier prior to analyses and percent moistures determined. Delta$^{13}$C and $\delta^{15}$N analysis was performed on subsamples of homogenized biota, terrestrial vegetation, and periphyton at the Stable Isotopes in Nature Laboratory at the University of New Brunswick, Fredericton, Canada using a Costech 4010 Elemental Analyzer interfaced with a Finnigan Delta Plus Mass Spectrometer. Isotope values were calculated as a ratio to a known standard similar to Chételat et al. (2010) and Post (2002).

### 2.4. Mercury analysis

#### 2.4.1. MeHg in biota

Subsamples of invertebrate homogenates were analyzed for MeHg and Hg(II) at the Center for Analytical Research on the Environment

(CARE) at Acadia University using methods of Edmonds et al. (2012). Samples of approximately 10.0 mg were weighed using a Sartorius ultra-microbalance and then digested in glass vials with Teflon caps using 25% methanol: potassium hydroxide. Aliquots of digest were ethylated with sodium tetraethyl borate ($NaBEt_4$) and volatile species captured on Tenax traps. In each sample, MeHg and Hg(II) were separated by gas chromatography (GC) and measured with atomic fluorescence spectrometry (AFS; Bloom and Fitzgerald, 1988; Edmonds et al., 2010). No samples were below the mean method detection limit (MDL; 3 times the standard deviation of blank concentrations) of 2.65 ng/g (based on a sample mass of 10.0 mg dry wt.).

*2.4.2. THg in fish*

Subsamples of large (n = 168) and small (n = 94) char were analyzed for THg using a Milestone Direct Mercury Analyzer (DMA-80) at the University of New Brunswick in Saint John (UNBSJ) in 2010 and at CCIW in 2011, using the methods described in Clayden et al. (2013). No samples were below the mean MDL of 0.04 μg/g (n = 59; UNBSJ) or 0.07 μg/g (n = 45; CCIW), based on a mean sample weight of 10.0 mg dry wt. Twenty large char were also analyzed for MeHg and Hg(II) using the AFS analysis described above and THg concentrations in 2010 and 2011 char were on average 92 ± 28 and 99 ± 16% MeHg, respectively.

*2.4.3. THg and MeHg in water*

Water samples were analyzed at the Low-Level Mercury Analytical Laboratory at CCIW using standard US EPA Methods 245.7 and 1630 for THg and MeHg, respectively (EPA, 1998, 2005). Briefly, THg water samples were analyzed by potassium bromide (KBr) oxidation, stannous chloride ($SnCl_2$) reduction, and detection by AFS. No water samples were below the calculated MDL of 0.04 ng/L. MeHg analysis in water was performed using a Brooks Rand MERX automated MeHg analyzer. Briefly, analysis was carried out by distillation, ethylation with NaBEt4, GC separation, and detection by AFS. Because blanks were low and consistent (average 0.005 ± 0.001 ng/L; n = 12), the calculated mean MDL was 0.005 ng/L and no samples were below this MDL. When a more conservative MDL of 0.01 ng/L is considered, 17 out of 86 samples were below the detection limit. All data were included in mean calculations and statistical analyses.

*2.5. Quality control*

Details on quality assurance measures for all laboratory analyses can be found in the supplemental information (SI). Briefly, percent recovery of certified reference materials (CRMs) and spiked samples ranged from 90 to 106% across all laboratory analyses; relative percent differences between method duplicates ranged from <1 to 7% across all laboratory analyses.

*2.6. Data analysis*

*2.6.1. Data handling and corrections*

All statistical analyses and graphing were done with SPSS 19.0 and Sigma Plot 11.0. Data were visually inspected for normality and homoscedasticity using Kolmogorov–Smirnov and Levene's Tests, respectively. When appropriate, data were $Log_{10}$ transformed. Mann–Whitney U tests showed no among-year differences in Hg in large char, small char, benthic invertebrates, or pelagic invertebrates for each lake and therefore data were combined within taxa and across years for subsequent statistical analyses. It should be noted that tests for between-year differences in invertebrate Hg concentrations were only run in four out of the six lakes due to low sample sizes (n < 3) for some years (see Table 3). Due to low sample sizes, data for all zooplankton size classes within a lake were considered "pelagic" invertebrates for all statistical analyses. Similarly, data for all chironomid samples (herbivores and predators, larva and adults) were combined and analyzed as "benthic" invertebrates.

Due to highly variable percent moistures in invertebrates (e.g. 44–83% in chironomids), all statistical analyses of biological data were done using dry wt. Hg concentrations. In addition, no isotope data were lipid corrected; C:N ratios were generally low (ranging from 3.2 to 3.9 in large char; 3.6 to 4.1 in small char; and 4.7 to 5.2 in benthic invertebrates) and no relationships were found between C:N ratios and $\delta^{13}C$ values across all lakes in large char (linear regression; $p = 0.813$; $r^2 = 0.002$), small char ($p = 0.579$; $r^2 = 0.001$), or benthic invertebrates ($p = 0.260$; $r^2 = 0.023$). All mentions of small fish Hg concentrations refer to muscle THg, unless otherwise stated.

*2.6.2. Stable isotope data analyses*

To assess the contribution of benthic and pelagic food sources in the chars' diet, standard one-isotope linear mixing models were applied (IsoSource, IsoError: Phillips and Gregg, 2003; Chételat et al., 2010; see SI for more details). Percentages above 100% are likely due to interspecies variation in $\delta^{13}C$ fractionation (Post, 2002) and, although they suggested benthic feeding by char, they should be interpreted with caution given the limitations of mixing models. Invertebrate $\delta^{13}C$ values were corrected for trophic fractionation (0.5 and 0.4‰ for chironomids and zooplankton, respectively) before entering data into the model, as per Chételat et al. (2010). In each lake, separate models were run for large and small char.

*2.6.3. Hg concentrations and biomagnification regressions*

Before comparing char THg concentrations between lakes, variability in Hg due to fish size was removed using the residual errors of within-lake linear regressions between log Hg concentrations and char fork lengths ($THg_{length}$; see Gantner et al., 2010b; Swanson et al., 2010; relationships between Hg concentrations and fish fork length, weight and age can be found in the SI). These residuals were compared between lakes using Kruskal–Wallis H Tests (and Mann–Whitney U post hoc tests). Similar to fish, residuals resulting from linear regressions between MeHg concentrations and $\delta^{15}N_{adj}$ (see below) in invertebrates were also compared between lakes.

Linear regressions between Log Hg (dry wt.; THg in fish and MeHg in invertebrates; Wyn et al., 2009) and $\delta^{15}N$ (unadjusted) were run within each lake to determine the slope (average Hg biomagnification through food web) and intercept. Regression slopes were calculated using benthic invertebrate and char data only (zooplankton were excluded) because the latter species fed mainly on benthic carbon sources (as recommended by Borgå et al., 2012). In these models, whole body THg estimates were used instead of muscle THg for all small and large char (as per Peterson et al., 2005; see SI). Triplicate periphyton scrapings were used to determine baseline $\delta^{15}N$ in each lake and these values were subtracted from those of other biota ($\delta^{15}N_{adj}$) before among-lake comparisons were done, and isotope fractionation from prey to predator was assumed to be consistent across lakes. The ANCOVA model Log Hg [THg (char, whole-body estimates) and MeHg (invertebrates), dry wt.] = lake + $\delta^{15}N_{adj}$ + $\delta^{15}N_{adj}$*lake was run and the interaction term used to determine whether slopes were significantly different across lakes.

*2.6.4. Predictors of Hg concentrations in biota and biomagnification slopes in food webs*

Before biotic Hg concentrations were compared between lakes, Hg concentrations were first standardized to a common length (for fish, 35.1 cm and 6.9 cm for large and small char, respectively) or trophic level ($\delta^{15}N_{adj}$; for invertebrates, 3.9 and 4.0‰ for benthic and pelagic groups, respectively) using ANCOVA models (see SI for details). The resulting least squared means for Hg ($LSM_{THg}$ for fish; $LSM_{MeHg}$ for invertebrates) were used in all correlation and regression analyses for these groups.

A paired Mann–Whitney *U* test was run to detect differences between Hg concentrations in surface and deep water samples across lakes. Given the inherent collinearity between the 23 water chemistry

variables and 3 physical lake features examined, all of these data (log-transformed and standardized to a mean of 0 and a standard deviation of 1; Pett et al., 2003) were entered into a principal component analysis (PCA) and the data were reduced to five principal components (PCs). PCA reduces large data sets of related variables by identifying vectors within the data; the extracted PCs account for the greatest variability in a data set (Graham, 2003). After extraction, PCs can be used in multiple regression models without statistical issues from multicollinearity between predictor variables; in addition, the orthogonal nature of the PCs produces stable regression results despite potential collinearity between components (Graham, 2003). Within each component, the dominant variables were identified based on the rotated loadings (representative of the correlation between each variable and a component; Pett et al., 2003); any variable with a loading >|0.7| was considered dominant (see Table 2). Backward elimination ($\alpha < 0.10$) stepwise multiple regressions were run across lakes to determine which PCs best predicted regression (Log Hg vs. $\delta^{15}N_{adj}$) slopes or intercepts, or LSM Hg concentrations of the four biotic groups (large char, small char, benthic invertebrates, and pelagic invertebrates). All models were compared using a version of Akaike's Information Criterion which corrects for small sample sizes ($AIC_C$; Burnham and Anderson, 2004).

It should be noted that North Lake was a consistent outlier in PC regression analyses (based on Cook's Distance; Leech et al., 2008) due to its large catchment area (relative to lake size; 165). However, given that this study only examined 6 lakes and that this large catchment area appears to be related to other water chemistry variables, North Lake was included in all analyses. In 9-Mile Lake, one of the soluble reactive phosphorus (SRP; n = 14) measures (253 μg/L) was a statistical outlier and was removed from mean calculations (see SI). Alpha ($\alpha$) was set at 0.05 for all statistical analyses.

**Table 1**
Mean (±SD) THg and MeHg concentrations (ng/L) and % MeHg in surface and deep water from lakes sampled in 2010 and 2011 near Resolute Bay, Cornwallis Island, Canada.

| Lake | Year | n | MeHg (ng/L) | THg (ng/L) | % MeHg |
|---|---|---|---|---|---|
| | | | Surface samples | | |
| Meretta | 2010 | 5 | 0.062 ± 0.017 | 0.707 ± 0.165 | 9.0 ± 2.5 |
| | 2011 | 3 | 0.044 ± 0.012 | 0.704 ± 0.107 | 6.3 ± 1.9 |
| | Total | 8 | 0.055 ± 0.017 | 0.706 ± 0.137 | 8.0 ± 2.5 |
| Resolute | 2010 | 4 | 0.015 ± 0.002 | 0.405 ± 0.088 | 3.8 ± 0.9 |
| | 2011 | 3 | 0.017 ± 0.002 | 0.717 ± 0.355 | 2.6 ± 1.0 |
| | Total | 7 | 0.016 ± 0.002 | 0.539 ± 0.272 | 3.3 ± 1.1 |
| Char | 2010 | 5 | 0.009 ± 0.002 | 0.349 ± 0.156 | 2.8 ± 1.0 |
| | 2011 | 3 | 0.008 ± 0.001 | 0.480 ± 0.063 | 1.8 ± 0.2 |
| | Total | 8 | 0.009 ± 0.001 | 0.399 ± 0.140 | 2.4 ± 1.0 |
| Small | 2010 | 4 | 0.019 ± 0.002 | 0.584 ± 0.138 | 3.3 ± 0.9 |
| | 2011 | 3 | 0.031 ± 0.012 | 0.661 ± 0.085 | 4.8 ± 1.6 |
| | Total | 7 | 0.024 ± 0.010 | 0.617 ± 0.117 | 4.0 ± 1.4 |
| North | 2010 | 4 | 0.023 ± 0.011 | 0.887 ± 0.175 | 2.6 ± 1.0 |
| | 2011 | 3 | 0.015 ± 0.002 | 0.787 ± 0.081 | 1.9 ± 0.2 |
| | Total | 7 | 0.019 ± 0.009 | 0.844 ± 0.143 | 2.3 ± 0.8 |
| 9-Mile | 2010 | 3 | 0.020 ± 0.019 | 0.440 ± 0.060 | 5.1 ± 5.5 |
| | 2011 | 3 | 0.010 ± 0.003 | 0.584 ± 0.103 | 1.9 ± 0.9 |
| | Total | 6 | 0.015 ± 0.013 | 0.512 ± 0.109 | 3.5 ± 3.9 |
| | | | Deep samples | | |
| Meretta | 2010 | 5 | 0.068 ± 0.053 | 0.667 ± 0.091 | 10.1 ± 7.2 |
| | 2011 | 3 | 0.056 ± 0.007 | 0.741 ± 0.100 | 7.7 ± 2.0 |
| | Total | 8 | 0.064 ± 0.041 | 0.635 ± 0.095 | 9.2 ± 5.7 |
| Resolute | 2010 | 4 | 0.021 ± 0.004 | 0.388 ± 0.064 | 5.5 ± 1.0 |
| | 2011 | 3 | 0.018 ± 0.003 | 0.548 ± 0.090 | 3.3 ± 1.1 |
| | Total | 7 | 0.020 ± 0.004 | 0.457 ± 0.110 | 4.6 ± 1.5 |
| Char | 2010 | 5 | 0.010 ± 0.003 | 0.240 ± 0.051 | 4.2 ± 1.3 |
| | 2011 | 3 | 0.010 ± 0.002 | 0.522 ± 0.052 | 1.9 ± 0.5 |
| | Total | 8 | 0.010 ± 0.003 | 0.343 ± 0.156 | 3.3 ± 1.6 |
| Small | 2010 | 4 | 0.018 ± 0.003 | 0.490 ± 0.042 | 3.7 ± 0.5 |
| | 2011 | 3 | 0.027 ± 0.003 | 0.673 ± 0.077 | 4.0 ± 0.3 |
| | Total | 7 | 0.022 ± 0.005 | 0.569 ± 0.111 | 3.8 ± 0.4 |
| North | 2010 | 4 | 0.021 ± 0.013 | 0.908 ± 0.159 | 2.4 ± 1.4 |
| | 2011 | 3 | 0.020 ± 0.003 | 0.795 ± 0.085 | 2.6 ± 0.3 |
| | Total | 7 | 0.021 ± 0.01 | 0.860 ± 0.137 | 2.4 ± 1.0 |
| 9-Mile | 2010 | 3 | 0.008 ± 0.004 | 0.432 ± 0.023 | 2.0 ± 0.9 |
| | 2011 | 3 | 0.025 ± 0.014 | 0.609 ± 0.143 | 4.3 ± 3.1 |
| | Total | 6 | 0.016 ± 0.013 | 0.521 ± 0.133 | 3.1 ± 2.4 |

## 3. Results

### 3.1. Chemical and physical lake features

As is common in high Arctic systems, the lakes in our study generally had low primary productivity and nutrients (chl *a*, DOC, and PON; see SI for detailed tables). Water chemistry variables did, however, vary across lakes. For example, North Lake had 10 to 12 times higher concentrations (~0.12 mg/L) of nitrate and nitrite ($NO_3^-/NO_2^-$) when compared to all other lakes (0.01 mg/L in all cases; not statistically tested). Resolute Lake had the highest mean conductivity (360.3 μs/cm) and ion concentrations (18.94 mg/L, sodium (Na), surface sample), especially when compared to North Lake (lowest mean values of 167.3 μs/cm, 3.98 mg/L, respectively). Although Meretta Lake has a history of sewage inputs from 1949 to 1998 (Chételat and Amyot, 2009; Antoniades et al., 2011), it had similar chl *a* (1.29 ± 1.04 μg/L), DOC (1.74 ± 0.14 mg/L), DIC (15.84 ± 1.55 mg/L), and POC (0.24 ± 0.08 mg/L) concentrations in 2010 and 2011 to all other lakes.

Although MeHg concentrations in deep water samples (range 10–64 pg/L across lakes) were typically higher than those of surface waters within the same lake (8–55 pg/L across lakes; Table 1), no statistical differences between surface and deep water MeHg or %MeHg (p = 0.884 and 0.479, respectively; Mann–Whitney *U* Test) were found. North Lake had the lowest % MeHg (2.3 ± 0.8% and 2.4 ± 1.0% for surface and deep samples, respectively), implying a lower methylation rate. In contrast, Meretta Lake had the highest MeHg concentrations (0.055, 0.064 ng/L, surface and deep samples, respectively) and %MeHg (8.0, 9.2%, surface and deep samples, respectively). Similar to MeHg, no differences in aqueous THg concentrations were found between shallow and deep water samples across lakes (p = 0.497; Mann–Whitney *U* Test).

**Table 2**
Rotated component matrix of the 5 principal components (PCs) extracted using a principal component analysis. Rotations were converged in 9 iterations using varimax rotation with Kaiser Normalization.

| Variable | PC 1 | PC 2 | PC 3 | PC 4 | PC 5 |
|---|---|---|---|---|---|
| Depth | −.019 | −.835* | −.042 | −.391 | .386 |
| Lake Area | .011 | −.558 | .089 | −.036 | .824* |
| $A_C$:$A_L$ | −.975* | .117 | .171 | −.079 | .023 |
| THg | −.884* | .278 | −.348 | .135 | .045 |
| MeHg | −.120 | .189 | −.034 | .966* | −.123 |
| %MeHg | .222 | .365 | .235 | .873* | −.006 |
| $NO_3NO_2$ | −.889* | −.036 | −.390 | −.229 | .057 |
| $NH_3$ | −.148 | .708* | .383 | .574 | .002 |
| Conductivity | .589 | .081 | .797* | .103 | .013 |
| pH | .606 | −.172 | .602 | −.480 | −.099 |
| Alkalinity | .894* | .121 | .405 | −.133 | .069 |
| Cl | .256 | .349 | .849* | .286 | −.103 |
| $SO_4$ | −.217 | −.725* | .607 | .163 | .181 |
| Chl α | −.731* | .102 | −.459 | −.494 | .021 |
| DOC | .072 | .912* | .055 | .400 | −.030 |
| DIC | .902* | .092 | .384 | −.149 | .091 |
| Ca | .385 | −.478 | .777* | −.044 | .129 |
| Mg | .746* | .600 | .199 | .086 | .190 |
| Na | .419 | .165 | .860* | .203 | −.126 |
| K | .321 | −.208 | .919* | −.077 | .054 |
| POC | −.023 | .993* | .027 | −.099 | −.056 |
| PON | −.020 | .982* | .094 | .047 | −.158 |
| $SiO_2$ | .615 | .422 | .257 | −.604 | −.115 |
| SRP | .043 | .318 | .749* | −.372 | .445 |
| TN | −.557 | .792* | −.186 | .149 | .078 |
| TP | .294 | .691 | −.184 | .542 | .329 |
| Eigenvalue | 11.597 | 8.805 | 3.454 | 2.919 | 1.225 |
| Variance explained (%): | 28 | 28 | 23 | 15 | 5 |
| Sum variance explained (%): | 28 | 56 | 80 | 95 | 100 |

* Indicates dominant variables within each PC (based on loadings greater than 0.7).

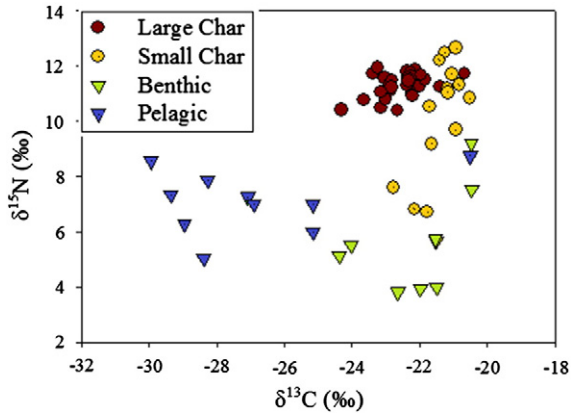


**Fig. 2.** $\delta^{15}N$ and $\delta^{13}C$ (‰) of Meretta Lake biota collected in both 2010 and 2011. Benthic samples include predatory and herbivorous larval and emerging chironomids. Pelagic samples include all zooplankton size classes (<250 μm, 250–500 μm, >500 μm).

Five principal components (PCs) were identified in the PCA of the lakes' physical and chemical characteristics and the loadings of individual variables for each are shown in Table 2. *PC1* was dominated by $A_C$:$A_L$, $THg_{water}$, $NO_3^-/NO_2^-$, alkalinity, DIC, chl *a*, and Mg; *PC2* was dominated by nutrients ($NH_3$, $SO_4$, DOC, POC, PON) and lake depth; *PC3* was dominated by cations (K, Na, Ca, Cl), SRP, and conductivity; *PC4* was dominated by $MeHg_{water}$ and $\%MeHg_{water}$; and *PC5* was dominated by lake area. Individually, *PC1*, *PC2*, *PC3*, *PC4*, and *PC5* accounted for 28%, 28%, 23%, 15%, and 5% of the variability in the data, respectively. It should be noted that the *PC 1* was largely driven by North Lake, which has an $A_C$:$A_L$ ratio (165) that is up to 30 times larger than those of the other 5 lakes (ratio range, 5.4 to 14.7; see Fig. 1).

### 3.2. Food web characterization

Delta$^{15}$N values in periphyton (baseline) samples were 2.4 ± 0.5, 3.7 ± 1.0, 1.6 ± 0.6, 1.0 ± 0.4, 2.5 ± 0.8, and 2.9 ± 0.6‰ in Meretta, Resolute, Char, Small, North, and 9Mile lakes, respectively. Baseline-corrected $\delta^{15}N$ values of large char ranged from 9.16 ± 0.54 to 11.31 ± 0.59‰ in fish from 9Mile and Char Lakes, respectively. Uncorrected $\delta^{15}N$ values in large char were between 9.70 (Char Lake) and 7.33‰ (9-Mile Lake) higher than their respective periphyton values. Within a lake, the $\delta^{15}N$ of large char indicated that they occupied the highest trophic level. Large char had mean $\delta^{15}N$ values that were 0.7 to 3.3‰ higher than those of small char, but their mean $\delta^{13}C$ values were similar (generally < 1‰ difference). As expected, pelagic invertebrates were low in $\delta^{13}C$, with mean values that were 2.8 to 8.0‰ lower when compared to benthic invertebrates from the same system. Isotope values of each trophic group can be found in the SI (Table SI-7).

According to the linear isotope mixing model using $\delta^{13}C$ values, large and small char consumed more benthic than pelagic carbon (e.g. Fig. 2). Benthic feeding was 115 ± 10, 136 ± 1, 150 ± 21, and 147 ± 39% in Resolute, Char, Small, and 9-Mile lakes, respectively for large char. In both Meretta and North Lakes, fish also mainly relied on benthic sources (76 ± 6 and 97 ± 8%, respectively) as opposed to pelagic carbon (23 ± 6 and 3 ± 8%, respectively). Small char diets were 94 ± 8, 94 ± 7, 114 ± 1, 171 ± 49, 79 ± 18, and 97 ± 24% benthic carbon for Resolute, Meretta, Char, Small, North, and 9-Mile lakes, respectively (see Table SI-8 for more details).

### 3.3. Hg concentrations

#### 3.3.1. Invertebrates

Residual MeHg concentrations in benthic ($p = 0.013$) and pelagic ($p < 0.001$) invertebrates were significantly different among lakes. However, only 9 (out of 30) post hoc lake-to-lake comparisons of invertebrate Hg concentrations were significant (see SI), likely due to limited samples sizes and lower statistical power (e.g. power ~0.2 for benthic invertebrates and ~0.9 for fish).

With the exception of Meretta Lake, benthic invertebrates were 9 to 27 times higher in MeHg than pelagic invertebrates in each lake (see Table 3). Benthic invertebrate $LSM_{MeHg}$ was significantly ($p = 0.017$) and negatively related to the *PC3* ($R^2_{adj} = 0.74$; Fig. 3). Several of the dominant variables in this PC (K, Na, Cl, and conductivity) were also individually significantly ($p = 0.002$–$0.046$; $r^2 = 093$–$0.67$) and negatively related to benthic $LSM_{MeHg}$ (see Fig. 4).

Pelagic $LSM_{MeHg}$ was significantly related to *PC1*, *PC2*, and *PC4* ($p = 0.015$, $R^2_{adj} = 0.975$); however, individually, only *PC4* was significantly ($p = 0.021$, $r^2 = 0.77$) related to Pelagic $LSM_{MeHg}$. This relationship was largely driven by the high pelagic MeHg concentration in Meretta Lake and, when removed, Pelagic $LSM_{MeHg}$ was no longer significantly related to *PC4* ($y = 0.31x - 1.72$; $r^2 = 0.11$; $p = 0.579$; see SI for graphs with PCs and individual parameters).

#### 3.3.2. Hg concentrations in char

In general, Hg concentrations in large char were low; only 4 fish from North Lake (0.51 to 1.38 μg/g, wet wt.) and 2 fish from Char Lake (0.49 and 0.47 μg/g, wet wt.) met or exceeded Health Canada's 0.5 μg/g (wet wt.) consumption guideline (Health Canada, 2007; wet weight Hg concentrations calculated using individual fish % moisture). North Lake had the highest mean Hg concentration in large char (unadjusted, 1.28 ± 0.99 μg/g, dry wt.), and Small Lake fish had the lowest mean Hg concentration (0.38 ± 0.17 μg/g, dry wt.; see Table 3). Mercury concentrations in large char differed in all between-lake comparisons ($p \leq 0.030$, comparing $THg_{Length}$ residuals) except for those of Small and Resolute Lakes ($p = 0.661$), the systems with the lowest char Hg concentrations, and those between Meretta, North, and Char lakes ($p = 0.948$–$0.118$), systems with higher fish THg. Relationships

**Table 3**
Mean (±SD) Hg concentrations in biota (THg in fish muscle (μg/g), MeHg in invertebrates (ng/g); unadjusted dry wt.) from six lakes near Resolute Bay, Cornwallis Island, Canada. NA = not applicable. Whole bodies were analyzed for a subset of small char; these data were converted to muscle equivalents as described in the Methods.

| | Year | n | Meretta | n | Resolute | n | Char | n | Small | n | North | n | 9-Mile |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Large char | 2010 | 20 | 0.87 ± 0.35 | 10 | 0.24 ± 0.12 | 9 | 0.38 ± 0.16 | 25 | 0.37 ± 0.18 | 17 | 1.22 ± 1.21 | 17 | 0.75 ± 0.39 |
| | 2011 | 10 | 0.49 ± 011 | 20 | 0.67 ± 0.32 | 4 | 1.72 ± 0.13 | 15 | 0.40 ± 0.16 | 15 | 1.35 ± 0.70 | 15 | 0.79 ± 0.60 |
| | Total | 30 | 0.75 ± 0.34 | 30 | 0.53 ± 0.33 | 12 | 0.79 ± 0.66 | 40 | 0.38 ± 0.17 | 32 | 1.28 ± 0.99 | 32 | 0.77 ± 0.49 |
| Small char | 2010 | 7 | 0.30 ± 0.15 | 7 | 0.26 ± 0.06 | 7 | 0.15 ± 0.03 | 9 | 0.35 ± 0.12 | 9 | 0.62 ± 0.25 | 6 | 0.27 ± 0.07 |
| | 2011 | 6 | 0.33 ± 0.04 | 8 | 0.23 ± 0.11 | 5 | 0.29 ± 0.07 | 7 | 0.29 ± 0.07 | 7 | 0.67 ± 0.27 | 9 | 0.18 ± 0.05 |
| | Total | 13 | 0.31 ± 0.11 | 15 | 0.24 ± 0.09 | 12 | 0.25 ± 0.09 | 16 | 0.33 ± 0.10 | 16 | 0.64 ± 0.25 | 15 | 0.22 ± 0.08 |
| Benthic inverts. | 2010 | 3 | 131.7 ± 121.9 | 6 | 64.0 ± 36.5 | 3 | 93.8 ± 31.5 | 1 | 174.2 | 4 | 195.1 ± 61.4 | 2 | 160 |
| | 2011 | 7 | 138.6 ± 97.4 | 6 | 79.0 ± 37.6 | 5 | 122.2 ± 73.3 | 4 | 82.1 ± 22.7 | 3 | 151.5 ± 22.3 | 3 | 160.5 ± 43.7 |
| | Total | 10 | 136.5 ± 98.2 | 12 | 71.5 ± 36.2 | 8 | 111.5 ± 59.7 | 5 | 100.5 ± 45.6 | 7 | 176.4 ± 50.9 | 5 | 160.3 ± 54.7 |
| Pelagic inverts. | 2010 | 5 | 63.9 ± 33.2 | 6 | 16.7 ± 8.2 | 1 | 8.7 | 4 | 6.6 ± 4.3 | 2 | 32.2 | 4 | 5.6 ± 2.9 |
| | 2011 | 8 | 192.2 ± 157.4 | 6 | 22.1 ± 16.3 | 3 | 30.2 ± 16.3 | 5 | 11.1 ± 2.8 | 3 | 27.5 ± 21.4 | 6 | 20.2 ± 9.0 |
| | Total | 13 | 142.9 ± 138.0 | 12 | 19.4 ± 12.6 | 4 | 24.8 ± 17.1 | 9 | 9.1 ± 4.1 | 5 | 29.4 ± 17.8 | 10 | 14.3 ± 10.2 |

Note: % moisture in large and small char ranged from 61.4 to 91.5% (mean 73 ± 15%) across lakes.

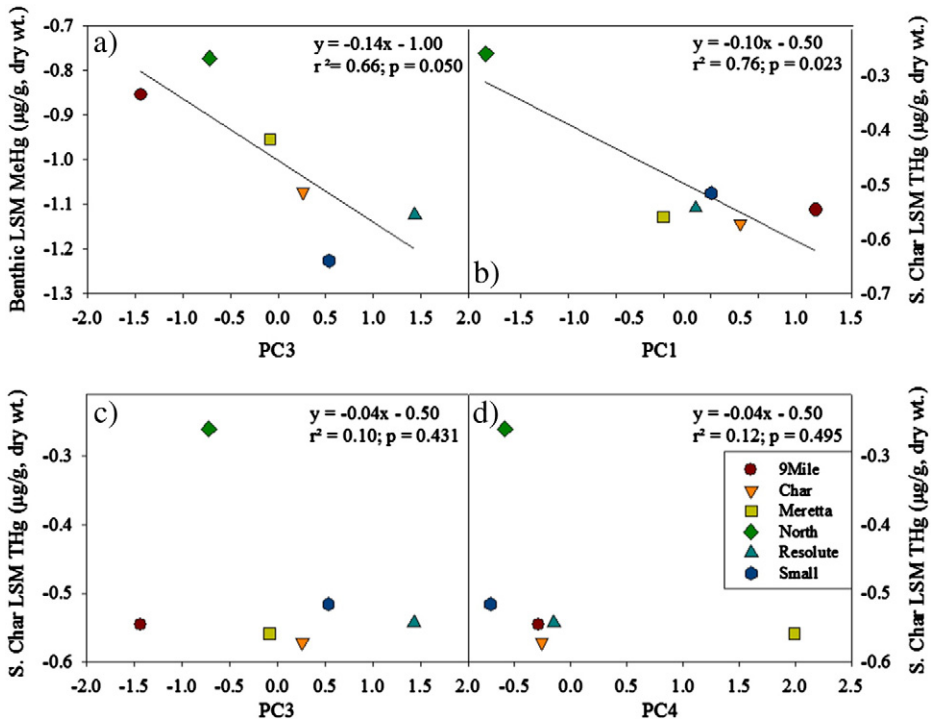


**Fig. 3.** Relationships between biotic $LSM_{Hg}$ and individual PCs representing water chemistry and physical lake variables for the six lakes near Resolute Bay, Cornwallis Island (see Table 2 for PC loadings). a. Benthic LSM MeHg — benthic invertebrates; b, c, d. S. CharLSM THg — small char (muscle estimates). When North Lake was removed, the relationship between small fish LSM and *PC1* was non-significant ($p = 0.980$; $r^2 < 0.01$).

between Hg concentrations in large char and their biological characteristics can be found in the SI. Post-hoc tests for individual lake comparisons were all non-significant for small char $THg_{Length}$ residuals except for North Lake comparisons ($p \leq 0.001$ in all cases; see SI). Similar to large char, small char from North Lake had the highest mean Hg concentration (0.64 ± 0.25 µg/g, dry wt), but 9-Mile had the lowest (0.22 ± 0.08 µg/g, dry wt. in muscle; See Table 3). Relationships between Hg concentrations (whole body) in large and small char and their dietary carbon ($\delta^{13}C$) can also be found in the SI.

Although $THg_{Length}$ residuals for large char varied significantly between some lakes, no PCs were found to significantly predict their $LSM_{THg}$ concentrations. In contrast, $LSM_{THg}$ of small fish had a significant negative relationships with *PC1* and *PC3* ($p = 0.017$; $R^2_{adj} = 0.975$; see SI). When individual variables with high loadings in the *PC1* were examined,

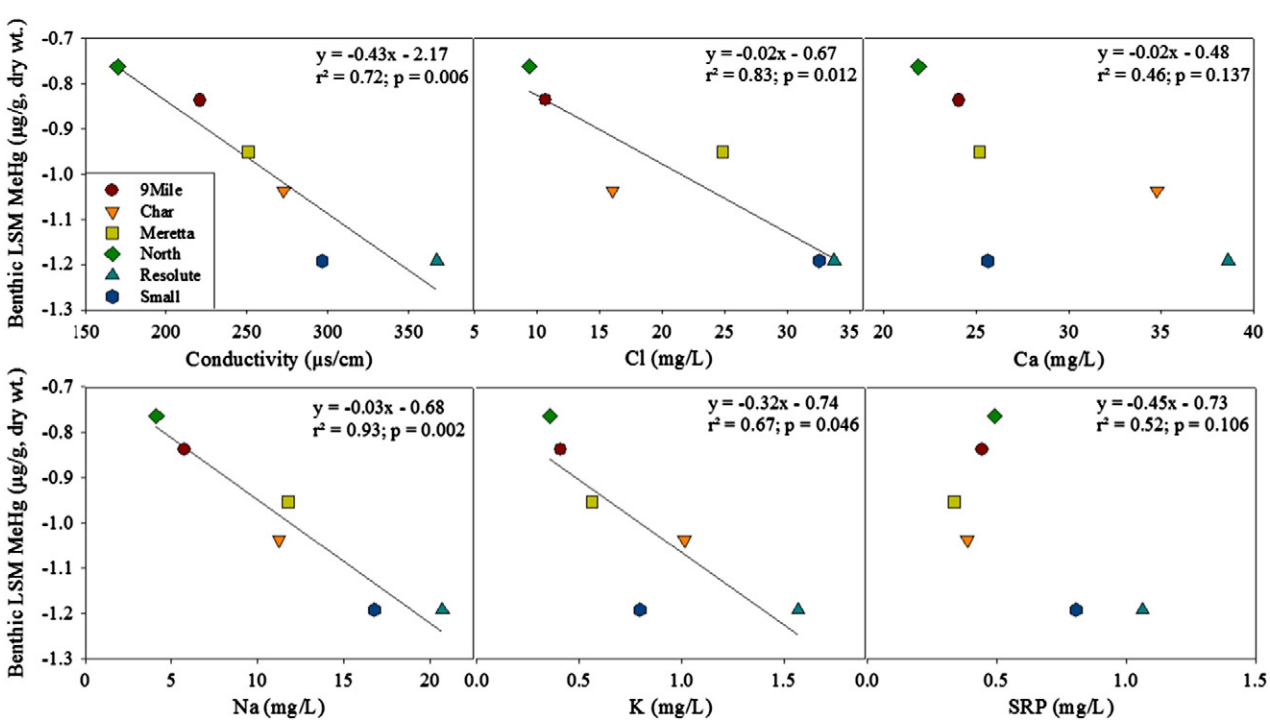


**Fig. 4.** Relationships between benthic $LSM_{MeHg}$ (µg/g, dry wt.) and the individual water chemistry variables dominating *PC1* for six lakes near Resolute Bay, Cornwallis Island.

small char $LSM_{THg}$ was significantly and positively related to $A_C$:$A_L$ ratios ($r^2 = 0.98$; $p < 001$), and $NO_3^-/NO_2^-$ ($r^2 = 0.98$; $p < 0.001$), and chl *a* ($r^2 = 0.92$; $p = 0.002$) concentrations. When North Lake, an influential point, was removed, these relationships became non-significant ($p = 0.528$, $r^2 = 0.12$ for $A_C$:$A_L$, $p = 0.824$, $r^2 = 0.02$ for $NO_3/NO_2$, $p = 0.191$; $r^2 = 0.48$ for chl *a*; see SI for graphs). $LSM_{THg}$ of small char was not significantly related to any individual parameters from PC3 (see SI).

Regressions of log Hg (μg/g, dry wt.) versus $\delta^{15}N$ were significant ($p < 0.001$ for all lakes; $r^2 = 0.58$–$0.87$) and the slopes were positive, confirming biomagnification of Hg through each of these high Arctic food webs (Fig. 5). The slopes, which ranged from 0.09 in Meretta Lake to 0.18 in 9-Mile Lake, were significantly different between lakes (ANCOVA, $F = 497.7$, $p < 0.001$; interaction $F = 5.0$, $p < 0.001$; Log Hg and $\delta^{15}N_{adj}$; see SI for graph). However, despite the moderately strong relationship between slopes and *PC1* ($y = 0.14x + 0.02$; $r^2 = 0.39$; $p = 0.160$; see SI for graphs), no PCs significantly predicted the biomagnification slopes. In contrast, Hg biomagnification intercepts were significantly predicted by the combination of *PC1*, *PC3*, and *PC4* ($R^2_{adj} = 0.951$, $p = 0.029$).

## 4. Discussion

### 4.1. Aqueous concentrations of Hg

MeHg concentrations in these systems were higher in deep water samples compared to surface waters. Previous studies have suggested that Hg(II) methylation in hypolimnetic waters (Eckley and Hintelmann, 2006) and/or lake sediments (Lehnherr et al., 2012a) are important contributors to aqueous MeHg concentrations in these lakes. In addition, %MeHg in water samples, a proxy for methylation rate (Lehnherr et al., 2012b), was 1 to 2% higher in deep water than surface water samples. Overall, %MeHg in water in these six lakes was higher than snow from the surface of remote lakes in the French Alps (mean 2% MeHg; Marusczak et al., 2011) but lower when compared to high Arctic ponds (range 4% to 53%, median 19%; Lehnherr et al., 2012b), likely due to the differences in lake depth and other factors such as nutrient availability.

### 4.2. Concentrations and factors affecting mercury in invertebrates

In the current study, residual MeHg concentrations in benthic and pelagic invertebrates were significantly different between lakes. Benthic invertebrates were typically higher in MeHg when compared to pelagic zooplankton from the same lake; this is likely due to the higher MeHg concentrations in lake sediments, where chironomids live and feed, as compared to the water column (Chételat and Amyot, 2009; Rizzo et al., 2011). This has implications for dietary Hg exposures for char given that they feed mainly on benthic carbon in these systems. The higher MeHg in benthic invertebrates in these Arctic lakes is in contrast to remote Patagonian lakes where THg concentrations were 10-fold higher in pelagic invertebrates when compared to benthic primary consumers (Rizzo et al., 2011). Because the zooplankton communities of Cornwallis Island lakes are dominated by herbivorous copepods (Chételat et al., 2012), their Hg burdens are likely lower than those of zooplankton communities consisting of more predaceous species.

Of the six lakes in this study, MeHg concentrations in zooplankton were the highest in Meretta Lake (142.9 ng/g, dry wt.) and may be due to the atypical presence of *Daphnia* (mainly *Daphnia middendorffiana*), a planktonic crustacean that is larger and up to five times higher in MeHg concentrations than the copepods that dominate most high Arctic lakes (Chételat and Amyot, 2009). This difference in the zooplankton community may be due to the historical eutrophication of Meretta Lake (Chételat and Amyot, 2009; Antoniades et al., 2011). Although sewage inputs ceased in 1998 and its nutrient concentrations were similar to the other lakes in 2010 and 2011, it is possible that historical nutrient inputs had longer-term effects on Hg methylation and bioavailability that may account for the high MeHg concentrations in the zooplankton and water. Furthermore, untreated sewage effluent has been shown to be a source of Hg to various aquatic systems across Canada (Chambers et al., 1997; Bodaly et al., 1998) and historical inputs may have affected baseline Hg concentrations in Meretta Lake. MeHg concentrations in zooplankton from the other 5 lakes ranged from 9.1 to 29.4 ng/g (dry wt.), and are comparable to results from other studies on temperate (0.2 to 42 μg/g dry wt., Rizzo et al., 2011; 9 to 23 ng/g dry wt., Wyn et al., 2009) and Arctic (3 to 15 ng/g dry wt., Chételat and Amyot, 2009) lakes. Due to the relatively high MeHg concentrations and

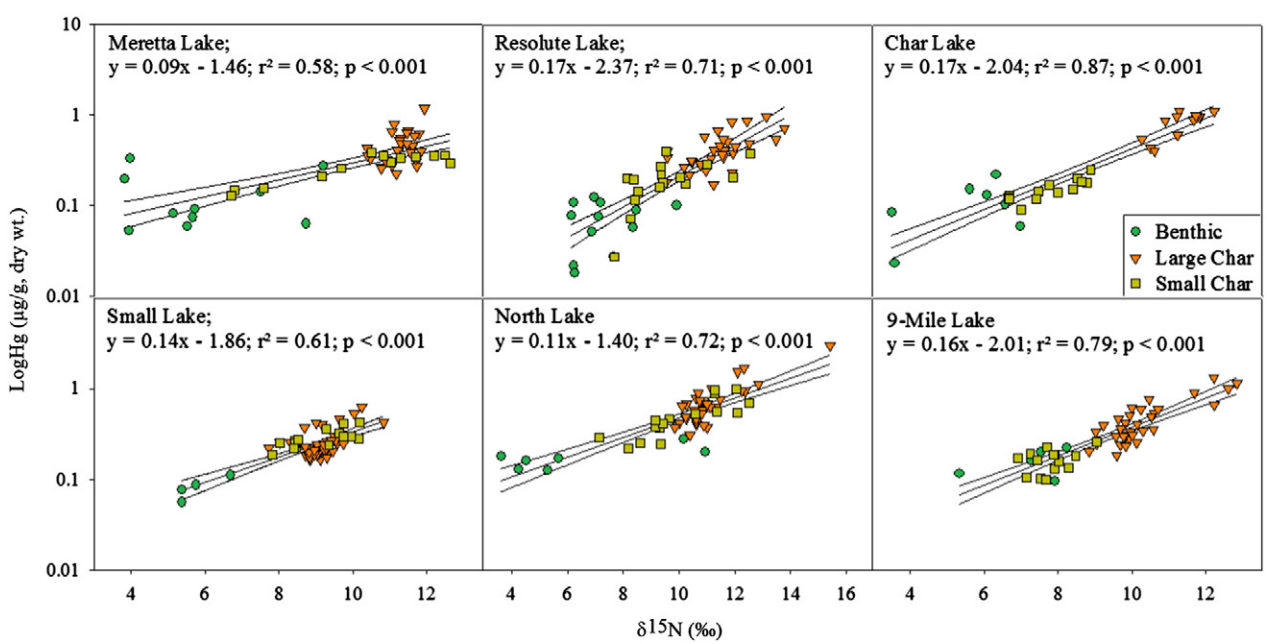


**Fig. 5.** Linear regressions (±95% CI) of unadjusted Log Hg (THg in fish (whole body estimates) and MeHg in benthic invertebrates; μg/g, dry wt.) versus unadjusted $\delta^{15}N$ (‰) values of biota from six lakes near Resolute Bay, Cornwallis Island.

percentage in water, Meretta Lake was an influential point in pelagic $LSM_{MeHg}$ relationships with *PC4*, the only individually significant predictor of zooplankton Hg estimates. Given the low sample size and historical nutrient inputs in Meretta Lake, further study is needed to understand how abiotic factors affect its MeHg concentrations in both water and pelagic invertebrates.

Unlike pelagic invertebrates, benthic $LSM_{MeHg}$ was negatively related to the *PC3* (and the individual ions that dominate the component). Aqueous ions have been shown to affect Hg bioavailability and methylation. Daguené et al. (2012) found reductions in Hg(II) uptake by bacteria in the presence of various cations and at concentrations comparable to those measured in the lakes in the current study. For example, $Ca^{2+}$ (2.2 mg/L) and $Mg^{2+}$ (2.4 mg/L) reduced Hg(II) uptake into bacteria by 65% and 68%, respectively, in laboratory bioassays (Daguené et al., 2012). Further, decreasing ion concentrations result in less competition for binding sites on particulate organic matter (POM) and dissolved organic matter (DOM), known ligands for MeHg in freshwater ecosystems (O'Driscoll and Evans, 2000), which would, in turn, increase MeHg binding to these ligands (Ravichandran, 2004; O'Driscoll et al., 2004; Chase et al., 2012) and, potentially, deposition of MeHg to sediments. In addition, aqueous iron facilitates MeHg photodegradation (Vost et al., 2012) and iron-mediated photo reactions are the principal mechanism for MeHg degradation in Arctic lakes (Hammerschmidt and Fitzgerald, 2006, 2010).

### 4.3. Concentrations and factors affecting mercury in Arctic char

Residual mercury concentrations ($THg_{Length}$) in char were significantly different between the six neighboring lakes, and unadjusted data were comparable to Hg concentrations in char from other Arctic systems (means of 0.111 to 0.227 μg/g, wet wt.; van der Velden et al., 2013a). However, in this study $THg_{Length}$ in char was not predicted by any PC, suggesting that the chemical and physical properties of lakes affect Hg in lower-trophic-level taxa more than in large char. It is possible that a larger range of aqueous nutrient concentrations is needed to rigorously examine whether the chemical characteristics of systems affect the concentrations of Hg in the older and larger individuals of Arctic char. Alternatively, ecological factors such as feeding habits may be the main determinants of Hg in this species.

In contrast to large char, $LSM_{THg}$ of small char was negatively related to *PC1*, *PC3*, and *PC4*; however, only the individual relationship with *PC1* (dominated by $A_C$:$A_L$, $NO_3^-/NO_2^-$, alkalinity, DIC, chl α and $THg_{water}$) was significant. Although North Lake strongly influences these relationships, results suggest that the effects of chemical and physical characteristics of the lakes were influential on Hg concentrations in younger but not older char. While most of the Hg in fish is believed to come from its diet (Hall et al., 1997), a recent study showed that at least 10% of Hg in young-of-the-year (YOY) yellow perch (*Perca flavescens*) comes from water (Hrenchuk et al., 2012). Although small char in the current study are unlikely to be YOY and aqueous contributions to Hg in older fishes are not well known, it is possible that some of the differences in Hg concentrations of the small char may be driven by among-lake differences in the aqueous bioavailability of Hg. For example, lab studies have shown that higher concentrations of DOC (2 to 5 mg/L) reduce the uptake of MeHg across the gills of Sacramento blackfish (*Orthodon microlepidotus*) by up to 85% (Choi et al., 1998). Further, previous studies have shown that nitrate decreases Hg methylation by acting as an electron receptor during microbial respiration of sulfate-reducing bacteria (Todorova et al., 2009; Lehnherr et al., 2012a, 2012b) and increases water column photo-demethylation of MeHg (Kim and Zoh, 2013). In contrast to our study, Mattieu et al. (2013) report positive relationships between Hg concentrations in largemouth (*Micropterus salmoides*) and smallmouth bass (*Micropterus dolomieui*) and lake alkalinity and $A_C$;$A_L$, two parameters that dominated the *PC1*. Given that many of the relationships in the current study are driven by North Lake's high $NO_3^-/NO_2^-$ concentrations and $A_C$:$A_L$, a wider range of lake characteristics is recommended in future studies to better examine the effects that these variables have on Hg concentrations in Arctic char.

### 4.4. Food web structure and Hg biomagnification

The six Arctic lakes we examined had positive and significant Hg biomagnification slopes (from 0.093 to 0.167) that were comparable to but slightly lower than those reported in other freshwater systems (~0.2), including ones from other parts of the Canadian Arctic (Swanson et al., 2010). However, unlike other studies (e.g. Kidd et al., 2012; Clayden et al., 2013; Lavoie et al., 2013), there were no significant relationships between the magnitude of Hg biomagnification and water chemistry or physical variables of these systems. Kidd et al. (2012) reported higher Hg biomagnification slopes in more productive lakes (based on positive relationships with TP), while Clayden et al. (2013) reported lower slopes in more nutrient rich systems. Results from this study found relatively strong (but non-significant) relationships between slopes and *PC1* (dominated by $A_C$:$A_L$; $r^2 = 0.39$) and *PC2* (dominated by nutrients; see SI for graphs); our analyses were likely limited by the smaller number of lakes and range of nutrient concentrations in these ultra-oligotrophic systems. Comparisons across systems that vary in their characteristics are critical for understanding broader drivers of Hg biomagnification (Lavoie et al., 2013).

Although we chose to only include organisms energetically linked (based on $\delta^{13}C$ values) in our calculations of Hg biomagnification slopes, most studies include all food web organisms in these models (e.g. Gantner et al., 2010a; van der Velden et al., 2013b). Despite the high variability in zooplankton MeHg concentrations in these Cornwallis lakes, regressions of Log Hg vs. $\delta^{15}N$ were still significant ($p < 0.001$ in all cases; data not shown) when the zooplankton data were included. The resulting regression slopes increased by 0.01 to 0.08 and intercepts decreased by 0.2 to 0.9 when compared to the models without zooplankton data. It should be noted that the residual errors of these regressions were not normal when zooplankton data were included.

### 4.5. Conclusions

The results of this study generally agree with earlier studies on high Arctic systems, but not with those done at more southerly latitudes. Char in the lakes on Cornwallis Island fed more on benthic than pelagic invertebrates (as per Gantner et al., 2010a), but the former had higher MeHg concentrations when compared to pelagic zooplankton, results that differ from sub-Arctic systems (Power et al., 2002). While Hg concentrations in biota were significantly predicted by their $\delta^{15}N$ in the current study, no predictors of these Hg biomagnification slopes were found, which is in contrast with studies on temperate lake food webs (Kidd et al., 2012; Clayden et al., 2013; Chen et al., 2004).

Although this study examined ultra-oligotrophic lakes with narrow ranges of nutrient and ion concentrations, analyses based on PCA showed that water chemistry and physical variables are important to consider when assessing Hg in organisms from high Arctic lakes. $LSM_{THg}$ in small char, for example, was related to the PC dominated by $A_C$:$A_L$, nitrate, and $THg_{water}$, implying that the bioaccumulation of MeHg (and the bioavailability of Hg) from the water may affect younger char more than large fish. The strong relationships between $LSM_{MeHg}$ concentrations in benthic invertebrates and aqueous ions (or *PC3*) suggest that ions may directly or indirectly control the bioavailability of MeHg to sediment-dwelling invertebrates in high Arctic lakes. While the relationship between MeHg in benthic invertebrates and aqueous ions (and the curious lack of such a relationship for pelagic invertebrates) for the lakes near Resolute Bay warrants further study, it is likely due to a combination of biogeochemical and biological processes influencing uptake into these lower-trophic-level consumers.

## Acknowledgments

Funding and logistical support for this study was provided by the Natural Sciences and Engineering Research Council of Canada through their Canada Research Chair (950-211803) and Discovery (312237) Programs, the Northern Contaminants Program of Aboriginal Affairs and Northern Development Canada (project # M26), Environment Canada (Water Science and Technology Directorate), and the Polar Continental Shelf Program (Natural Resources Canada) in Resolute. The authors sincerely thank Brandy and Debbie Iqaluk (Resolute Bay NU), Guenter Koeck (Austrian Academy of Sciences) and Paul Drevink (Universite de Quebec) for their assistance with field work. We also thank Angela Mercer, Amber Gleason, Greg Lawson and Sam Edmonds for their laboratory assistance.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.04.133.

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